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Title: LABORATORY EVALUATION & APPLICATION OF MICROWAVE ABSORPTION...PLANETARY ATMOS

## PROJECT ADMINISTRATION DATA

OCA contact: Jacquelyn L. Bendall	894-4820
Sponsor technical contact	Sponsor issuing office
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Security class (U,C,S,TS) : U Defense priority rating : N/A Equipment title vests with: Sponsor >\$5,00 REQUIRES PRIOR NASA APPROVAL IF NOT IN APPROVED PROPOSAL BUDGET. Administrative comments -ISSUED TO FXTEND PERIOD OF PERFORMANCE TO 12-31-96. DELIVERABLE SCHEDULE REVISED TO REFLECT NO-COST EXTENSION.

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Effective Completion Date 31-DEC-1996 (Performance) 31-MAR-1997 (Reports)

Closeout Action:	Y/N	Date Submitted
Final Invoice or Copy of Final Invoice	Y	
Final Report of Inventions and/or Subcontracts	Y	
Government Property Inventory and Related Certificate	Y	
Classified Material Certificate	N	
Release and Assignment	N	
Other	N	
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Comments

Distribution Required:

Project Director/Principal Investigator	Y
Research Administrative Network	Y
Accounting	Y
Research Security Department	N
Reports Coordinator	Y
Research Property Team	Y
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NOTE: Final Patent Questionnaire sent to PDPI

W/A E-21-643 NAGW-533

#### I. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoreticallyderived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studies, often leads to significant misinterpretation of available opacity data. For example, laboratory measurements performed by Joiner et al. (1989), under Grant NAGW-533, have shown that the millimeter-wave capacity of ammonia between 7.5 mm and 9.3 mm and also at the 3.2 mm wavelength is best described by a different lineshape than was previously used in theoretical predictions. The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by both radio occultation experiments and radio astronomical observations, and over a range of frequencies which correspond to those used in both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has been the goal of this investigation to conduct such measurements and to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profiles of constituents in those planetary atmospheres.

A key activity over this past grant year has continued to be laboratory

measurements of the microwave and millimeter-wave properties of the simulated atmospheres of the outer planets and their satellites. However, we have also focussed on development of a radiative transfer model of the Jovian atmosphere at wavelengths from 1 mm to 10 cm. This model utilizes our laboratory data and has also been used to evaluate the need for laboratory measurements of other possible absorbers. This modeling effort has led us to conduct a laboratory measurement of the millimeter-wave opacity of hydrogen sulfide ( $H_2S$ ) under simulated Jovian conditions. Similarly, our modeling effort suggests that it may be possible to detect  $H_2S$  in the atmosphere of Jupiter using a medium resolution observation at 1.4 mm. Since no sulfur compounds have yet been detected in the Jovian atmosphere, this would be an important observation. An observation is planned for November from the Caltech Submillimeter Observatory (CSO) in Hawaii. A description of this modeling effort, the laboratory experiment, and the proposed observation is given in Section II.

Recently, we completed measurement, calibration, and interpretive studies of the Venus microwave emission, and a paper entitled, "Observations of the Microwave Emission of Venus from 1.3 to 3.6 cm," by P.G. Steffes, M.J. Klein, and J.M. Jenkins (Steffes <u>et al</u>., 1990) has been published in <u>Icarus</u>. One important issue which was discussed in this paper is the discovery that the microwave absorptivity for gaseous  $H_2SO_4$  which was measured by Steffes (1985 and 1986) appears to differ from a theoretical spectrum newly computed by Janssen (personal communication) by a scale factor. That scale factor suggested that the theoretically-derived "dissociation factor" for gaseous  $H_2SO_4$  (i.e., the percentage of  $H_2SO_4$  which breaks down to form  $SO_3$  and  $H_2O$ ) may have been underestimated. This could result in an underestimation of the absorption from

gaseous  $H_2SO_4$ . Therefore, an experiment has been conducted to correctly evaluate the "dissociation factor" and thus allow unambiguous calibration of laboratory data for  $H_2SO_4$  opacity. A complete description of this experiment was given in <u>Semiannual Status Report #13</u> for Grant NAGW-533 (November 1, 1989 through April 30, 1990), and has been submitted as a paper to <u>Icarus</u> (Fahd and Steffes, 1990).

Yet another important source of information regarding the Venus atmosphere is the increasing number of high-resolution millimeter-wavelength emission measurements which have been recently conducted. (See, for example, de Pater <u>et al</u>., 1990) Correlative studies of these measurements with Pioneer-Venus radio occultation measurements (Jenkins and Steffes, 1990) and out longer wavelength emission measurements (Steffes <u>et al</u>., 1990) should provide the necessary data for characterizing temporal and spatial variations in the abundance of gaseous  $H_2SO_4$  and  $SO_4$ , and for modeling its role in the subcloud atmosphere. In fact, it appears from the results of Steffes <u>et al</u>., (1990) that long term temporal variations in subcloud  $SO_2$  abundance may indeed be occurring. However, unambiguous results require that we have dependable knowledge of the microwave and millimeter-wave opacity of gaseous and liquid  $H_2SO_4$ , and of gaseous  $SO_2$  under Venus conditions.

While some laboratory measurements of the microwave absorption properties of gaseous  $SO_2$  under simulated Venus conditions were made at 13 cm and 3.6 cm wavelengths by Steffes and Eshleman (1981b), no measurements have been made at shorter wavelengths. Since the 1.35 cm wavelength appears to be one of the better wavelengths for measuring the sub-cloud  $SO_2$  abundance (Steffes <u>et al.</u>, 1990), we have conducted laboratory measurements of the 1.35 cm (and 13 cm)

opacity of gaseous  $SO_2$ . The results and application of this work are discussed in Section III of this proposal/report.

In the next grant year we propose to measure the millimeter-wave absorptivity of both gaseous and liquid  $H_2SO_4$  and of gaseous  $SO_2$  under simulated Venus conditions (at the 3 mm wavelength). The system being developed for measurements of the 3 mm properties of liquid  $H_2SO_4$  is briefly described in Section III. As described in Section II, we will also complete laboratory measurements of the 1.4 mm (216 GHz) absorptivity of hydrogen sulfide ( $H_2S$ ) under simulated Jovian conditions and use the results for interpreting our November 1990 observation of Jupiter at that wavelength.

## II. OUTER PLANETS STUDIES

# A. Dual Wavelength Observation of Jupiter at 1.4 mm

Several different calculated emission spectra for Jupiter using various  $H_2S$  distributions for two different ammonia distributions are shown Figure 1. These emission spectra were developed using the Jovian atmospheric model which we described in <u>Semiannual Status Report #13</u> for Grant NAGW-533 (November 1, 1989 - April 30, 1990). An expanded view of the line at 216 GHz is shown in Figure 2. The spectra show that detection of  $H_2S$  is possible using a small bandwidth (on the order of 1 GHz or less) receiver. At least three radio telescopes with such receivers exist:

The Caltech Submillimeter Observatory (CSO) at Mauna Kea, Hawaii



Figure 1 : Calculated emission spectra for Jupiter using two different  $NH_3$  abundance profiles with no  $H_2S$ , solar abundance  $H_2S$ , and ten times solar abundance  $H_2S$ .



Figure 2: Expanded view of calculated emission at the 216 GHz absorption line of H<sub>2</sub>S.

The University of Massachusetts Millimeter Observatory

## The NRAO Observatory at Kitt Peak, Arizona

Of the three observatories capable of making a small bandwidth observation at 216 GHz, the CSO receiver is the most sensitive (by a factor of two or more). The CSO also provides the best location for making such an observation. The high altitude at Mauna Kea minimizes the effects of absorption from the earth's atmosphere. However, since the dip in emission due to  $H_2S$  is small, calculations must be made to see if the antenna and receiver are capable of measuring the dip.

The first step is to calculate the change in antenna temperature which will result from the expected 2K dip in the Jovian emission which is due to  $H_2S$ . It will be useful to first define several quantities used in this type of calculation. The Rayleigh-Jeans approximation, which is valid when (hv << kT) is commonly used at radio frequencies in order to simplify Planck's law. This approximation to Planck's law is given by

$$B = 2v^2 kT/c^2 = 2kT/\lambda^2.$$
(1)

This approximation produces errors on the order of 10% at millimeter wavelengths. Using the Rayleigh-Jeans approximation, the total spectral power density, S, radiated from a spheric blackbody of radius r at a distance d from the source is given as

$$S = \pi k T (r/d)^2 / \lambda$$
<sup>(2)</sup>

(Gulkis, 1987), where S is the flux density. The flux density is in units of power per unit surface area per unit frequency. A commonly used unit for flux density is the Jansky(jy) which is defined as  $10^{25}$  W m<sup>-2</sup> Hz<sup>-1</sup>. The change in antenna temperature,  $\Delta T$ , (in Kelvins) due to a certain flux density, S, may be calculated using

$$\Delta T = \frac{S}{2K} \cdot A_{eff}$$

(3)

where  $A_{\rm eff}$  is the effective area of the antenna. The effective area is defined as

$$A_{eff} = \sigma \cdot \pi r^2 \tag{4}$$

where  $\sigma$  is the efficiency of the dish and r is the radius of the dish. The factor of two in the denominator of Equation 3 is due to the fact that only one polarization is observed.

The  $\Delta T_{H_2S}$  from Jupiter (change in antenna temperature due to the dip in emission from the H<sub>2</sub>S) can be calculated using Equations 2-4. For Jupiter,  $r \approx 71.6 \cdot 10^3$ km and d  $\approx 4.2A$  U (astronomical units) or 6.28  $\cdot 10^8$  km. The diameter of the CSO is 10.4 m and an efficiency of 30% was assumed. The difference in emission assuming a temperature of 180K at 216 GHz and a temperature of 182K at 200 GHz was calculated. After subtracting out the difference in flux due to the difference in wavelength, the  $\Delta T_{H_2S}$  was found to be 0.76 K.

The sensitivity of the receiver,  $\Delta T_{rms}$ , must now be examined in order to determine whether or not it is capable of measuring the  $\Delta T_{H_2s}$ . The sensitivity equation of an ideal receiver is given as

$$\Delta T_{rms} = 2 \cdot \frac{T_s}{\sqrt{t \, dv}}$$

(5)

where  $\Delta T_{rms}$  is called the rms noise power and  $T_s$  (in units of Kelvins) is defined as the system temperature and is a measure of the noise power from the receiver, t is the integration time in seconds, and  $d\nu$  is the bandwidth of the receiver in Hz. The factor of two accounts for the fact that the CSO is a double side band (DSB) receiver. The full receiver bandwidth of the CSO is 500 MHz and operates between 200 and 260 GHz. Although the CSO is equipped to make observations with much greater resolution, the full bandwidth of the receiver will be utilized in order to achieve the necessary sensitivity.

The noise temperature of the CSO (for double side band) is around 100K and the system sensitivity is about 500 mJy for a one second integration time. Evaluating Equation 5 gives a  $\Delta T_{\rm rms}$  of about 0.01 K. Since this is much smaller than the 0.76 change in emission which is due to H<sub>2</sub>S, the CSO should provide the needed sensitivity in order to detect the H<sub>2</sub>S feature.

A proposal was written to the CSO and has been accepted. Three nights of observing time in November 1990 have been granted for use of the CSO.

#### B. Laboratory Measurement of Hydrogen Sulfide Absorption

Our current method for computing of the millimeter-wave absorption from  $H_2S$  uses a Van Vleck-Weisskopf formalism for the pressure-broadened lineshape. However, as in the case of ammonia, the actual absorption at higher pressures may be significantly different from the Van Vleck-Weisskopf theory. In addition, the linewidths for this molecule have never been measured. These parameters must be known accurately if reliable information is to be inferred from observing the effects of  $H_2S$  on the Jovian emission spectrum.

Our analysis shows that by using higher concentrations of  $H_2S$  in a hydrogenhelium atmosphere, absorption should be detectable in the laboratory. However, the absorption lines of  $H_2S$  occur at much higher frequencies (168, 216, and 300 GHz) than our previous laboratory experiments. Thus, the signal source used at these frequencies would have to be harmonically generated. The use of a frequency doubler in order to achieve harmonic generation would decrease the signal strength. The signal-to-noise ratio would therefore be lower than that of previous experiments. Since  $H_2S$  is extremely opaque near the line centers, it may be possible to measure its absorptivity using a long glass cell with a source and precision attenuator at one end and a detector at the other. This configuration is shown in Figure 3. This type of configuration would greatly simplify the laboratory measurement since a resonator would not be necessary.



Figure 3: Block diagram of absorption.

laboratory configuration for measuring H<sub>2</sub>S

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Some of the equipment, such as the signal source, horn antennas, and waveguides will be supplied by the Georgia Tech Research Institute (GTRI). This experiment will be conducted at room temperature since the equipment is too large to fit in our ultra-low temperature freezer. This laboratory data will be very important in analyzing the planned November 1990 observation of Jupiter in which a dip in the emission spectrum due to the 216 GHz line of  $H_2S$  will be searched for.

## **III. VENUS STUDIES**

# A. Laboratory Measurement of the 1.3 cm and 13.3 cm Opacity of SO<sub>2</sub> Under Simulated Venus Conditions

In our paper, "Observations of the Microwave Emission of Venus from 1.3 to 3.6 cm," (Steffes <u>et al</u>., 1990), we discussed the fact that the only gas which substantially affects the microwave emission from Venus at the 1.35 cm wavelength (besides  $CO_2$ ) is  $SO_2$ . This is due to the relatively low opacity of gaseous  $H_2SO_4$  at that wavelength and the relatively low abundance of gaseous  $H_2O$  present in the middle atmosphere. Since the  $CO_2$  abundance and temperature-pressure profile in the lower and middle atmosphere do not vary significantly with time, it is possible to infer variations in the  $SO_2$  abundance from variations in the 1.35 cm flux. However, once an accurate estimate of disk brightness is obtained, accurate estimates of  $SO_2$  abundance can only be made if the microwave absorbing properties of  $SO_2$  at that wavelength are well understood. While some laboratory measurements of the microwave absorption properties of gaseous  $SO_2$  under simulated Venus conditions were made at 13 cm and 3.6 cm (Steffes and Eshleman, 1981b), no measurements have been made at 1.35 cm. Thus, we have conducted such

a laboratory measurement at 1.35 cm.

The experimental approach used to measure the microwave absorptivity of gaseous  $SO_2$  in a  $CO_2$  atmosphere is similar to that used previously by Steffes (1986) for characterizing the absorption of  $H_2SO_4$  in a  $CO_2$  atmosphere. The absorptivity is measured by observing the effects of the test gas mixture on the quality factors (Q's) of microwave resonances between 2.2 and 22 GHz of two cavity resonators (see Figure 4). For relatively low-loss gas mixtures, the relation between the absorptivity of the gas mixture and its effect on the Q and the transmissivity (t) of a particular resonance is given by the equation:

$$\alpha = \left[Q_L^{-1} \left(1 - t^{-1/2}\right) - Q_C^{-1}\right] \frac{\pi}{\lambda}$$

(6)

where  $\alpha$  is absorptivity of the gas mixture in Nepers km<sup>-1</sup>. (Note: an attentuation constant, or absorption coefficient or absorptivity of 1 Neper km<sup>-1</sup> = 2 optical depths per km (or km<sup>-1</sup>) = 8.6866 dB km<sup>-=1</sup>, where the first notation is the natural form used in electrical engineering, the second is the prevalent form used in physics and astronomy, and the third is the common (logarithmic) form. The third form is used often in order to avoid a possible factor-of-two ambiguity in meaning.)  $Q_L$  is the quality factor of the resonance when the test gas mixture is present,  $Q_C$  is the quality factor of the resonance in a vacuum, t is the transmissivity of the resonance with the gas mixture is present. (Note that this new expression takes into account the effects of the coupling of the



Figure 4: Block diagram of Georgia Tech Planetary Atmospheres Simulator, as configured for measurements of microwave refraction and absorption of gases under simulated Venus conditions.

resonator on the resulting absorptivity measurement. We describe this effect, known as "dielectric loading," in Joiner <u>et al</u>., (1989).

This experiment was conducted by introducing a mixture of 8.3%  $SO_2$  (by volume) and 91.7%  $CO_2$  into the pressure vessel at room temperature. The Q and the transmissivity (0 < t < 1) of the two resonances (one at 2.24 GHz or 13.4 cm, and one at 21.7 GHz or 1.38 cm wavelength) are measured with the spectrum analyzer (Q is simply the ratio of resonance center frequency to resonance half-power bandwidth). The measurements were made at 1 Bar pressure increments with the total pressure ranging from 1 to 6 Bars. After the quality factors measured with the gas mixture present were compared with those measured in a vacuum, the absorptivity of the gas mixture was determined as per equation (6).

The results of these measurements are shown in Figure 5 and 6. In Figure 5, we plot the measured absorptivity of  $SO_2$  in a  $CO_2$  atmosphere (normalized by volume mixing ratio) at 2.24 GHz. For comparison, we also plot the absorptivity computed theoretically using the Van Vleck-Weisskopf formalism as per Steffes and Eshleman (1981a). In Figure 6, we plot the measured, normalized absorptivity at 21.7 GHz along with that computed using the Van Vleck-Weisskopf formalism. Our results at 2.24 GHz (13.4 cm) are consistent with results from Steffes and Eshleman (1981b) in that the measured opacity has been shown to be at least 50% larger than computed using the Van Vleck-Weisskopf formalism. Similarly, a pressure dependence of approximately  $p^{1.3}$  was found. However, at 21.7 GHz (1.38 cm) the results are quite consistent with the Van Vleck-Weisskopf formalism, except at the highest pressure (6 Bars). This result is <u>extremely</u> important in that it shows that the f<sup>2</sup> dependence of the SO<sub>2</sub> opacity which was proposed by



Figure 5:

Measured absorption coefficient (normalized by mixing ratio) at 2.24 GHz of SO, in a CO, atmosphere compared with theoretical calculation using the Van Vleck-Weisskopf model (solid line). Error bars shown indicate a  $\pm 1~\sigma$  about the mean value of the measured data. Measurements were made at 296 K with a 8.33% mixing ratio.



## Figure 6:

Measured absorption coefficient (normalized by mixing ratio) at 21.7 GHz of SO, in a CO, atmosphere compared with theoretical calculation using the Van Vleck-Weisskopf model (solid line). Error bars shown indicate a  $\pm 1$   $\sigma$  about the mean value of the measured data. Measurements were made at 296 K with a 8.33% mixing ratio.

Janssen and Poynter (1981) and adopted by Steffes and Eshleman (1981b) for frequencies below 100 GHz and pressures greater than 1 Bar is <u>not</u> valid. Since the Van Vleck-Weisskopf formulation <u>is</u> valid at 1.3 cm for pressures less than 6 Bars, it is possible to use it in determining estimated  $SO_2$  abundances from 1.3 cm emission measurements. However, since the 1.3 cm opacity appears to be lower than indicated by the expression presented in Steffes and Eshleman (1981b) it is more difficult to accurately determine limits to  $SO_2$  abundance. For example the  $SO_2$  abundance inferred by Steffes <u>et al</u>., (1990) from 1.35 cm emission was 45 (+90 or -45) ppm using the expression from Steffes and Eshleman (1981b). Using these laboratory results, the inferred abundance is about 90 (+180 or =90) ppm.

# B. Laboratory Measurement of the Millimeter-Wave Properties of Liquid Sulfuric Acid

Observations of the millimeter-wave emission of Venus at 115 GHz (2.6 mm) suggests significant spatial and possible diurnal variations in the continuum flux emission (de Pater et al., 1990). Such variations are undoubtedly due to variabilities in abundances of absorbing constituents, since substantive variations in temperature-pressure profiles in the middle and lower atmosphere have not been indicated by either <u>in-situ</u> or radio occultation studies. Potential constituents whose abundance variability might account for the flux variations include gaseous  $H_2SO_4$  and  $SO_2$ , and liquid  $H_2SO_4$  (the cloud condensate). Estimating the absorbing properties of any of these constituents at frequencies near 100 GHz is difficult since no laboratory measurements have been reported for Venus-like conditions. de Pater <u>et al</u>. (1990) predicted that the cloud condensate was most likely the major source of non-CO<sub>2</sub> opacity, basing their estimate on the

millimeter-wave opacity of water. However, since the microwave properties of water are substantially different from sulfuric acid (ref. Ho and Hall, in Cimino, 1982), a laboratory measurement is definitely needed.

In the next grant year, we will measure the absorption and refraction of liquid  $H_2SO_4$  (85%, solution by weight) at 100 GHz, using the free space measurement system shown in Figure 7. Note that the complex permittivity of the sample will be measured using a millimeter-wave network analyzer. In the next grant year we will also complete construction of our laboratory system for measurement of the 3 mm opacity of gaseous  $H_2SO_4$  and  $SO_4$  under simulated Venus conditions.

## IV. PUBLICATIONS AND\_INTERACTION WITH OTHER INVESTIGATORS

In this grant year, a paper regarding the observation, calibration, and interpretation of the Venus microwave emission spectrum was published in <u>Icarus</u> (Steffes <u>et al</u>., 1990). Similarly, we have submitted a paper to <u>Icarus</u> on the topic of the vapor pressure and equilibrium between gaseous and liquid  $H_2SO_4$  and its effect on models for the Venus atmosphere (Fahd and Steffes, 1990). We are also preparing papers on the topic of interpretation of the Jovian microwave and millimeter-wave emission spectrum and on our laboratory measurement of the 1.35 cm opacity of  $SO_2$ .

In early November 1989, we attended the 21st Annual Meeting of the Division for Planetary Sciences of the American Astronomical Society and presented papers on interpretation of the Jovian microwave and millimeter-wave emission spectrum



Figure 7: Sketch of free-space measurement system.

(Joiner and Steffes, 1989), on the vapor pressure and equilibrium between gaseous and liquid  $H_2SO_4$  (Fahd and Steffes, 1989), and on the temporal variation of the abundance of  $SO_2$  below the clouds of Venus (Steffes, 1989). Likewise, we are currently preparing papers for presentation at the October 1990 AAS/DPS meeting and accompanying Conference on Laboratory Research for Planetary Atmospheres (Charlottesville, VA).

Our work has been complemented by our involvement in the Pioneer-Venus Guest Investigator Program in which we have been involved in processing radio occultation data in order to obtain 13 cm absorptivity profiles for the Venus atmosphere (Jenkins and Steffes, 1990). This has kept us in close contact with a large number of Venus investigators. More informal contacts have been maintained with groups at the California Institute of Technology, with the Stanford Center for Radar Astronomy (Drs. V.R. Eshleman, G.L. Tyler, and T. Spilker, regarding Voyager results for the outer planets, and laboratory measurements), and with JPL (Drs. Robert Poynter, Samuel Gulkis, and Michael Klein, regarding radio astronomical observations of the outer planets and Venus). We have also worked with Dr. Imke de Pater (University of California-Berkeley) by using our laboratory measurements of atmospheric gases in the interpretation of radio astronomical observation of Venus and the outer planets. We have also studied possible effects of the microwave opacity of cloud layers in the outer planets' atmospheres. In this area, we have worked both with Dr. de Pater and with Dr. Paul Romani (Goddard SFC).

Dr. Steffes has also been active in the review of proposals submitted to the Planetary Atmospheres Program at NASA. We have also continued to serve the

planetary community through the distribution of reprints of our articles describing our laboratory measurements and their application to microwave and millimeter-wave data from planetary atmospheres. The results of these measurements have been used in the mission planning for radio and radar systems about the Galileo and Magellan missions, and more recently, for proposed experiments for the Cassini mission. Another source of close interaction with other planetary atmospheres principal investigators has been Dr. Steffes' membership in the Planetary Atmospheres Management and Operations Working Group (PAMOWG). Travel support for Dr. Steffes' attendance at PAMOWG meetings, as well as the AAS/DPS meetings has been provided by Georgia Tech in support of Planetary Atmospheres Research.

#### V. PROPOSED PROCEDURE AND LEVEL OF EFFORT

The proposed program will continue an ambitious effort to resolve many of the uncertainties involved in the interpretation of microwave and millimeter-wave absorptivity data from Venus, Jupiter, Saturn, Uranus, and Neptune. The next grant year (November 1, 1990 through October 31, 1991) will begin with our completion of laboratory measurements of the millimeter-wave opacity of gaseous  $H_2S$  (under Jovian conditions) and liquid  $H_2SO_4$  (Venus cloud constituent). We will use these results in completing development of microwave and millimeter-wave radiative transfer models for Venus and the outer planets. We will also observe the emission from Jupiter at and around the 1.4 mm wavelength from the Caltech Submillimeter Observatory (CSO-Mauna Kea, Hawaii) in order to set limits on the abundance of  $H_2S$  in the Jovian atmosphere. Finally, we will complete design and

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## REPORT

## TO THE

# NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

# SEMI-ANNUAL STATUS REPORT #1

for

## GRANT NAGW-533

## LABORATORY EVALUATION OF MICROWAVE ABSORPTION PROPERTIES UNDER SIMULATED CONDITIONS FOR PLANETARY ATMOSPHERES

# Paul G. Steffes, Principal Investigator

February 1, 1984 through July 31, 1984

Submitted by:

Professor Paul G. Steffes School of Electrical Engineering Georgia Institute of Technology Atlanta, Georgia 30332-0250

## I. INTRODUCTION

The need for laboratory measurement of the microwave absorption properties of atmospheric constituents, under simulated conditions for the planetary atmosphere being considered, was demonstrated by Steffes and Eshleman (1981 and 1982). The recognition of the need to make such measurements over a range of temperatures and pressures which correspond to the periapsis altitudes of radio occultation experiments, and over a range of frequencies which correspond to both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements.

## II. THE MEASUREMENT FACILITY

The overall configuration for such a facility is similar to those used previously by Steffes and Eshleman (1981 and 1982). As shown in figure 1, a resonator is used to detect losses at microwave frequencies from an introduced gas mixture. The major differences between the Georgia Tech atmospheric simulator and those used previously are as follows:

- The pressure containment vessel is large enough

   (0.33m diameter, 0.38m height) so that it will allow
   the use of resonators which operate at frequencies as
   low as 1.5 GHz. (20 cm wavelength) and yet still be
   heated to over 500K while maintaining pressures up to
   10 atmospheres.
- 2) The resonator used to measure absorption in the 1.5 to 8 GHz range (20 to 3.7 cm wavelengths) exhibits a higher "quality factor," or Q, due to special construction techniques. As a result, the system sensitivity is greater, resulting in lower measurement uncertainties.

-1-

- 3) The use of a digitally-refreshed spectrum analyzer further serves to increase the sensitivity of the system. As a consequence, when measurements are to be made of the microwave absorption from gases which are obtained from liquidderived vapors, a smaller amount of source liquid can be used, assuring that all will reach vapor phase, and yet the absorptivity is still measurable. Thus, uncertainties regarding vapor pressure behavior of the source liquids (which in the case of H<sub>2</sub>SO<sub>4</sub> are on a factor-of-ten scale) no longer affect the accuracy of the absorptivity measurement. In fact, accurate measurements of the vapor pressures accompanying liquid H<sub>2</sub>SO<sub>4</sub> under Venus atmospheric conditions (based on the absorptivity from vapors) can now be made.
- Signal source and spectrum analyzer frequency ranges have been extended to allow measurement up to 40 GHz.
- 5) A refridgerant capability is currently being developed which will allow operation as low as -140C (133K), so as to allow measurements under simulated conditions for the outer planets.

The development of the microwave/millimeter-wave atmospheric absorption simulator is now reaching completion. Much of the capital support required to construct this facility has been provided by the Georgia Tech Research Institute (GTRI), with additional equipment available from existing laboratories within the Georgia Tech School of Electrical Engineering. This facility will accomodate absorption measurements which will provide accurate calibration of radioscientific experiments aboard Mariner, PioneerVenus, Voyager, Galileo, and Venus Radar Mapper spacecraft. In addition, the capability to make absorption measurements in the 1 to 10cm wavelength range

-2-

will provide a basis for interpretation of a wide range of planetary radio astronomy data.

## III. INITIAL LABORATORY MEASUREMENTS

At the writing of this first semi-annual status report, we have begun the first of the laboratory measurements to investigate the absorptive properties of gaseous  $H_2SO_4$  under conditions simulating the Venus atmosphere in the 30 to 50 km altitude range (pressures from 1 to 8 Bar in a  $CO_2$  atmosphere, and temperatures reaching to 500 K). The initial measurements will be conducted at 13.3 cm and 3.6 cm wavelengths in order to allow direct application to radio occultation absorptivity data. Due to instrumental limitations on Venus atmospheric probe experiments, this will provide the only method for measurement of  $H_2SO_4$  vapor abundance in the Venus atmosphere.

In the past, introduction of gaseous H<sub>2</sub>SO<sub>4</sub> into a microwave measurement system was accomplished by allowing liquid sulfuric acid to reach vapor pressure equilibrium within the test chamber. (See, for example, Steffes and Eshleman, 1982.) However, such an approach requires good knowledge of H<sub>2</sub>SO<sub>4</sub> vapor pressure accompanying liquid sulfuric acid. Unfortunately, large uncertainties in the vapor pressure behavior of sulfuric acid have resulted in correspondingly large uncertainties in the absorptivity data gathered in this way. The higher sensitivities available with the Georgia Tech simulator allow the use of an alternate approach in order to avoid this problem. At the beginning of the experiment, a precisely measured volume of sulfuric acid liquid is introduced into the chamber. The amount of liquid is small enough so that the pressure created by its <u>complete</u> vaporization is well below even the most conservative estimates for the saturation vapor pressure of sulfuric acid. Thus, the mixing ratio of gaseous H<sub>2</sub>SO<sub>4</sub> in the test atmosphere will be known to a reasonably good accuracy, since

-3-

all of the introduced liquid will be vaporized. (It should be noted, however, that theoretical calculations for the percentage of  $H_2SO_4$  molecules which dissociate to form relatively transparent  $SO_3$  and  $H_2O$  are still used.)

After the initial measurements which will determine the pressure and temperature dependences (and the magnitude) of the absorption from gaseous H<sub>2</sub>SO<sub>4</sub> in a CO<sub>2</sub> atmosphere, we will then conduct an experiment using a large reservoir of liquid H<sub>2</sub>SO<sub>4</sub> as the source of H<sub>2</sub>SO<sub>4</sub> vapor. Since the microwave absorption is proportional to the H<sub>2</sub>SO<sub>4</sub> mixing ratio, we will be able to infer (from the absorption we observe) the equilibrium mixing ratio (and therefore, partial pressure) of gaseous H<sub>2</sub>SO<sub>4</sub> above liquid sulfuric acid. This will be of great help to investigators trying to model convection and cloud condensation in the Venus atmosphere, especially given the current uncertainties in the vapor pressure behavior of H<sub>2</sub>SO<sub>4</sub>. (For further discussion, see Esposito <u>et al.</u>, 1983.)

## IV. FURTHER MEASUREMENTS AND INTERACTION WITH OTHER INVESTIGATORS

The second half of the current contract year will see further measurements of the microwave absorption properties of gaseous sulfuric acid. Since the instrumentation for our facility will operate to 40 GHz, measurements at higher frequencies will be especially attractive. Such measurements require high-Q resonators which operate at the frequencies of interest (15 GHz and 30 GHz are of special interest due to VLA operations at those frequencies). A resonator operating in the 8 to 24 GHz range could be easily constructed, but such a resonator has already been built at the Stanford Center for Radar Astronomy (V. R. Eshleman, Director). Since the group at Stanford is currently without the required instrumentation for operation above 12 GHz, they have been invited to participate in measurements at the Georgia Tech facility. In the 24 to 40 GHz frequency range the cylindrically

-4-

structured resonators used at lower frequencies become operationally inefficient. Instead, Fabry-Perot type resonators are generally used. The components for such a resonator currently exist at Georgia Tech, and some attempt will be made to integrate this resonator with our atmospheric simulator.

In the general area of planetary atmospheres, we have had interaction with a number of workers both within the Georgia Tech Schools of Physics, Geophysical Sciences, and Chemistry, and with workers at other institutions. Topics of these discussions have ranged from simulator hardware construction to the abundance and structure of sulfuric acid dimers and hydrates. We hope to be able to present initial results from this work to our colleagures at the Annual Meeting of the Division for Planetary Sciences of the American Astronomical Society this October.

## V. SUMMARY

In the first six months of this program we have successfully assembled a highly-flexible microwave/millimeter-wave absorption simulator for planetary atmospheres, and begun its use to measure the absorption from gaseous  $H_2SO_4$ under Venus atmospheric conditions. We will continue these measurements over the next six months with the goal of using the results for interpretation of radio occultation data and radio astronomical data, and for solving basic questions as to the distribution and function of sulfur-bearing compounds in the Venus atmosphere.

-5-

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Figure 1: Georgia Tech Planetary Atmospheres Simulator, as Configured for Measurements of Microwave Absorption of gaseous  ${}^{\rm H}2{}^{\rm SO}4$  under Venus Atmospheric Conditions

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#### REPORT

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## TO THE

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

## ANNUAL STATUS REPORT

for

GRANT NAGW-533

## LABORATORY EVALUATION OF MICROWAVE

ABSORPTION PROPERTIES UNDER SIMULATED

#### CONDITIONS FOR PLANETARY ATMOSPHERES

Paul G. Steffes, Principal Investigator

February 1, 1984 through January 31, 1985

## Submitted by

Professor Paul G. Steffes School of Electrical Engineering Georgia Institute of Technology Atlanta, Georgia 30332-0250

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#### I. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often lead to significant misinterpretation of available opacity data. Steffes and Eshleman (1981) showed that under environmental conditions corresponding to the middle atmosphere of Venus, the microwave absorption due to atmospheric  $SO_2$  was 50 percent greater than that calculated from Van Vleck-Weiskopff theory. Similarly, the opacity from gaseous  $H_2SO_4$  was found to be a factor of 7 greater than theoretically predicted for conditions of the Venus middle atmosphere (Steffes and Eshleman, The recognition of the need to make such measurements over a range 1982). of temperatures and pressures which correspond to the periapsis altitudes of radio occultation experiments, and over a range of frequencies which correspond to both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements.

In the initial year of Grant NAGW-533, this facility has been developed, and then operated, in order to evaluate the microwave absorbing properties of gaseous sulfuric acid  $(H_2SO_4)$  under Venus atmospheric conditions. The results have then been applied to measurements from Mariner 5, Mariner 10, and Pioneer-Venus Radio Occultation experiments, to determine abundancies of

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gaseous sulfuric acid in the Venus atmosphere, with accuracies exceeding those achieved with <u>in-situ</u> instruments. (Steffes <u>et al</u>, 1984). Measurements of the microwave properties of the vapors accompanying liquid  $H_2SO_4$  have also resulted in more accurate estimates of the vapor pressure behavior of sulfuric acid, which are critical for modeling the behavior and structure of the Venus atmosphere (Ibid).

The initial results of this work were presented at the 16th Annual Meeting of the Division for Planetary Sciences of the American Astronomical Society (Steffes <u>et al</u>, 1984). Response from colleagues has been extremely positive, with suggestions for joint research and/or publications coming from Jet Propulsion Laboratory, Stanford University, and the University of Massachusetts. Currently, four journal publications are being prepared to document this work.

Plans for future work include, first, completing the shorter wavelength (approximately 1 cm) absorptivity measurements for  $H_2SO_4$  and application of these measurements to radio astronomical measurements of Venus, including measurements made with the NRAO Very Large Array (VLA). The measurement facility would then be reconfigured, so as to allow measurement of the micro-wave absorption of ammonia ( $NH_3$ ), methane ( $CH_4$ ), and other potential microwave absorbers under simulated conditions for the outer plantes (Jupiter, Saturn, Uranus, and Neptune). These results would then be applied to Voyager radio occultation data, as well as a wide range of radio astronomical data for these planets, in order to establish accurate atmospheric abundance profiles for these gases on the outer planets.

# II. THE GEORGIA TECH RADIO ASTRONOMY AND PROPAGATION (R.A.P.) FACILITY

As part of the Electromagnetics Group of the School of Electrical Engineering, the Radio Astronomy and Propagation (R.A.P.) Laboratory has been developed to facilitate research related to radio propagation in telecommunications and remote sensing of planetary atmospheres. The overall configuration for our planetary atmosphere simulator is similar to those used previously by Steffes and Eshleman (1981 and 1982). As shown in Figure 1, a resonator is used to detect losses at microwave frequencies from an introduced gas mixture. The major differences between the Georgia Tech atmospheric simulator and those used previously are as follows:

- (1) The pressure containment vessel is large enough (0.33 m diameter, 0.38 m height) so that it will allow the use of resonators which operate at frequencies as low as 1.5 GHz (20 cm wavelength), and yet still be heated to temperatures over 500 K while maintaining pressures up to 10 atmospheres.
- (2) The resonator used to measure absorption in the 1.5 to 8 GHz range (20 to 3.7 cm wavelengths) exhibits a higher "quality factor," or Q, due to special construction techniques. These techniques include the use of stainless steel to construct the resonator, and then plating with nickel, copper, and silver. This not only maximizes "Q," but insures that the Q will remain high in spite of drastic thermal shifts. As a result, the system sensitivity is greater, resulting in lower measurement uncertainties.
- (3) The use of a digitally-refreshed spectrum analyzer further serves to increase the sensitivity of the system. As a consequence, when measurements are to be made of the microwave absorption from gases

which are obtained from liquid-derived vapors, a smaller amount of source liquid can be used, allowing operation at lower temperatures without condensation.

- (4) Uncertainties of mixing ratios of gases obtained from liquid-derived vapors have been greatly reduced for two reasons. First, the amounts of liquid used to generate these gases can be determined with a volume accuracy of  $\pm$  0.005 ml. Thus when an amount of liquid becomes vapor, it becomes possible to compute the partial pressure due to that vapor from the ideal gas equation and the measured change in liquid-derived vapors is by measuring the refractivity of those vapors. Since the index of refraction is proportional to the vapor abundance, our ability to accurately measure the refractivity of such vapors can also be used to determine the resulting vapor abundance/pressure. This has been especially useful for gases such as  $H_2SO_4$ , where little accurate vapor pressure data has been available.
- (5) Signal source and spectrum analyzer frequency ranges have been extended to allow measurement up to 40 GHz. A resonator operating in the 8 to 26 GHz range is also being constructed.
- (6) A refridgerant capability can be added which will allow operation as low as -140 C (133 K), so as to allow measurements under simulated conditions for the outer planets.

The initial development of the microwave/millimeter-wave atmospheric absorption simulator has now reached completion. Much of the capital support required to construct this facility has been provided by the Georgia Tech Research Corporation (GTRC), with additional equipment available from existing

laboratories within the Georgia Tech School of Electrical Engineering. This facility has now provided absorption measurements which are being used to accurately calibrate the radioscientific data from Mariner 5, Mariner 10, Pioneer-Venus, and, in the future, the Venus Radar Mapper spacecraft. In addition, absorption measurements in the 1.1 to 10 cm wavelength range are providing a basis for interpretation of a wide range of Venus radio astronomical observations.

In the second year of this program, we hope to be able to complete measurements of the vapor pressure behavior of gaseous  $H_2SO_4$ , as well as its microwave absorption behavior in the 26 to 40 GHz (7.5 mm to 1.1 cm) frequency range, under simulated Venus conditions. This will require the integration of a 26 to 40 GHz Fabry-Perot type resonator into our system. Subsequently we will begin system reconfiguration for simulation of the atmospheres of the outer planets. This will require moving the simulator from a heated environment (simulating Venus) to a refrigerated environment (simulating outer planets). It will also require changes in the simulator components so as to be compatible to the extremely flammable constituents (predominantly hydrogen) of the outer planets. Measurements in the 1.5 to 40 GHz range will then be made.

#### **III. LABORATORY MEASUREMENTS**

A wide range of laboratory measurements of the microwave absorbing properties of gaseous  $H_2SO_4$  in a predominantly  $CO_2$  atmosphere have been made under temperature and pressure conditions (temperatures to 575 K and pressures to 6 atm.) simulating the middle atmosphere (30-50 km altitude) of Venus. The initial measurements were made at frequencies of 2.24 and 8.44 GHz (13.4 cm and 3.6 cm wavelengths) in order to allow direct application to absorptivity

data from Pioneer-Venus, Mariner 5, and Mariner 10 radio occulatation experiments.

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Since absorptivity at radio frequencies is generally directly proportional to the abundance (either by number or by volume) of the absorbing constituent, the results of our measurements, shown in Figures 2 and 3, are plotted as absorptivities normalized by  $H_2SO_4$  mixing ratio. Figure 2 shows the normalized absorptivity of gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere (taken around 570 K temperature and at 2.24 GHz frequency) as a function of pressure. Similarly, Figure 3 shows similar measurements taken around 525 K and at a frequency of 8.42 GHz. The  $H_2SO_4$  mixing ratios used to normalize the plots were obtained using vapor pressure data from Figure 4, which were obtained by the measurement techniques described in Section II. Several significant discoveries have been made as a result of these measurements:

- (1) While generally confirming the initial laboratory measurements made by Steffes and Eshleman (1982) of  $H_2SO_4$  vapor absorption at a single temperature and pressure, it has been found that the microwave absorption of  $H_2SO_4$  in a  $CO_2$  atmosphere has a <u>significant</u> pressure dependence at both the 13 cm and 3.6 cm wavelengths. This is in sharp contrast to the theoretical calculation presented in Cimino (1982) (actually performed by Janssen and Poynter).
- (2) A new relation for  $H_2SO_4$  vapor pressure has been developed which generally confirms the work of Roedel (1979) and Ayers <u>et al</u> (1980). (ln p(atm) = 9.42 - 7330/T.) However, our findings are different enough that they significantly affect the condensation temperatures and altitudes predicted for Venus. This will be of great help to investigators trying to model convection and cloud condensation in the Venus atmosphere, especially given the current uncertainties in

the vapor pressure behavior of  $H_2SO_4$ . (For further discussion, see Esposito et al, 1983.)

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- (3) Frequency dependences which vary significantly with pressure have been found for the absorption from gaseous  $H_2SO_4$ , ranging from f<sup>1.23</sup> at 1 atm pressure to f<sup>1.7</sup> at 6 atm pressure. This will significantly effect comparisons of radio occultation data with radio astronomical data from Venus.
- (4) Simple multiplicative expressions for the absorption from gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere at 13 cm and 3.6 cm wavelengths have been developed.  $(\alpha_{13} \approx 6.1 \times 10^9 (P)^{1/2} T^{-3} \text{ and } \alpha_{3.6} \approx 3.13 \times 10^{10} (P)^{0.85} T^{-3}$  where  $\alpha$  is in dB km<sup>-1</sup>, P is atmospheres and T is in Kelvins.) These will be highly useful in converting microwave absorptivity profiles to  $H_2SO_4$  abundance profiles.

In addition to the measurements at 13 cm and 3.6 cm wavelengths, measurements in the 1.1 to 3.6 cm wavelength range are currently being conducted and should yield similar valuable data for interpretation of a wide range of radio astronomical data.

# IV. APPLICATION OF LABORATORY RESULTS

We are now beginning a wide range of analytical studies based on the laboratory data. These studies cover an area which is so wide that complete documentation would be difficult at this point in time. Examples of application of the data can be seen in Figures 5 and 6. Figure 5 shows the 13 cm wavelength microwave absorption which would arise from a 20 ppm abundance of gaseous  $H_2SO_4$  in the Venus atmosphere which condenses to form clouds at 48 km altitude. It can be seen from this figure that gaseous  $H_2SO_4$  is the predominant microwave absorber in the 35 to 48 km altitude range as compared to other absorbing constituents. It can also be seen that abundances on the order of 20 to 30 ppm could account for most of the absorption measured in a number of radio occultation measurements.

Figure 6 shows abundances of gaseous sulfuric acid, at several altitudes, obtained by attributing all of the 13 cm opacity measured in the 3 radio occultation experiments shown, to gaseous sulfuric acid. Also shown are curves showing saturation vapor abundance as a function of altitude. Note that the vapor will not condense at a given altitude unless its abundance equals or exceeds the saturation abundance. The saturation vapor abundance from Gmitro and Vermeulen (1964) would imply that no significant cloud formation could occur except at altitudes well above 50 km. This, of course, conflicts with in-situ atmospheric probe findings. (See, for example, Knollenberg and Hunten, 1980.) The saturation vapor abundance from Ayers et al. (1980) would require cloud formation below the 45 km altitude, which likewise conflicts with in-situ findings. The saturation abundances inferred from our measurements are consistent with cloud formation in the 46 to 48 km range, for the given abundances of gaseous H<sub>2</sub>SO<sub>4</sub>, except for a single peak measurement from the Mariner 10 experiment. However, it should be noted that the average Mariner 10 absorptivity measurements were well below that figure (Lipa and Tyler, 1979).

Thus, abundances of gaseous sulfuric acid below the main Venus cloud layer on the order of 20-35 ppm are indicated from radio occultation 13 cm absorptivity measurements. However, positional and temporal variations in the measured opacity at 13 cm (from radio occultation experiments) and at 3.6 cm (from radio astronomical and radio occultation experiments) indicate as much as factor of 2 variations in the gaseous  $H_2SO_4$  abundance in the 30-50 km altitude range. The variations are suggestive of significant atmospheric changes,

such as would be induced by volcanic activity. Further study into this issue is being conducted.

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#### V. PUBLICATIONS AND INTERACTION WITH OTHER INVESTIGATORS

Three presentations of the initial results of this work have been made. The major presentation was given at the 16th Annual Meeting of the Division for Planetary Sciences of the American Astronomical Society (Steffes <u>et al.</u>, 1984--Abstract attached, see Appendix 1). Presentations have also been made to the Georgia Tech Community and to the Atlanta IEEE. Response from colleagues has been extremely positive, with suggestions for joint research and/or publications coming from the Jet Propulsion Laboratory, Stanford University, and the University of Massachusetts.

Four publications are currently being prepared:

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- (1) A paper presenting our laboratory results for the microwave absorption and vapor pressure behavior of gaseous sulfuric acid under simulated conditions for the Venus atmosphere.
- (2) An accompanying paper which applies our laboratory results to a wide range of microwave absorption data and to atmospheric structure data from Venus, and results in abundance profiles for  $H_2SO_4$  in the Venus atmosphere.
- (3) A paper written jointly with Drs. Michael J. Klein and Michael A. Janssen of JPL, discussing variations of the microwave emission from Venus at 3.6 cm over the past 20 years, and the implications, in light of our laboratory results, for volcanism related atmospheric changes.
- (4) A paper which will discuss the vapor pressure behavior of  $H_2SO_4$ . In addition to the analysis/research being conducted jointly with

Drs. Klein and Janssen, joint measurements with the Stanford University Center for Radar Astronomy (V. R. Eshleman, Director) of the microwave properties of gaseous sulfuric acid have been made, using the Georgia Tech facility. Also, in the general are of planetary atmospheres, we have had interaction with a number of workers both within the Georgia Tech Schools of Physics, Geophysical Sciences, and Chemistry, and with workers at other institutions. Topics of these discussions have ranged from simulator hardware construction to the abundance and structure of sulfuric acid dimers and hydrates.

#### VI. CONCLUSION

The work conducted during the first year of NASA Grant NAGW-533 has resulted in laboratory measurements, which when applied to data from the Venus atmosphere, give new insight into the abundance and structure of sulfuric acid vapor in the Venus atmosphere. Because of instrumental limitations of <u>in-situ</u> probes, this has become the major technique for determining the  $H_2SO_4$  abundance profiles. Application of the laboratory results to a wider range of radio astronomical data may even provide evidence for active volcanism on Venus.

Future work under this grant will include absorptivity measurements under simulated Venus conditions for a wider range of wavelengths. This will allow for analysis and interpretation of an even wider range of radio astronomical data. Upon completion of these measurements, the system will be reconfigured so as to allow for the beginning of microwave absorptivity measurements of several gaseous constituents under simulated conditions for the outer planets. Such measurements will be used to interpret data from Voyager radio occultation experiments, radio astronomical observations of the outer planets, and, in the future, radio absorption measurements from the Galileo spacecraft and probe.

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VIII. KEY FIGURES

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Figure 1: Gerogia Tech Planetary Atmospheres Simulator, as configured for measurements of microwave absorption of gaseous H<sub>2</sub>SO<sub>4</sub> under Venus atmospheric conditions.





PRESSURE (atm)

Figure 2: Measured microwave absorption (normalized by number mixing ratio) for gaseous  $H_2SO_4$  in a CO<sub>2</sub> atmosphere at 2.24 GHz. Mixing ratio was obtained from data in figure 4. Dashed line represents a best fit multiplicative expression for absorption.

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Figure 3: Measured microwave absorption (normalized by number mixing ratio) for gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere at 8.42 GHz. Mixing ratio was obtained from data in figure 4.Dashed line represents best-fit multiplicative expression.

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Figure 5: Comparative 13 cm opacity of several constituents in the middle atmosphere of Venus, assuming the pressence of gaseous(g)  $H_2SO_4$  (20ppm), gaseous SO\_(150ppm), gaseous H\_2O (1000 ppm),  $CO_2(95\%)$  and liquid<sup>2</sup>(1)  $H_2SO_4$  (100 mg per cubic meter in the 48 to 50 km altitude range). We assume the cloud-related gases to be depleted above the cloud layer (50 km and above).

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Figure 6: Saturation densities for gaseous sulfuric acid  $(H_2SO_4)$  under conditions for the Venus middle atmosphere. Dashed lines represent data from Ayers <u>et al.</u> (1980) and Gmitro and Vermeulen (1964). Solid line represents data from this work. The illustrated points represent the abundances of gaseous  $H_2SO_4$  required to explain the radio occultation results at 13 cm, using the laboratory measurements described.

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# IX. APPENDICES

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of Gaseous Sulfuric Acid Under Simulated Venus Conditions

P. G. Steffes, P. S. Stellitano, R. C. Lott (Georgia Institute of Technology)

Based on initial laboratory studies, it has been suggested by Steffes and Eshleman (1982, Icarus 51, 322-333) that sulfuric acid vapor contributes significantly to the microwave opacity observed in the 30 to 50 km altitude range of the Venus atmosphere. The development of a facility at Georgia Tech capable of making highly accurate measurements of the microwave opacity of gas mixtures in the 1 to 15 cm wavelength range, under pressure and temperature conditions simulating this altitude range of the Venus atmosphere (pressures ranging to 8 atm, temperatures to 550K), has resulted in absorptivity measurements of gaseous  $H_2SO_4$  in a CO<sub>2</sub> atmosphere which are used for interpretation of both radio occultation and radio astronomical microwave absorptivity data. Besides providing abundance profiles for gaseous H<sub>2</sub>SO<sub>4</sub> in the middle Venus atmosphere, these measurements have also been used to infer the partial pressure of gaseous H<sub>2</sub>SO<sub>A</sub> above liquid H<sub>2</sub>SO<sub>A</sub>, based on the microwave absorptivity of the vapors. Both results are used in modelling the sulfur compound chemistry of the Venus middle atmosphere.

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American Astronomical Society Abstract submitted for the DPS Meeting

Date Submitted \_\_\_\_\_ FORM VERSION 1/84 APPENDIX 2: Communications with Reprentatives regarding the planetary exploration program.

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# March 7, 1984

Congressman Wyche Fowler 1210 Longworth House Office Building Washington, DC 20515

Dear Congressman Fowler:

As a supporter of the Space Sciences and Solar System Exploration, I thought I would take this opportunity to extend our congratulations and heartfelt thanks for your support to these programs, and to alert you to potential problems with the FY 1985 budget for the solar system exploration program within NASA as released by OMB. We were encouraged to see inclusion of start-up funding for the MGCO (Mars Geoscience and Climatology Orbiter) mission, but were deeply concerned by the significant reduction in funding for the Research and Analysis program. A large portion of these funds is provided to university scientists to support research at the highest levels of excellence. It has been pointed out by William L. Quaide, Chief Scientist of NASA's Solar System Exploration Division, that an augmentation of \$15 million will be needed to maintain the supporting level included in the FY 84 appropriation. An augmentation for the research and analysis program is included in the president's budget, but only for \$6 million, which would fall far short of the 1984 level.

The future of U.S. leadership in this field, which you have characterized as "prestigious and technology-expanding," lies not simply in the development by industry of spacecraft hardware, but in the elements of basic research and analysis, which involve students and faculty at American universities. Your support of such research in the past has been greatly appreciated. We hope to see it continue in the current and future sessions of Congress.

Sincerely, \_ //

Paul G. Steffes Assistant Professor

PGS/jd

cc: D.T. Paris, Director School of Electrical Engineering

PLEASE RESPOND TO:

WASHINGTON OFFICE: ONGWORTH HOUSE OFFICE BUILDING WASHINGTON, D.C. 20515 (202) 225-3801

DISTRICT OFFICE: ( 425, WILLIAM-OLIVER BUILDING 32 PEACHTREI STREET ATLANTA, GEORGIA 30303 (404) 588-8207 WYCHE FOWLER, JR. STH DISTRICT, GEORGIA

COMMITTEE ON

-22-

Congress of the United States

House of Representatives Washington, D.C. 20515 March 22, 1984 SELECT COMMITTEE ON INTELLIGENCE

Mr. Paul Steffes Assistant Professor Georgia Institute of Technology School of Electrical Engineering Atlanta, Georgia 30332

Dear Mr. Steffes:

Thank you for your letter concerning appropriations for the solar system exploration program, specifically funding for the Research and Analysis program.

The House Committee on Science and Technology recently completed consideration for the NASA Authorization Bill. \$59.5 million had been allocated in the Fiscal Year 1984 budget for Research and Analysis under planetary exploration. The Reagan Administration had requested \$54.5 million for FY'85. However, the Committee decided to recommend \$64.5 million. Of the extra \$10 million, \$2 million is slated to go towards lab equipment, and \$8 million for basic planetary research at universities. The bill goes to the Rules Committee next week, and should reach the full House within the next two weeks.

As you know, I strongly support the role of the United States as the world leader in the space sciences. In pursuit of this goal, I contacted the members of the Committee of Science and Technology, urging them to strengthen Congressional commitment to our space science programs. I was pleased by their response in the NASA Authorization mark-up. I have enclosed a copy of this letter for your interest.

I appreciate your interest in this important issue, and hope you will continue to inform me of other matters in which you have an interest.

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#### REPORT

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# TO THE

# NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

# SEMIANNUAL STATUS REPORT #3

for

GRANT NAGW-533

#### LABORATORY EVALUATION AND APPLICATION OF

#### MICROWAVE ABSORPTION PROPERTIES UNDER SIMULATED

## CONDITIONS FOR PLANETARY ATMOSPHERES

Paul G. Steffes, Principal Investigator

February 1, 1985 through July 31, 1985

Submitted by

Professor Paul G. Steffes School of Electrical Engineering Georgia Institute of Technology Atlanta, Georgia 30332-0250

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#### I. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often lead to significant misinterpretation of available opacity data. Steffes and Eshleman (1981) showed that under environmental conditions corresponding to the middle atmosphere of Venus, the microwave absorption due to atmospheric SO2 was 50 percent greater than that calculated from Van Vleck-Weiskopff theory. Similarly, the opacity from gaseous  $H_2SO_4$  was found to be a factor of 7 greater than theoretically predicted for conditions of the Venus middle atmosphere (Steffes and Eshleman, 1982). The recognition of the need to make such measurements over a range of temperatures and pressures which correspond to the periapsis altitudes of radio occultation experiments, and over a range of frequencies which correspond to both radio occultation experiments and radio astonomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements.

In the initial year of Grant NAGW-533 (i.e., February 1, 1984 through January 31, 1985), this facility was developed, and then operated, in order to evaluate the microwave absorbing properties of gaseous sulfuric acid  $(H_2SO_4)$  at 13.4 and 3.6 cm wavelengths, under Venus atmospheric conditions. The

results were then applied to measurements from Mariner 5, Mariner 10, and Pioneer-Venus Radio Occultation experiments, to determine abundancies of gaseous sulfuric acid in the Venus atmosphere, with accuracies exceeding those achieved with in-situ instruments. Measurements of the microwave properties of the vapors accompanying liquid H<sub>2</sub>SO<sub>4</sub> also resulted in more accurate estimates of the vapor pressure behavior of sulfuric acid, which are critical for modeling the behavior and structure of the Venus atmosphere. The results of this work are to be published in a paper entitled, "Laboratory Measurements of the Microwave Opacity and Vapor Pressure of Sulfuric Acid Vapor under Simulated Conditions for the Middle Atmosphere of Venus," which will be published in Icarus. (Note: As specified in the NASA Provisions for Research Grants and Cooperative Agreements, preprints of the original manuscript were sent to the NASA Technical Officer and the NASA Scientific and Technical Information Facility on February 2, 1985. A copy of the revised manuscript, i.e., postpeer review, is attached as Appendix 3.)

During the first six months of the current grant year (February 1, 1985 through July 31, 1985), our work has concentrated on making laboratory measurements of the microwave absorption from gaseous  $H_2SO_4$  at wavelengths from 1 to 3 cm under simulated Venus conditions. Additional measurements of the vapor pressure behavior of sulfuric acid have also been made. Our goal will be to apply these results to radio astronomical observations of Venus which have been made in the wavelength range, in order to better model the structure of  $H_2SO_4$  abundance in the Venus atmosphere, and to resolve temporal variations of its abundance on a planet-wide basis. In addition, we have begun the process of redesigning and refitting the laboratory system so as to permit measurements of the microwave absorptivity of ammonia (NH<sub>3</sub>), methane

(CH<sub>4</sub>), and other potential microwave absorbers under simulated conditions for the outer planets (Jupiter, Saturn, Uranus, and Neptune). In May, a paper entitled, "Laboratory Measurements of Microwave Absorption from Gaseous Atmospheric Constituents under Conditions for the Outer Planets," was presented at the <u>Conference on the Jovian Atmospheres</u>, held at the Goddard Institute for Space Studies in New York, in which feedback from other experimenters was sought in order to optimize the yield from our outer planets simulations. (See Appendix 1.) This feedback has helped significantly in our design of the outer planets atmospheric simulator.

Our goals for the second half of the current grant year (August 1, 1985 through January 31, 1986) include the analysis and application of the recently obtained 1 to 3 cm wavelength range absorptivity data for gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere. The unique frequency and pressure dependences exhibited by gaseous  $H_2SO_4$  in these wavelength ranges may finally explain what were thought to be inconsistencies between absorption measurements at 13.3 and 3.6 cm wavelengths and those obtained from radio astronomical observations in the 1 to 3 cm wavelength range (see Steffes and Eshleman, 1982). In addition, this new absorptivity data will allow determination of the <u>minimum</u> altitude at which gaseous  $H_2SO_4$  can exist while still being consistent with models for total atmospheric opacity based on radio emission studies. The other major effort will be to complete work on reconfiguring the laboratory so as to allow measurements of the microwave opacity, in the 1 to 20 cm wavelength range, of constituent gases under simulated conditions for the outer planets. We hope to conduct the first of such measurements in December.

# II. THE GEORGIA TECH RADIO ASTRONOMY AND PROPAGATION (R.A.P.) FACILITY

The basic configuration of the planetary atmosphere simulator developed at Georgia Tech for use in measurement of the microwave absorptivity of gases under simulated conditions for planetary atmospheres is described at length in the first <u>Annual Status Report</u> for Grant NAGW-533 (February 1, 1984 through January 31, 1985). It is also discussed at length in Steffes (1985), which is attached as Appendix 3. The major new addition made to the laboratory apparatus is a microwave resonator capable of making absorptivity measurements in the 8 to 27 GHz range (1 to 3.7 cm wavelengths). This significantly extends the operating range beyond the previous 1.5 to 8.5 GHz (3.6 to 20 cm) operating range. As shown in Figure 1, the system is currently configured so as to be used in measuring microwave absorption (2 to 27 GHz) from gaseous  $H_2SO_4$ under simulated conditions for the middle atmosphere of Venus (total pressures from 1 to 6 atm, temperatures from 500 to 575 K).

Adding a resonator capable of operating in the 8 to 27 GHz frequency range was more difficult than expected, especially considering the high temperatures and pressures involved. It was decided to use coaxial cable to interconnect the high frequency resonator to the microwave instrumentation, since hermetically-sealed coaxial feed-through connectors were available which would allow operation to 30 GHz while still maintaining the pressure integrity of the chamber. The type of coaxial cable required for the higher frequency resonator was different from that used in the 1.5 to 8.5 GHz range, in that a solid metallic jacket is required to prevent signal leakage, as opposed to the metallic braid used in lower frequency cables. Unfortunately, even when hightemperature solder was used, connector failure occurred for these cables at

temperatures above 500 K. This was due to the extrusion of the PTFE (Teflon) dielectric material within the cable placing mechanical stresses on the connectors. As a result, a limited production air-dielectric cable was obtained which employs a PTFE spline to space the center conductor from the solid metallic outer conductor. The connectors for this specialized cable have been difficult to obtain since special production runs (and accompanying set-up charges) are usually required. Fortunately, we were able to combine the production of our connectors with those from other customers so as to avoid these charges. The result has been a system capable of successfully measuring the microwave absorption from gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere at wavelengths from 1 to 20 cm, at pressures from 1 to 6 atmospheres, and at temperatures from 500 to 570 K. (Lower temperatures can, of course, be achieved but the resulting gaseious  $H_2SO_A$  abundance would be too small so as to provide measurable absorptivity.) It was originally hoped that yet another resonator could be placed within the resonator so as to provide measurements in the 27 to 40 GHz frequency range (0.75 to 1.1 cm wavelength range). Because of the minute losses being measured, a Fabry-Perot type resonator would be required. Unfortunately, the volume required for a Fabry-Perot resonator and the difficulties in coupling signals to it through pressuresealed waveguides have made that impractical.

The pressure vessel and its accompanying resonators are designed to be usable over an extremely wide range of temperatures. The extremely high temperatures (up to 600 K) developed for the Venus atmosphere simulations by the oven/temperature chamber (see Figure 1) can be replaced by extremely low temperatures created by a freezer/temperature chamber for simulations of the outer planets, as shown in Figure 2. Such a freezer has been recently

procured for use in such measurements. The unit, a Revco/Rheem ULT-7120D, has an internal volume of 193 liters (which is capable of containing the pressure vessel with resonators), and is capable of maintaining temperatures as low as 153 K. Since access to the freezer is through the top of the unit, a special lifting frame and pulley system has been constructed which allows easy movement of the pressure vessel and accompanying resonators in and out of the freezer. The result is a system capable of measuring microwave absorption from 1.5 to 27 GHz (1 to 20 cm wavelength) at pressures to 8 atmospheres and at temperatures as low as 153 K. The minimal measurable absorption absorptivity, or sensitivity, for this system when operated at 170 K is shown in Figure 3, as a function of frequency.

Another more critical limitation on how low the temperatures can be taken (while still being able to measure microwave absorption) is the saturation vapor pressure of the gas being tested. As shown in Table 1, the lowest temperature for which sufficient quantities of gaseous  $NH_3$  would still exist so that the microwave absorption in an  $H_2$  atmosphere would be measurable would be approximately 155 K.

A final critical issue which greatly affects the use of the simulator in outer planets simulations is the use of hydrogen  $(H_2)$  at pressures reaching 8 atmospheres. In previous simulations, small leakages from the pressure vessel presented little or no danger to the experimenters. The use of hydrogen will require a special set of procedures and a special ventilation system to be used. Initial pressure tests of the system, as configured for outer planets simulation, will be conducted with nitrogen  $(N_2)$ . Not only will this be a safer way to test the system, but it will also allow measurement of the collisional microwave absorption from  $N_2$ , which may be the source of the

3.6 cm (8.4 GHz) opacity detected by Voyager 1 radio occultation studies of the Titan atmosphere. (See Lindal <u>et al.</u>, 1985.) We hope to begin these tests by December.

# III. RESULTS OF LABORATORY MEASUREMENTS AND THEIR APPLICATION

Our goal for the first six months of the current grant year has been to complete laboratory measurements of the microwave absorption from gaseous  $H_2SO_4$  in the 1 to 3 cm wavelength range, under simulated conditions for the middle atmosphere of Venus, and to obtain additional measurements of the vapor pressure behavior of gaseous  $H_2SO_4$  above liquid sulfuric acid. The additional vapor pressure measurements made have been consistent with the vapor pressure expression derived from earlier data. It should be noted, however, that the expression has been revised from its earlier form to reflect a higher percentage dissociation of the  $H_2SO_4$  vapor above liquid sulfuric acid into  $H_2O$  and  $SO_3$ . The complete set of vapor pressure measurements and the best-fit expression are plotted in Steffes (1985). (See Appendix 3, Figure 4.)

The measurements of microwave absorption in the 1 to 3 cm wavelength range are currently being evaluated, but one major result has been the extremely low absorptivity observed in the 1.1 to 1.8 cm wavelength range. For example, measurements made at 21.63 GHz (1.38 cm wavelength) show an opacity for a .36% mixture of gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere (with a total pressure of 6 atmospheres and temperature of 570 K) of less than 9 dB/km. This is far below the opacity predicted by using previous measurements at 2.2 GHz and 8.4 GHz and assuming a simple  $f^2$  dependence such as exhibited by  $SO_2$  and  $CO_2$ . In fact, this even implies that using the measured frequency dependences for the microwave absorption from  $H_2SO_4$  in a  $CO_2$  atmosphere in the

2.2 to 8.4 GHz range to predict absorption at 21.6 GHz (1.38 cm wavelength) would result in overstating the absorption at 1.38 cm by at least a factor of 2.

Such behavior implies that the contribution of gaseous  $H_2SO_A$  to the overall non-CO2 opacity inferred from radio astronomical observations at the 1.35 cm is minimal. Such a result is not surprising in light of the calculations done by Janssen and Klein (1981) which attribute nearly all of the non- $CO_2$  opacity at 1.35 cm in the Venus atmosphere to  $SO_2$ . This behavior is likewise consistent with the calculated  $H_2SO_4$  resonance frequencies computed (R. Poynter, J.P.L., personal communication. by Poynter. Note that the results of the resonance calculations are shown in Cimino, 1982.) These calculations are based on rotational constants computed using measurements of H<sub>2</sub>SO<sub>4</sub> resonances in the 60 to 120 GHz range by Kuczkowski <u>et al.</u> (1981). It is significant that there is a notable absence of  $H_2SO_4$  resonances in the 15 to 30 GHz frequency range, which would be consistent with our measurements. A computation of the microwave spectrum from  $H_2SO_4$ , made by Allen, which is presented in Cimino (1982), showed significantly more absorption from  $H_2SO_A$ in this same frequency range than we have actually measured. This can be explained by the fact that a pressure-broadened linewidth parameter of 7.2 MHz was assumed to  $H_2SO_4$  in a  $CO_2$  atmosphere. It appears that using a smaller broadening parameter will result in a spectrum which will be consistent with our measurements.

Thus, our laboratory measurements indicate that while radio occultation measurements of 13 and 3.6 cm, and radio astronomical measurements at wavelengths longer than 2 cm, measure absorption which is predominated by gaseous  $H_2SO_4$ ; opacity measured by radio astronomical observations in the 1 to 2 cm

wavelength range is predominated by  $SO_2$  at higher altitudes, and by  $CO_2$  at lower altitudes. We intend to present these results at the October 1985 AAS/DPS meeting.

# IV. PUBLICATIONS AND INTERACTION WITH OTHER INVESTIGATORS

In the first six months of the current grant year, a paper was completed and revised for publication in Icarus, describing results and applications of experiments performed during the first year of the grant (P. G. Steffes, 1985, "Laboratory Measurements of the Microwave Opacity and Vapor Pressure of Sulfuric Acid Vapor under Simulated Conditions for the Middle Atmosphere of Venus," attached as Appendix 3.) In May, a paper was presented at the Conference on Jovian Atmospheres, at the Goddard Institute for Space Studies (New York) entitled, "Laboratory Measurements of Microwave Absorption from Gaseous Atmospheric Constituents under Simulated Conditions for the Outer Planets." (See Abstract, Appendix 1.) This paper described our plans and capabilities for simulating outer planets atmospheres and measuring microwave properties of those atmospheres. Much positive feedback was received with regard to the need for such measurements, and discussions were held with I. de Pater of UC/Berkeley and A. Kliore of JPL regarding application of such data to radio astronomical observations, and Galileo probe measurements, respectively. Contact with both is expected to continue as the measurements are made. Contacts have also been maintained with groups at the Stanford Center for Radar Astronomy (V. Eshleman, director), and JPL (Drs. Michael J. Klein and Samuel Gulkis).

During the second half of the current grant year, an Invited Paper will be presented at the International Association of Meteorology and Atmospheric

Physics (IAMAP) Assembly (August 12, 1985) entitled, "Microwave Absorption from Cloud-Related Gases in Planetary Atmospheres," which summarizes our work over the past 18 months and its applications. In addition, a paper will be presented at the October meeting of the Division of Planetary Sciences of the American Astronomical Society (DPS/AAS) on the complete microwave spectrum of  $H_2SO_4$  in a  $CO_2$  atmosphere (from 1 to 15 cm) and its application to understanding the microwave emission spectrum from Venus. We may also submit a short paper to <u>Icarus</u> on this same subject. We have also maintained contact with our congressional delegation, keeping them aware of our work, and the need for continued support to the solar system exploration program. (See Appendix 2.)

#### V. CONCLUSION

During the first half of the current grant year (February 1, 1985 through July 31, 1985) we have completed measurements of the microwave absorption from gaseous  $H_2SO_4$  in the 1 to 3 cm wavelength range, as well as having applied results from earlier measurements at 3.6 and 13.4 cm wavelengths to microwave absorptivity data from radio occultation measurements at those wavelengths, in order to derive abundance profiles for gaseous  $H_2SO_4$ . We also completed design and began reconfiguration of the system so as to conduct outer planets simulations.

In the second half of the grant year, we intend to complete the outer planets simulator construction, and make some preliminary measurements of the microwave absorption from nitrogen  $(N_2)$  to simulate Titan, and from ammonia  $(NH_3)$  in a hydrogen atmosphere  $(H_2)$  in order to simulate Jovian atmospheres. We will likewise work on the application of our newly derived 1 to 3 cm  $H_2SO_4$ spectrum to a wide range of radio astronomical data.

#### VI. REFERENCES

Cimino, J. B. (1982). The composition and vertical structure of the lower cloud deck on Venus. Icarus 51, 334-357.

Janssen, M. A. and Klein, M. J. (1981). Constraints on the composition of the Venus atmosphere from microwave measurements near 1.35 cm wavelength. <u>Icarus</u> 46, 58-69.

Kuczkowski, R. L., Suenram, R. D., and Lovas, F. J. (1981). The microwave spectrum, structure, and dipole moment of sulfuric acid. J. Amer. Chem. Soc. 103, 2561-2566.

Steffes, P. G. and Eshleman, V. R. (1981). Laboratory measurements of the microwave opacity of sulfur dioxide and other cloud-related gases under simulated conditions for the middle atmosphere of Venus. Icarus 48, 180-187.

Steffes, P. G. and Eshleman, V. R. (1982). Sulfuric acid vapor and other cloud-related gases in the Venus atmosphere: abudances referred from observed radio opacity. Icarus 51, 322-333.

VII. KEY FIGURES

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Figure 1: Blockdiagram of updated Georgia Tech Planetary Atmospheres Simulator, as configured for measurements of the microwave properties of gaseous  $H_2SO_4$  under simulated conditions for the Venus atmosphere over the 1 to 20 cm wavelength range.


Figure 2: Block diagram of Georgia Tech Planetary Atmospheres Simulator, as configured for measurements of microwave refraction and absorption of gases under simulated conditions for the outer planets.



# TABLE I

Constituent Gas	Abundance of Gas in H <sub>2</sub> atmosphere (6 atm total pressure) required so as to measure microwave absorption at lowest Temperature	Partial Pressure of Gas (in atm.) required so as to exhibit detectable absorption in H <sub>2</sub> atmosphere. (P <sub>H</sub> = 6 atm.) H <sub>2</sub>	Lowest Possible Temperature for which required abundance can be achieved (K)
NH <sub>3</sub>	60 ppm	$3.6 \times 10^{-4}$	155
H <sub>2</sub> -H <sub>2</sub> and H <sub>2</sub> -He* (collisional)	80% H <sub>2</sub> : 20% He	6	20
CH <sub>4</sub> **	33%	2	120
CO	Not Detectable in the 1 to 20 cm range by this system		

# MINIMUM GASEOUS ABUNDANCES NECESSARY SO AS TO BE MEASURABLE BY THE SYSTEM

\* Will provide baseline reference for measurements of other constituents

\*\* The theoretically predicted absorption for methane under these conditions is approximately 80% below the minimum detectable absorption for the system. However, it was felt that this is the largest mixing ratio for which hydrogen broadening would still predominate.

# VIII. APPENDICES

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APPENDIX I : Abstract of paper presented at the Conference on the Jovian Atmospheres (Goddard Institute for Space Studies, New York City, May 1985.)

# Laboratory Measurements of Microwave Absorption from Gaseous Atmospheric Constituents under Conditions for the Outer Planets

## P. Steffes (Georgia Institute of Technology)

Quite often, the interpretive work on the microwave and millimeterwave absorption profiles, which are inferred from radio occultation measurements or radio astronomical observations of the outer planets, employs theoretically-derived absorption coefficients to account for contributions to the observed opacity from gaseous constituents. Variations of the actual absorption coefficients from those which are theoretically-derived, especially under the environmental conditions characteristic of the outer planets, can result in significant errors in the inferred abundances of the absorbing constituents. The recognition of the need to make laboratory measurements of the absorptivity of gases such as NH3, CH4, and  $H_20$  in a predominantly  $H_2$  atmosphere, under temperature and pressure conditions simulating the outer planets' atmospheres, and at wavelengths corresponding to both radio occultation and radio astronomical observations, has led to the development of a facility capable of making such measurements at Georgia Tech. We describe the laboratory measurement system, the measurement techniques, and the proposed experimental regimen for Summer 1985; with the goal of obtaining feedback from interested investigators on the relative priorities of the various proposed measurements to be made on specific constituents at specific wavelengths.

#### MARK O. HATFIELD, OREGON, CHAIRMAN

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# J. KEITH KENNEDY, STAFF DIRECTOR FRANCIS J. SULLIVAN, MINORITY STAFF DIRECTOR

**APPENDIX II:** 

# United States Senate

COMMITTEE ON APPROPRIATIONS WASHINGTON, DC 20510

May 30, 1985

Professor Paul G. Steffes Georgia Institute of Technology School of Electrical Engineering Atlanta, Georgia 30332

Dear Professor Steffes:

Thank you for contacting me concerning NASA.

I believe that the exploration of space and the utilization of knowledge gained from such efforts would prove to be of inestimable value to all people. It is important that we proceed with the development of space exploration technology and I will support such efforts in the Congress.

Again, thank you for contacting me.

Sincerely,

Mack Mattingly

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MM/jbh

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**APPENDIX 3:** 

## LABORATORY MEASUREMENTS OF THE MICROWAVE OPACITY AND VAPOR PRESSURE OF SULFURIC ACID VAPOR UNDER SIMULATED CONDITIONS FOR THE MIDDLE ATMOSPHERE OF VENUS

Paul G. Steffes

School of Electrical Engineering Georgia Institute of Technology Atlanta, Georgia 30332

Submitted to Icarus

February 1985 Revised, July 1985

Manuscript Pages: 28 Figures: 6

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Proposed running head: Sulfuric Acid Vapor at Venus

Mail proofs to:

Professor Paul G. Steffes School of Electrical Engineering Georgia Institute of Technology Atlanta, Georgia 30332 (404) 894-3128

#### ABSTRACT

Microwave absorption observed in the 35 to 48 km altitude region of the Venus atmosphere has been attributed to the presence of gaseous sulfuric acid  $(H_2SO_4)$  in that region. This has motivated the laboratory measurement of the microwave absorption at 13.4 and 3.6 cm wavelengths from gaseous  $H_2SO_4$  in a CO2 atmosphere under simulated conditions for that region. As part of the same experiments, upper limits on the saturation vapor pressure of gaseous  $H_2SO_4$  have also been determined. The measurements for microwave absorption have been made in the 1 to 6 atmosphere pressure range, with temperatures in the 500 to 575 K range. Using a theoretically-derived temperature dependence, the best-fit expression for absorption from gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere at the 13.4 cm wavelength is 9.0 x  $10^9 q(P)^{1/2} T^{-3} (dB \text{ km}^{-1})$ , where q is the  $H_2SO_4$  number mixing ratio, P is the pressure in atmospheres, and T is the temperature in Kelvins. The best-fit expression for absorption at the 3.6 cm wavelength is 4.52 x  $10^{10}$  q(P)  $^{0.85}$  T<sup>-3</sup> (dB km<sup>-1</sup>). The inferred H<sub>2</sub>SO<sub>4</sub> vapor pressure above liquid  $H_2SO_4$  corresponds to  $\ln p = 8.84 - 7220/T$  where p is the H<sub>2</sub>SO<sub>4</sub> vapor pressure (in atm) and T is the temperature in Kelvins. These results suggest that abundances of gaseous  $H_2SO_4$  on the order of 15 to 30 ppm could account for the microwave absorption observed by radio occultation experiments at 13.3 and 3.6 cm wavelengths. They also suggest that such abundances would correspond to saturation vapor pressure existing at or above the 46 to 48 km range, which correlates with the observed cloud base. It is suggested that future measurements of absorption in the 1 to 3 cm wavelength range will provide additional tools for monitoring variations in  $H_2SO_4$  abundance via radio occultation and radio astronomical observations.

#### I. INTRODUCTION

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different from those of the planetary atmosphere being studied, often lead to significant misinterpretation of available opacity data. Steffes and Eshleman (1981) showed that under environmental conditions corresponding to the middle atmosphere of Venus, the microwave absorption at 13 cm and 3.6 cm wavelengths due to atmospheric SO<sub>2</sub> was approximately 50 percent greater than that calculated from Van Vleck-Weiskopff theory. Similarly, the opacity from gaseous  $H_2SO_4$  was found to be a factor of 7 greater than theoretically predicted for conditions of the Venus middle atmosphere (Steffes and Eshleman, 1982). The recognition of the need to make such measurements over a range of temperatures and pressures which correspond to the altitudes at which significant opacity has been detected, and over a range of frequencies which correspond to both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements.

In its initial year of operation, this facility has been used to evaluate the microwave absorbing properties and limits to the saturation vapor pressures for gaseous sulfuric acid  $(H_2SO_4)$  under simulated conditions for the middle atmosphere of Venus. This paper describes the methodology and results

of laboratory measurements of the absorptivity of gaseous  $H_2SO_4$  in a predominantly  $CO_2$  atmosphere at wavelengths corresponding to those used in spacecraft radio occultation experiments (13.4 and 3.6 cm) and for pressures at which these experiments detected significant atmospheric opacity. Also, we describe the results of measurements of the saturation vapor pressure of sulfuric acid. We also discuss the effect these measurements have on modeling  $H_2SO_4$  in the Venus atmosphere. These results are then applied to measurements from Mariner 5, Mariner 10, and Pioneer-Venus Radio Occultation experiments, to determine limits on abundances of gaseous sulfuric acid in the Venus middle atmosphere. We conclude by outlining plans for laboratory measurements of the opacity of gaseous  $H_2SO_4$  at shorter wavelengths (1-3 cm) and for application of these measurements to a wider range of absorptivity data.

#### **II. EXPERIMENTAL APPROACH**

The experimental approach used to measure the microwave absorptivity of gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere is similar to that used previously by Steffes and Eshleman (1981 and 1982). As can be seen in Figure 1, the absorptivity is measured by observing the effects of the introduced gas mixture on the Q, or quality factor, of a cavity resonator at its particular resonances near 2.24 GHz and 8.42 GHz. The changes in the Q of the resonator which are induced by the introduction of an absorbing gas mixture can be monitored by the high resolution microwave spectrum analyzer, since Q is simply the ratio of the cavity resonant frequency to its half-power bandwidth. For relatively low-loss gas mixtures, the relation between the absorptivity of the gas mixture and its effect on the Q of the resonator is straightforward:

$$\alpha \simeq (Q_{\rm L}^{-1} - Q_{\rm C}^{-1}) \pi / \lambda \tag{1}$$

where  $\alpha$  is absorptivity of the gas mixture in Nepers km<sup>-1</sup>. (Note, for example, that an attenuation constant or absorption coefficient or absorptivity of 1 Neper km<sup>-1</sup> = 2 optical depths per km (or km<sup>-1</sup>) = 8.686 dB km<sup>-1</sup>, where the first notation is the natural form used in electrical engineering, the second is the usual form in physics and astronomy, and the third is the common (logarithmic) form. The third form is often used in order to avoid a possible factor-of-two ambiguity in meaning.)  $Q_{\rm L}$  is the quality factor of the cavity resonator when the gas mixture is present,  $Q_{\rm C}$  is the quality factor of the cavity resonator in a vacuum, and  $\lambda$  is the wavelength (in km) of the test signal in the gas mixture.

In order to obtain a gas mixture with a sufficient amount of  $H_2SO_4$  vapor so that the microwave absorption is detectable, the system must be operated at temperatures exceeding 450 K. While this is suboptimal in that the temperatures at the altitudes where radio occultation experiments have detected microwave opacity range from 350 K to 450 K, temperature dependences measured for similar gases (such as  $SO_2$ ) can be used to estimate temperature effects in that range. Two different approaches are used to infer  $H_2SO_4$  vapor pressure. With the first, the volume of liquid sulfuric acid which is vaporized to generate the gaseous  $H_2SO_4$  is determined to a high accuracy (up to  $\pm 0.005$  ml). It is then possible to compute an upper limit for the partial pressure of gaseous  $H_2SO_4$  using the ideal gas equation, the measured change in liquid volume, and published densities for  $H_2SO_4$  liquid. The second method for accurately determining amounts of liquid-derived vapors measures the refractivity of those vapors. Since the index of refraction (relative to unity) is proportional to the vapor abundance, the system's ability to accurately measure the

refractivity of such vapors can also be used to infer the relative vapor abundance/pressure. Note that it is not yet possible to use this approach for accurate determination of the absolute vapor pressure from gaseous  $H_2SO_4$ , since accurate refractivity data for gaseous  $H_2SO_4$  is not currently available. While neither method is able to accurately resolve dissociation of  $H_2SO_4$  into  $H_2O$  and  $SO_3$ , an upper limit for  $H_2SO_4$  vapor abundance can be inferred, using the first method, which is accurate to  $\pm 2$  percent at a temperature of 500 K.

#### III. MEASUREMENT TECHNIQUE

As shown in Figure 1, a flask is filled with a precisely known volume of liquid sulfuric acid (99 percent by weight) at room temperature. For nearly all of the experiments, the initial volume used was 2.5 ml (measured at room temperature). Smaller quantities were also tried, with no significant difference in results. The volume measurements are made using a 1 ml syringe with .01 ml gradiations. For volumes less than 1 ml, repeatable accuracies of better than .005 ml have been obtained. The entire system is then heated and allowed to thermally stabilize at the chosen experimental temperature (approximately 500-575 K), which requires approximately six hours. The temperature is monitored using a thermocouple which is placed in the center of the pressure vessel, near the microwave resonator, which is one of the coldest places in the system. Thermal equilibrium within the system is achieved by preheating the system for 6 to 8 hours before beginning the experiments. Since the thermal time constant is approximately 2 hours and 15 minutes, this assures a relatively constant temperature within the chamber after heating. Another technique used in monitoring the heating of the resonator within the chamber is to monitor the resonant frequencies of the resonator. As the

resonator is heated, the resonant frequencies drop due to thermal expansion. Thus, when thermal stability is reached, the resonant frequencies likewise After thermal stability is reached, a vacuum is drawn in the stabilize. pressure vessel containing the microwave cavity resonator, and the bandwidth and center frequency of the 13.4 cm and 3.6 cm resonances are then measured. A valve is then opened which allows the sulfuric acid vapor eluting from the flask to fill the pressure vessel (0.031 cubic meters of open volume with resonator in place) and reach vapor pressure equilibrium with the liquid  $H_2SO_4$ . Note that all components which contact the gaseous sulfuric acid are maintained at the same temperature as the flask, so as to avoid condensation. In addition, because of the high temperatures and corrosive vapor involved, all tubing, valves, and the pressure chamber itself are fabricated from stainless steel. Gaskets and cables are fabricated from either viton or PTFE. These steps not only insure the survival of components under the test conditions, but also reduce possible reactions of the components with the sulfuric acid vapor and possible outgassing of vapors related to the materials from which the components are fabricated.

As  $H_2SO_4$  vapor fills the chamber, changes in the resonance center frequency are observed. These changes, which reach over 400 kHz at the 13.4 cm resonance, are related to the  $H_2SO_4$  vapor abundance. After approximately 10 minutes, the frequency shift ceases, as vapor pressure equilibrium is reached. The valve to the reservoir flask is then closed, and  $CO_2$  is admitted to the chamber containing the  $H_2SO_4$  vapor. For this experiment, a total pressure of 6 atm was used. The  $CO_2$  gas is admitted to the chamber at a sufficiently slow rate so as not to significantly affect the temperature within the chamber. The bandwidth of the cavity is then measured and compared

with its value when the chamber was evacuated in order to determine the absorptivity of the  $CO_2/H_2SO_4$  gas mixture at 6 atm total pressure. The total pressure is then reduced to 4 atm by venting, and the bandwidth is again Subsequent measurements are likewise made at lower pressures in measured. order to determine absorptivities at those pressures. The pressure vessel is then evacuated and the bandwidth again measured so as to assure no variation (either due to thermal shift or chemical reaction) of the Q of the evacuated resonator has occurred. After the system has been allowed to cool, the volume of the remaining sulfuric acid liquid (at room temperature) is measured and compared with the initial volume measured (at room temperature) in order to set an upper limit on  $H_2SO_4$  vapor present in the gas mixture tested. This approach has the advantage that the same gas mixture is used for the absorptivity measurements at the various pressures. Thus, even though some uncertainty may exist as to the mixing ratio of the initial mixture, the mixing ratios at subsequent pressures will be the same, and thus the uncertainty for any derived pressure dependence will only be due to the accuracy limits of the absorptivity measurements, and not uncertainty in the mixing ratio. (This assumes that the mixing ratio is small, so that foreign-gas broadening predominates, as is the case for our measurements.) Similarly, measurements of the frequency dependence of the absorptivity from a  $CO_2/H_2SO_4$  mixture will likewise be immune to mixing ratio uncertainty, as long as foreign-gas broadening predominates.

While the overall equipment configuration and experimental approach for these measurements is similar to that used by Steffes and Eshleman (1982) for measurement of the absorptivity of gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere at a single pressure and temperature, several major differences between the current system and those used previously exist:

- (1) The pressure containment vessel is large enough (0.33 m diameter, 0.38 m height) so that it will allow the use of resonators which operate at frequencies as low as 1.5 GHz (20 cm wavelength), and yet still be heated to temperatures over 500 K while maintaining pressures up to 10 atmospheres.
- (2) The resonator used to measure absorption in the 2.2 to 8.4 GHz range (13.4 to 3.6 cm wavelengths) exhibits a higher "quality factor," or Q, due to special construction techniques. These techniques include the use of stainless steel to construct the resonator, and then plating with nickel, copper, and silver. This not only maximizes Q, but insures that the Q will remain high in spite of drastic thermal shifts. As a result, the system sensitivity is greater, resulting in lower measurement uncertainties.
- (3) The use of a digitally-refreshed spectrum analyzer further serves to increase the sensitivity of the system. As a consequence, when measurements are to be made of the microwave absorption from gases which are obtained from liquid-derived vapors, a smaller amount of source liquid can be used, allowing operation at lower temperatures without condensation.
- (4) Uncertainties of mixing ratios of gases obtained from liquid-derived vapors have been greatly reduced for two reasons. First, the amounts of liquid used to generate these gases can be determined with a volume accuracy of up to  $\pm 0.005$  ml. Thus, when an amount of liquid becomes vapor, it becomes possible to accurately compute an upper limit for the partial pressure due to that vapor using the ideal gas equation and the measured change in liquid volume.

Secondly, the high sensitivity and high stability of the microwave spectrum analyzer allow accurate measurements of the refractivity of introduced vapors, which can also be used to infer relative vapor abundance.

#### IV. MEASUREMENT RESULTS

A wide range of laboratory measurements of the microwave absorpting properties of gaseous  $H_2SO_4$  in a predominantly  $CO_2$  atmosphere have been made under temperature and pressure conditions (temperatures to 575 K and pressures to 6 atm.) simulating the middle atmosphere (35-50 km altitude) of Venus. The measurements reported here were made at frequencies of 2.24 and 8.44 GHz (13.4 and 3.6 cm wavelengths) in order to allow direct application to absorptivity data from Pioneer-Venus, Mariner 5, and Mariner 10 radio occultation experiments. However, measurements over a wider range of wavelengths are currently underway.

Since absorptivity at radio frequencies is generally directly proportional to the abundance (either by number or by volume) of the absorbing constituent, the results of our measurements, shown in Figures 2 and 3, are plotted as absorptivities normalized by  $H_2SO_4$  mixing ratio. Figure 2 shows the normalized absorptivity of gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere (taken at 570 K temperature and at 2.24 GHz frequency) as a function of pressure. Similarly, Figure 3 shows similar measurements taken around 525 K and at a frequency of 8.42 GHz. The  $H_2SO_4$  mixing ratios used to normalize the plots were obtained by dividing the vapor pressure data from Figure 4 (which were obtained by the measurement techniques described in Sections II and III) by the sum of the  $H_2SO_4$  vapor pressure plus the  $CO_2$  vapor pressure when the

mixture was formed (6 atm for our experiments). The error bars shown in Figures 2 and 3 represent  $1\sigma$  errors in the absorptivity measurements, but do not include uncertainties in mixing ratios. Since the same mixing ratio is used for all measurements at a given temperature, a small scale factor error might be present.

It should be noted that while the techniques used to set upper limits on the  $H_2SO_4$  vapor pressure obtained from the 99.0 percent concentration (by weight) liquid  $H_2SO_A$  solutions used can determine these limits quite accurately, they do not represent direct measurement of H<sub>2</sub>SO<sub>4</sub> vapor pressure. For the results presented in Figure 4, it is assumed that 47 percent of the vaporized  $H_2SO_4$  dissociates to form gaseous  $SO_3$  and  $H_2O$ , as computed by Gmitro and Vermeulen (1964) for 99% solutions. Since neither SO3 nor H2O exhibit measurable microwave absorption in the quantities which would be present (see Steffes and Eshleman 1981, and Ho et al. 1966), this dissociation results in an increase in the measured absorptivity (normalized by number mixing ratio) due to gaseous  $H_2SO_4$ . Also it is assumed that the portion of the vaporized liquid which actually becomes gaseous sulfuric acid is proportional to the number density of sulfuric acid molecules in the liquid solution. It should also be noted that the vapor pressures plotted are for a 99 percent (by weight) solution of sulfuric acid, except for the 4 points taken at the lowest temperatures, where a 95.9 percent solution was used. The predicted difference in  $H_2SO_4$  vapor pressure caused by this lower concentration is a reduction of less than 25 percent (see Gmitro and Vermeulen, 1964). Thus, it was felt that this reduction would only slightly affect our best fit expression plotted for the temperature dependence of the  $H_2SO_4$  vapor pressure:

$$\ln p = 8.84 - 7220/T$$
 (2)

where p is the  $H_2SO_4$  vapor pressure in atmospheres and T is the temperature in Kelvins. The error bars shown for each point in Figure 4 represent 1  $\sigma$  variations in temperature (horizontal axis) and in inferred vapor pressure (vertical axis). The variations in temperature measurement result from limits on thermocouple accuracy, while variations in inferred vapor pressure result from the accuracy limitations on the volume measurement of the vaporized liquid.

Another potential source of error could be opacity from molecules, other than  $H_2SO_4$ ,  $H_2O$ , or  $SO_3$  which accompany sulfuric acid vapor. Two such molecules would be the sulfuric acid dimer (i.e.,  $2-H_2SO_4$ ) and the sulfuric acid hydrate ( $H_2SO_4$   $H_2O$ ). However, electric deflection studies by Kay (1984) concluded that  $H_2SO_4$  dimers are nonpolar, which would suggest little or no microwave opacity. Additionally, no hydrated sulfuric acid molecules were detected above liquid sulfuric acid at 593 K, suggesting a minimal abundance of such molecules.

In Figures 2 and 3, best-fit multiplicative expressions are plotted for the 13.4 and 3.6 cm absorption from gaseous  $H_2SO_4$ . (Note: By "multiplicative" we mean a function which is directly proportional to some power of pressure.) Combining the measured pressure dependence, with an estimated thermal dependence of  $T^{-3}$  (a  $T^{-3.1}$  dependence has been measured for  $SO_2$ , see Steffes and Eshleman, 1981) gives the following result for absorption from gaseous  $H_2SO_4$  in a  $CO_2$  atmospheres at the 13.4 cm wavelength:

$$\alpha_{13} (dB km^{-1}) \approx 9.0 \times 10^9 q(P)^{1/2} T^{-3} (3)$$

where q is the  $H_2SO_4$  number mixing ratio, P is the pressure in atmospheres, and T is the temperature in Kelvins. A similar expression for absorption from

gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere at the 3.6 cm wavelength has also been estimated:

$$\alpha_{3.6} (dB \ km^{-1}) \simeq 4.52 \ x \ 10^{10} q(P)^{0.85} T^{-3}$$
 (4)

Inspection of these results shows several significant findings:

- (1) A new relation for  $H_2SO_4$  vapor pressure has been developed which generally confirms the work of Roedel (1979) and Ayers <u>et al.</u> (1980) in that the vapor pressures inferred are a factor of ten below those predicted by Gmitro and Vermeulen (1964). However, the findings with regard to the temperature dependence of the  $H_2SO_4$  vapor pressure are different enough from previous work so as to significantly affect the saturation abundances and cloud formation altitudes which one would predict for the Venus atmosphere. Clearly, additional measurements in the 350-450 K range are needed.
- (2) While generally confirming the initial laboratory measurements made by Steffes and Eshleman (1982) of  $H_2SO_4$  vapor absorption at a single temperature and pressure, it has been found that the microwave absorption of  $H_2SO_4$  in a  $CO_2$  atmosphere has a <u>significant</u> pressure dependence at both the 13 cm and 3.6 cm wavelengths. This is in sharp contrast to the theoretical calculation presented in Cimino (1982).
- (3) Frequency dependences which vary significantly with pressure have been found for the 3-13 cm wavelength absorption from gaseous  $H_2SO_4$ , ranging from f<sup>1.23</sup> at 1 atm pressure to f<sup>1.7</sup> at 6 atm pressure. This will significantly effect comparisons of radio occultation data with radio astronomical data from Venus.

(4) Simple multiplicative expressions for the absorption from gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere at 13 cm and 3.6 cm wavelengths have been developed which are highly useful in converting microwave absorptivity profiles to  $H_2SO_4$  abundance profiles.

In addition to the measurements at 13 cm and 3.6 cm wavelengths, measurements in the 1 to 3 cm wavelength range are currently being conducted and should yield similar valuable data for interpretation of a wide range of radio astronomical data.

#### V. APPLICATION OF LABORATORY RESULTS TO VENUS OPACITY DATA

A wide range of analytical studies of the Venus atmosphere based on the laboratory data has now begun. These studies cover an area which is so wide that complete documentation would be difficult at this time. Examples of application of the data can be seen in Figures 5 and 6. Figure 5 shows the 13.4 cm wavelength microwave absorption which would arise from a 13.5 ppm abundance of gaseous  $H_2SO_4$  in the middle atmosphere of Venus which condenses to form clouds at 48 km altitude. It can be seen from this figure that gaseous  $H_2SO_4$  is the predominant microwave absorber in the 35 to 48 km altitude range as compared to other absorbing constituents. For this comparison, we use equation (3) to compute the opacity from gaseous  $H_2SO_4$ . For  $SO_2$ ,  $H_2O$ , and  $CO_2$ , measured absorption coefficients from Steffes and Eshleman (1981) and Ho et al. (1966) are used. For liquid  $H_2SO_4$ , we employ opacities from Cimino et al. (1980). It can also be seen from this figure that abundances on the order of 15 to 30 ppm could account for most of the 13 cm absorption measured in a number of radio occultation measurements. In

addition, it should be noted that if the frequency dependences measured in the 3 to 13 cm wavelength range are used to extrapolate to 1.35 cm, the resulting vertical opacity from a 15 to 30 ppm abundance of gaseous  $H_2SO_4$  at altitudes below 48 km would exceed that attributable to constituents other than  $CO_2$  and  $SO_2$  ( $\tau_{other} = 3.6$ , for further discussion, see Steffes and Eshleman, 1982). Thus, it is likely that dissociation of gaseous  $H_2SO_4$  occurs at lower altitudes (below 30 km). Actual limits on gaseous  $H_2SO_4$  abundance, based on opacities inferred from 1.35 cm brightness and temperature measurements must await laboratory measurement of the 1.35 cm absorptivity of gaseous  $H_2SO_4$ .

Figure 6 shows abundance of gaseous sulfuric acid, at several altitudes, obtained by attributing all of the 13 cm opacity measured by the 3 radio occultation experiments shown, to gaseous sulfuric acid. Also shown are curves showing saturation vapor abundance as a function of altitude. Note that the vapor will not condense at a given altitude unless its abundance equals or exceeds the saturation abundance. The saturation vapor abundance obtained using vapor pressure data from Gmitro and Vermeulen (1964) would imply that for the  $H_2SO_4(g)$  abundances shown, no significant cloud formation could occur except at altitudes well above 50 km. This, of course, conflicts with in-situ atmospheric probe findings. (See, for example, Esposito et al., 1983.) The saturation vapor abundance from Ayers et al. (1980) would require cloud formation below the 45 km altitude for the  $H_2SO_4(g)$  abundance shown, which likewise conflicts with in-situ findings. The saturation abundances inferred from our measurements are consistent with cloud formation in the 46 to 48 km range, for the given abundances of gaseous  $H_2SO_4$ , except for a single peak measurement from the Mariner 10 experiment. However, it should be noted that the average Mariner 10 absorptivity measurements were well below that figure (Lipa and Tyler, 1979).

While the correlation between the inferred abundance of gaseous sulfuric acid (inferred by ascribing to it most of the observed 13 cm opacity in the sub-cloud region) and the abundance required so that saturation, and cloud formation, would begin in the 46 to 48 km altitude range gives further support to the hypothesis that observed 13 cm opacity is indeed caused by gaseous sulfuric acid below the clouds, it cannot be said to constitute proof thereof. However, the strong correlation between the shape of the observed 13 cm opacity profiles (some variations below 48 km altitude, but little measurable opacity above 50 km--see, for example, Fjeldbo <u>et al</u>. (1971) or Cimino (1982)), and the opacity profile shown in Figure 5 gives further evidence to support that theory. A similar correlation exists between observed opacity at 3.6 cm (see Cimino, 1982) and that attributed to gaseous sulfuric acid which follows our saturation abundance curve (see Figure 6) above the 48 km altitude.

Thus, abundances of gaseous sulfuric acid below the main Venus cloud layer on the order of 15-30 ppm are indicated from radio occultation 13 cm absorptivity measurements. However, positional and temporal variations in the measured opacity at 13 cm (from radio occultation experiments) and at 3.6 cm (from radio astronomical and radio occultation experiments) indicate as much as a factor of 2 variation in the gaseous  $H_2SO_4$  abundance in the 30-50 km altitude range. While some portion of these apparent variations in atmospheric opacity may be due to interpretive or calibration inaccuracies, they suggest significant atmospheric changes with time and position. Such variabilities as observed by the relatively localized radio occultation measurements at 13.3 cm and 3.6 cm wavelengths, may be due to global circulation (see Kliore <u>et al.</u>, 1984). However, longer term variations of the full

disk brightness temperature at wavelengths shortward of 4 cm may require variations on a global scale, such as might be induced by volcanism. (See, for example, Esposito (1984) or Prinn (1984).) Further study into this issue is being conducted.

#### VI. CONCLUSIONS AND SUGGESTIONS FOR FUTURE WORK

The laboratory measurements made of the microwave absorption and vapor pressure of sulfuric acid vapor under simulated Venus conditions, when applied to the measured opacities and temperature structure for the Venus atmosphere, give new insights into the abundance and structure of sulfuric acid vapor in the Venus atmosphere. Because of instrumental limitations of <u>in-situ</u> probes (see Von Zahn <u>et al.</u>, 1983), this may become the major technique for determining  $H_2SO_4$  abundance profiles in the Venus atmosphere.

Future measurements will allow us to interpret a wider range of microwave absorptivity data, and with higher accuracies. Measurements currently planned or underway include:

- (1) Measurement of absorptivity of gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere (1 to 6 atm total pressure) in the 1 to 3 cm wavelength range.
- (2) Measurement of the temperature dependence of  $H_2SO_4$  microwave opacity.
- (3) Measurement of upper limits for  $H_2SO_4$  vapor pressure in the 350-500 K temperature range, as constrained by system sensitivity.

Completion of these measurements will permit study and interpretation of an extremely wide range of microwave opacity data for the Venus atmosphere, with accompanying results in the area of temporal and spatial constituent abundance variations. Finally, upon completion of these measurements, the system will

be reconfigured so as to allow for microwave absorptivity measurements of several gaseous constituents under simulated conditions for the outer planets. Such measurements can be used to interpret data from Voyager radio occultation experiments, radio astronomical observations of the outer planets, and in the future, radio absorption measurements from the Galileo spacecraft and probe.

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#### FIGURE CAPTIONS

- Figure 1: Block diagram of atmospheric simulator, as configured for measurements of microwave absorption of gaseous  $H_2SO_4$  under venus atmospheric conditions.
- Figure 2: Measured microwave absorptivity (normalized by number mixing ratio) for gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere as a function of pressure at 2.24 GHz (13.4 cm wavelength). Error bars are  $\pm$  1  $\sigma$ . Dashed line represents a best-fit multiplicative expression for absorptivity.
- Figure 3. Measured microwave absorption (normalized by number mixing ratio) for gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere as a function of pressure at 8.42 GHz (3.6 cm wavelength). Error bars are  $\pm$  1  $\sigma$ . Dashed line represents a best-fit multiplicative expression for absorptivity.
- Figure 4. Vapor pressure of gaseous  $H_2SO_4$  above liquid sulfuric acid as a function of the inverse of temperature. The illustrated points are from the laboratory measurements. Vapor pressure expressions (dotted lines) from Ayers <u>et al</u>. (1980) and Gmitro and Vermeulen (1964) are then compared with a best-fit expression for our measurements (solid line). Error bars for temperature and pressure are  $\pm 1 \sigma$ .
- Figure 5. Comparative 13.4 cm opacity of several constituents in the middle atmosphere of Venus, assuming the presence of gaseous  $(g)H_2SO_4$  (13.5 ppm), gaseous  $SO_2$  (150 ppm), gaseous  $H_2O$  (1000 ppm),  $CO_2$  (95%), and liquid ( $\ell$ )  $H_2SO_4$  (100 mg per cubic meter in the 48 to 50 km altitude range). We assume the cloud-related gases to be depleted above the cloud layer (50 km and above).
- Figure 6. Saturation abundances for gaseous sulfuric acid  $(H_2SO_4)$  under conditions for the Venus middle atmosphere. Dashed lines represent saturation abundance data from Ayers <u>et al.</u> (1980) and Gmitro and Vermeulen (1964). Solid line represents saturation abundance data from this work. The illustrated points represent the abundances of gaseous  $H_2SO_4$  required to explain radio occultation results at 13 cm from Mariner 5 (Fjeldbo <u>et al.</u>, 1971), Mariner 10 (Lipa and Tyler, 1979), and Pioneer Venus Orbit 18 (Cimino, 1982), using the laboratory measurements described.



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Fig. 1 (Steffes)



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Fig. 2 (Steffes)





Fig. 4 (Steffes)



Fig. 5 (Steffes)



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Fig. 6 (Steffes)



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Fig. 6 (Steffes)

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#### REPORT

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#### TO THE

#### NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

# ANNUAL STATUS REPORT (Includes Semiannual Status Report #4)

for

GRANT NAGW-533

#### LABORATORY EVALUATION AND APPLICATION OF

#### MICROWAVE ABSORPTION PROPERTIES UNDER SIMULATED

#### CONDITIONS FOR PLANETARY ATMOSPHERES

Paul G. Steffes, Principal Investigator

February 1, 1985 through January 31, 1986

Submitted by

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## PAGE

#### 1. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often lead to significant misinterpretation of available opacity data. Steffes and Eshleman (1981) showed that under environmental conditions corresponding to the middle atmosphere of Venus, the microwave absorption due to atmospheric SO2 was 50 percent greater than that calculated from Van Vleck-Weiskopff theory. Similarly, the opacity from gaseous  $H_2SO_4$  was found to be a factor of 7 greater than theoretically predicted for conditions of the Venus middle atmosphere (Steffes and Eshleman, 1982). The recognition of the need to make such measurements over a range of temperatures and pressures which correspond to the periapsis altitudes of radio occultation experiments, and over a range of frequencies which correspond to both radio occultation experiments and radio astonomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements.

In the initial year of Grant NAGW-533 (i.e., February 1, 1984 through January 31, 1985), this facility was developed, and then operated, in order to evaluate the microwave absorbing properties of gaseous sulfuric acid  $(H_2SO_4)$  at 13.4 and 3.6 cm wavelengths, under Venus atmospheric conditions. The

results were then applied to measurements from Mariner 5, Mariner 10, and Pioneer-Venus Radio Occultation experiments, to determine abundances of gaseous sulfuric acid in the Venus atmosphere, with accuracies exceeding those achieved with <u>in-situ</u> instruments. Measurements of the microwave properties of the vapors accompanying liquid  $H_2SO_4$  also resulted in more accurate estimates of the vapor pressure behavior of sulfuric acid, which are critical for modeling the behavior and structure of the Venus atmosphere. The results of this work are being published in a paper entitled, "Laboratory Measurements of the Microwave Opacity and Vapor Pressure of Sulfuric Acid Vapor under Simulated Conditions for the Middle Atmosphere of Venus," in the journal, <u>Icarus</u> (Steffes, 1985).

During the first six months of the current grant year, our work concentrated on making laboratory measurements of the microwave absorption from gaseous  $H_2SO_4$  at wavelengths from 1 to 22 cm under simulated Venus conditions. Additional measurements of the vapor pressure behavior of sulfuric acid were also made. During the second half of this year, we have applied these results to radio astronomical observations of Venus which have been made in the same wavelength range, in order to better model the structure of  $H_2SO_4$  and  $SO_2$  abundance in the Venus atmosphere, and to resolve temporal variations of their abundances on a planet-wide basis. The results of this effort have been especially rewarding in that the unique frequency and pressure dependences measured for the absorption from gaseous  $H_2SO_4$  in these wavelengths and those obtained in the 1 to 3 cm wavelength range. We describe these effects, and the resulting limitations they place on abundances of gaseous  $H_2SO_4$  and  $SO_2$  in

the Venus atmosphere, in a paper entitled, "Evaluation of the Microwave Spectrum of Venus in the 1.3 to 22 cm Wavelength Range Based on Laboratory Measurements of Constituent Gas Opacities," which is being prepared for submission to Astrophysical Journal. In addition, we have completed the process of redesigning and refitting the laboratory system so as to permit measurements of the microwave absorptivity of ammonia  $(NH_3)$ , methane  $(CH_4)$ , and other potential microwave absorbers under simulated conditions for the outer planets (Jupiter, Saturn, Uranus, and Neptune). In May, a paper entitled, "Laboratory Measurements of Microwave Absorption from Gaseous Atmospheric Constituents under Conditions for the Outer Planets," was presented at the Conference on the Jovian Atmospheres, held at the Goddard Institute for Space Studies in New York, in which feedback from other experimenters was sought in order to optimize the yield from our outer planets simulations. (See Appendix 1.) This feedback helped significantly in our design of the outer planets atmospheric simulator.

Our plans for future work includes, as time and resources permit, further analysis and application of our laboratory results for the microwave absorption from gaseous  $H_2SO_4$  in the Venus atmosphere. Our <u>long term</u> goal would be a detailed analysis of available multi-spectral microwave opacity data from Venus including data from the Pioneer-Venus Radio Occultation experiments and earth-based radio and radar astronomical observations, such as the kinds which have been performed at the NRAO Very Large Array (VLA) and at stations in the Deep Space Network (DSN). This would provide a chance to determine both spatial and temporal variations in the abundances of both  $H_2SO_4$  and  $SO_2$  in the Venus atmosphere. However, our most immediate priority for the next grant year will be completion of the measurement of the microwave (1.3 to 27 GHz)

opacity of H<sub>2</sub>, NH<sub>3</sub>, and CH<sub>4</sub> under simulated conditions for the outer planets. We likewise would hope to be able to pursue a program of analysis and application of these results to microwave data for the outer planets such as Voyager Radio Occultation experiments and earth-based radio astronomical observations.

## II. THE GEORGIA TECH RADIO ASTRONOMY AND PROPAGATION (R.A.P.) FACILITY

The basic configuration of the planetary atmospheres simulator developed at Georgia Tech for use in measurement of the microwave absorptivity of gases under simulated conditions for planetary atmospheres is described at length in the first <u>Annual Status Report</u> for Grant NAGW-533 (February 1, 1984 through January 31, 1985). It is also discussed at length in Steffes (1985). The major new addition made to the laboratory apparatus is a microwave resonator capable of making absorptivity measurements in the 8 to 27 GHz range (1.1 to 3.7 cm wavelengths). This significantly extends the operating range beyond the previous 1.3 to 8.5 GHz (3.6 to 22 cm) operating range. As shown in Figure 1, the system was initially configured so as to be used in measuring microwave absorption (1.3 to 27 GHz) from gaseous  $H_2SO_4$  under simulated conditions for the middle atmosphere of Venus (total pressures from 1 to 6 atm, temperatures from 500 to 575 K).

Adding a resonator capable of operating in the 8 to 27 GHz frequency range was more difficult than expected, especially considering the high temperatures and pressures involved. It was decided to use coaxial cable to interconnect the high frequency resonator to the microwave instrumentation, since hermetically-sealed coaxial feed-through connectors were available which would allow operation to 30 GHz while still maintaining the pressure integrity of the chamber. The type of coaxial cable required for the higher frequency

resonator was different from that used in the 1.3 to 8.5 GHz range, in that a solid metallic jacket is required to prevent signal leakage, as opposed to the metallic braid used in lower frequency cables. Unfortunately, even when hightemperature solder was used, connector failure occurred for these cables at temperatures above 500 K. This was due to the extrusion of the PTFE (Teflon) dielectric material within the cable placing mechanical stresses on the connectors. As a result, a limited production air-dielectric cable was obtained which employs a PTFE spline to space the center conductor from the solid metallic outer conductor. The connectors for this specialized cable have been difficult to obtain since special production runs (and accompanying set-up charges) are usually required. Fortunately, we were able to combine the production of our connectors with those from other customers so as to avoid these charges. The result has been a system capable of successfully measuring the microwave absorption from gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere at wavelengths from 1.2 to 22 cm, at pressures from 1 to 6 atmospheres, and at temperatures from 500 to 570 K. (Lower temperatures can, of course, be achieved but the resulting gaseious  $H_2SO_4$  abundance would be too small so as to provide measurable absorptivity.)

The pressure vessel and its accompanying resonators are designed to be usable over an extremely wide range of temperatures. The extremely high temperatures (up to 600 K) developed for the Venus atmosphere simulations by the oven/temperature chamber (see Figure 1) can be replaced by extremely low temperatures created by a freezer/temperature chamber for simulations of the outer planets, as shown in Figure 2. Such a freezer has been procured for use in such measurements. The unit, a Revco/Rheem ULT-7120D, has an internal volume of 193 liters (which is capable of containing the pressure vessel with

resonators), and is capable of maintaining temperatures as low as 150 K. Since access to the freezer is through the top of the unit, a special lifting frame and pulley system has been constructed which allows easy movement of the pressure vessel and accompanying resonators in and out of the freezer. Another critical issue has been the behavior of sealing structures such as gaskets or 0-rings. We have found that at low temperatures, materials such as teflon or viton become too brittle to be used as gaskets. Thus we have tested alternate materials for use in this application. The result is a system capable of measuring microwave absorption from 1.3 to 27 GHz (1.1 to 22 cm wavelength) at pressures to 8 atmospheres and at temperatures as low as 150 K. The minimal measurable absorption absorptivity, or sensitivity, for this system when operated at 170 K is shown in Figure 3, as a function of frequency.

Another more critical limitation on how low the temperatures can be taken (while still being able to measure microwave absorption) is the saturation vapor pressure of the gas being tested. As shown in Table 1, the lowest temperature for which sufficient quantities of gaseous  $NH_3$  would still exist so that the microwave absorption in an  $H_2$  atmosphere would be measurable would be approximately 155 K.

A final critical issue which greatly affects the use of the simulator in outer planets simulations is the use of hydrogen  $(H_2)$  at pressures reaching 8 atmospheres. In previous simulations, small leakages from the pressure vessel presented little or no danger to the experimenters. The use of hydrogen will require a special set of procedures and a special ventilation system to be used. Initial pressure tests of the system, as configured for outer planets simulation, are currently being conducted with nitrogen  $(N_2)$ . Not

only is this a safer way to test the system, but it also allows measurement of the collisional microwave absorption from  $N_2$ , which may be the source of the 3.6 cm (8.4 GHz) opacity detected by Voyager 1 radio occultation studies of the Titan atmosphere. (See Lindal <u>et al.</u>, 1985.) It is noteworthy that funding for laboratory safety equipment needed in making such measurements has been provided by the Georgia Institute of Technology in support of planetary atmospheres research at Georgia Tech.

## III. RESULTS OF LABORATORY MEASUREMENTS AND THEIR APPLICATION

Our goal for the first six months of the current grant year was to complete laboratory measurements of the microwave absorption from gaseous  $H_2SO_4$  in the 1.2 to 22 cm wavelength range, under simulated conditions for the middle atmosphere of Venus, and to obtain additional measurements of the vapor pressure behavior of gaseous  $H_2SO_4$  above liquid sulfuric acid. The additional vapor pressure measurements made have been consistent with the vapor pressure expression derived from earlier data. Our measured results for the microwave absorption from gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere at 575 K are shown in Figure 4. The data points presented are for an  $H_2SO_4$  mixing ratio of 0.4 percent at total pressures up to 6 atmospheres. Best-fit curves for the absorption spectra from 1.3 GHz (22.5 cm) to 25 GHz (1.2 cm) at pressures of 6 atm and 1 atm are also shown. Inspection of these results reveals three major results.

The first major result has been the extremely low absorptivity observed in the 1.2 to 1.8 cm wavelength range. For example, measurements made at 21.63 GHz (1.38 cm wavelength) show an opacity for a .36% mixture of gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere (with a total pressure of 6 atmospheres and

temperature of 570 K) of less than 9 dB/km. This is far below the opacity predicted by using previous measurements at 2.2 GHz and 8.4 GHz and assuming a simple  $f^2$  dependence such as exhibited by  $SO_2$  and  $CO_2$ . In fact, this even implies that using the measured frequency dependences for the microwave absorption from  $H_2SO_4$  in a  $CO_2$  atmosphere in the 2.2 to 8.4 GHz range to predict absorption at 21.6 GHz (1.38 cm wavelength) would result in overstating the absorption at 1.38 cm by at least a factor of 2.

Such behavior implies that the contribution of gaseous  $H_2SO_4$  to the overall non-CO2 opacity inferred from radio astronomical observations at the 1.35 cm is minimal. Such a result is not surprising in light of the calculations done by Janssen and Klein (1981) which attribute nearly all of the non-CO<sub>2</sub> opacity at 1.35 cm in the Venus atmosphere to SO<sub>2</sub>. This behavior is likewise consistent with the calculated  $H_2SO_4$  resonance frequencies computed by Poynter. (R. Poynter, J.P.L., personal communication. Note that the results of the resonance calculations are shown in Cimino, 1982.) These calculations are based on rotational constants computed using measurements of H<sub>2</sub>SO<sub>4</sub> resonances in the 60 to 120 GHz range by Kuczkowski et al. (1981). It is significant that there is a notable absence of  $H_2SO_4$  resonances in the 15 to 30 GHz frequency range, which would be consistent with our measurements. A computation of the microwave spectrum from H<sub>2</sub>SO<sub>4</sub>, made by Allen, which is presented in Cimino (1982), showed significantly more absorption from  $H_2SO_4$ in this same frequency range than we have actually measured. This can be explained by the fact that a pressure-broadened linewidth parameter of 7.2 MHz was assumed to  $H_2SO_4$  in a  $CO_2$  atmosphere. It appears that using a smaller broadening parameter will result in a spectrum which will be consistent with our measurements.

The second major result has been the discovery of a peak in the absorptivity from gaseous  $H_2SO_4$  at a wavelength of approximately 2.2 cm, even at pressures as high as 6 atm. This does a lot to explain why observations of the Venus brightness temperature at 2 cm (495 K--from Pollack and Morrison, 1970), which were initially felt to be inconsistent with measurements at 1.35 cm (474 K--from Muhleman <u>et al.</u>, 1979), are indeed <u>consistent</u>, because there is at least one absorber whose opacity <u>decreases</u> with increasing frequency, in that wavelength range. In fact, we have developed a model for the microwave emission spectrum of Venus, using our laboratory results for gaseous  $H_2SO_4$ , and previous results for  $SO_2$  and  $CO_2$  which provides a better fit to the ensemble of Venus microwave observations than any model yet proposed. We are currently preparing a paper to be submitted to <u>The</u> Astrophysical Journal, which will present this model.

The third major result has been the low absorptivity measured at 22.3 cm. This is important since the well known problem of the falloff of brightness temperatures at wavelengths longward of 20 cm (see, for example, Muhleman <u>et al.</u>, 1979) is still unsolved, and might have been explained by a large opacity from a gaseous constituent. Our negative result indicates that no atmosphere constituent can be responsible for this effect.

Thus, as can be seen in Figure 5, our laboratory measurements indicate that while radio occultation measurements of 13 and 3.6 cm, and radio astronomical measurements at wavelengths longer than 2 cm, measure absorption which is predominated by gaseous  $H_2SO_4$ ; opacity measured by radio astronomical observations in the 1 to 2 cm wavelength range is predominated by  $SO_2$  at higher altitudes, and by  $CO_2$  at lower altitudes.

## IV. PUBLICATIONS AND INTERACTION WITH OTHER INVESTIGATORS

At the beginning of the current grant year, a paper was completed and revised for publication in Icarus, describing results and applications of experiments performed during the first year of the grant (P. G. Steffes, 1985, "Laboratory Measurements of the Microwave Opacity and Vapor Pressure of Sulfuric Acid Vapor under Simulated Conditions for the Middle Atmosphere of Venus.") In May, a paper was presented at the Conference on Jovian Atmospheres, at the Goddard Institute for Space Studies (New York) entitled, "Laboratory Measurements of Microwave Absorption from Gaseous Atmospheric Constituents under Simulated Conditions for the Outer Planets." (See Abstract, Appendix 1.) This paper described our plans and capabilities for simulating outer planets atmospheres and measuring microwave properties of those atmospheres. Much positive feedback was received with regard to the need for such measurements, and discussions were held with I. de Pater of UC/Berkeley and A. Kliore of JPL regarding application of such data to radio astronomical observations, and Galileo probe measurements, respectively. Contact with both is expected to continue as the measurements are made. Contacts have also been maintained with groups at the Stanford Center for Radar Astronomy (V. Eshleman, director), and JPL (Drs. Michael J. Klein and Samuel Gulkis). We have also had discussions with members of the Galileo Cloud Particle Size Spectrometer Team with regards to allowing the use of our simulator for testing of certain devices.

In August, an Invited Paper was presented at the International Association of Meteorology and Atmospheric Physics (IAMAP) Assembly (August 12, 1985) entitled, "Microwave Absorption from Cloud-Related Gases in Planetary

Atmospheres," which summarizes our work over the past 2 years and its applications. In addition, a paper was presented at the October meeting of the Division of Planetary Sciences of the American Astronomical Society (DPS/AAS) on the complete microwave spectrum of  $H_2SO_4$  in a  $CO_2$  atmosphere (from 1.2 to 22 cm) and its application to understanding the microwave emission spectrum from Venus (abstract attached as Appendix 3). The response from this presentation was extremely strong, from both the radar community as well as from radio astronomers. Dr. T. W. Thompson of JPL expressed a strong interest in the work and results and suggested cooperation on a Pioneer-Venus related project involving analysis of radar data. Dr. James B. Garvin of NASA-Goddard expressed strong interest in our results with regard to his interaction with the Soviet Vega mission to Venus. He was especially interested in our results for gaseous  $H_2SO_4$  abundances, since gaseous  $H_2SO_4$  was detected by the Vega probes. He was also interested in our abilities to make microwave measurements under simulated Venus conditions to support the Venus Radar Mapper program and similar Soviet synthetic aperture radar programs.

It should be noted that travel funds for all of the conferences and meetings attended were provided by the State of Georgia and the Georgia Institute of Technology, in support of planetary atmospheres research at Georgia Tech. We have also maintained contact with our congressional delegation, keeping them aware of our work, and the need for continued support to the solar system exploration program. (See Appendix 2.)

#### V. CONCLUSION

During the current grant year (February 1, 1985 through January 31, 1986) we have completed measurements of the microwave absorption from gaseous  $H_2SO_4$ in the 1.2 to 22.3 cm wavelength range, as well as having applied results from earlier measurements at 3.6 and 13.4 cm wavelengths to microwave absorptivity data from radio occultation measurements at those wavelengths, in order to derive abundance profiles for gaseous  $H_2SO_4$ ,  $SO_2$ , and  $CO_2$ .

Currently, we are completing the outer planets simulator construction, and beginning some preliminary measurements of the microwave absorption from nitrogen  $(N_2)$  to simulate Titan. In the next grant year we hope to measure absorption from ammonia  $(NH_3)$  and methane  $(CH_4)$  in a hydrogen atmosphere  $(H_2)$ in order to simulate Jovian atmospheres. We will likewise continue work on the application of our newly derived absorptivity spectra to a wide range of radio astronomical data.

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# VII. KEY FIGURES

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Figure 1: Blockdiagram of updated Georgia Tech Planetary Atmospheres Simulator, as configured for measurements of the microwave properties of gaseous  $H_2SO_4$  under simulated conditions for the Venus atmosphere over the 1 to 20 cm wavelength range.



gure 2: Block diagram of Georgia Tech Planetary Atmospheres Simulator, as configured for measurements of microwave refraction and absorption of gases under simulated conditions for the outer planets.







## TABLE I

Constituent Gas	Abundance of Gas in H <sub>2</sub> atmosphere (6 atm total pressure) required so as to measure microwave absorption at lowest Temperature	Partial Pressure of Gas (in atm.) required so as to exhibit detectable absorption in H <sub>2</sub> atmosphere. (P <sub>H2</sub> = 6 atm.)	Lowest Possible Temperature for which required abundance can be achieved (K)
NH <sub>3</sub>	60 ppm	$3.6 \times 10^{-4}$	155
H <sub>2</sub> -H <sub>2</sub> and H <sub>2</sub> -He* (collisional)	80% H <sub>2</sub> : 20% He	6	20
CH <sub>4</sub> **	33%	2	120
CO	Not Detectable in the 1 to	20 cm range by this syst	em

## MINIMUM GASEOUS ABUNDANCES NECESSARY SO AS TO BE MEASURABLE BY THE SYSTEM

\* Will provide baseline reference for measurements of other constituents

\*\* The theoretically predicted absorption for methane under these conditions is approximately 80% below the minimum detectable absorption for the system. However, it was felt that this is the largest mixing ratio for which hydrogen broadening would still predominate.

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VIII. APPENDICES

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APPENDIX I : Abstract of paper presented at the Conference on the Jovian Atmospheres (Goddard Institute for Space Studies, New York City, May 1985.)

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## Laboratory Measurements of Microwave Absorption from Gaseous Atmospheric Constituents under Conditions for the Outer Planets

#### P. Steffes (Georgia Institute of Technology)

Quite often, the interpretive work on the microwave and millimeterwave absorption profiles, which are inferred from radio occultation measurements or radio astronomical observations of the outer planets, employs theoretically-derived absorption coefficients to account for contributions to the observed opacity from gaseous constituents. Variations of the actual absorption coefficients from those which are theoretically-derived, especially under the environmental conditions characteristic of the outer planets, can result in significant errors in the inferred abundances of the absorbing constituents. The recognition of the need to make laboratory measurements of the absorptivity of gases such as NH3, CH4, and  $H_2O$  in a predominantly  $H_2$  atmosphere, under temperature and pressure conditions simulating the outer planets' atmospheres, and at wavelengths corresponding to both radio occultation and radio astronomical observations, has led to the development of a facility capable of making such measurements at Georgia Tech. We describe the laboratory measurement system, the measurement techniques, and the proposed experimental regimen for Summer 1985; with the goal of obtaining feedback from interested investigators on the relative priorities of the various proposed measurements to be made on specific constituents at specific wavelengths.

MARK O. HATFIELD, OREGON, CHAIRMAN

APPENDIX II:

VENS. ALASKA P. WEICKER, JA., CONNECTICUT L. MCCLURE, IDAHO XALT. NEVADA IRN. UTAH JCHRAN, MISSISSIPH NOREWS. NORTH DAKOTA BDNOR, SOUTH DAKOTA BDORGI, NEW HAMPSHIRE PECTER, PENNSYLVANIA DOMENICI, NEW MEXICO L. VETTU VENNEOU

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J. KEITH KENNEDY, STAFF DIRECTOR FRANCIS J. SULLIVAN, MINORITY STAFF DIRECTOR United States Senate

COMMITTEE ON APPROPRIATIONS WASHINGTON, DC 20510

May 30, 1985

Professor Paul G. Steffes Georgia Institute of Technology School of Electrical Engineering Atlanta, Georgia 30332

Dear Professor Steffes:

Thank you for contacting me concerning NASA.

I believe that the exploration of space and the utilization of knowledge gained from such efforts would prove to be of inestimable value to all people. It is important that we proceed with the development of space exploration technology and I will support such efforts in the Congress.

Again, thank you for contacting me.

Sincerely,

Mack Mattingly

MM/jbh

<u>Constraints on Constituent Abundances in the Venus Atmosphere</u> from the Microwave Emission Spectrum in the 1 to 20 cm Wavelength Range

P. G. Steffes, D. H. Watson (Georgia Institute of Technology)

We have recently completed laboratory measurements of the microwave opacity of gaseous  $H_2SO_4$  under simulated conditions for the Venus atmosphere in the 1 to 20 cm wavelength range. These measurements, when combined with previous laboratory measurements of the absorption properties of  $CO_2$  and  $SO_2$  in this wavelength range, are used to develop new limits on constituent abundances, based on the observed microwave emission from Venus. We also discuss limits on the long term variations of  $SO_2$  and gaseous  $H_2SO_4$  abundances in the Venus atmosphere, based on the results of the laboratory measurements, and on the reported variations of the Venus microwave emission spectrum.

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#### SUBMIT ABSTRACT TO:

Darrell F. Strobel/DPS Department of Earth and Planetary Sciences Johns Hopkins University Baltimore, MD 21218

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American Astronomical Society

Date Submitted \_\_\_\_

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GEORGIA INSTITUTE OF TECHNOLOGY SCHOOL OF ELECTRICAL ENGINEERING ATLANTA, GEORGIA 30332 July 10, 1986

EPHONE: (404) 694. 3128

Dr. Kenneth Fox Discipline Scientist Solar System Exploration Division Code EL National Aeronautics and Space Administration Washington, DC 20546

Dear Dr. Fox:

As specified in the NASA Provisions for Research Grants and Cooperative Agreements, I am enclosing three (3) copies of <u>Semiannual Status Report #5 for</u> <u>Grant NAGW-533</u> (Laboratory Evaluation and Application of Microwave Absorption Properties under Simulated Conditions for Planetary Atmospheres). As required, I am also forwarding two (2) copies of the report to the NASA Scientific and Technical Information Facility.

While this report, like others preceeding it, begins with the sentence, "Radio absorptivity data ... can be used to infer abundances of microwave absorbing atmospheric constituents ... as long as reliable information regarding the microwave absorbing properties of potential constituents is available", you'll find that it contains much new information regarding both the development of our outer planets atmospheric simulator and results of our Titan atmosphere simulations.

Our current activity involving the measurement of the microwave properties of ammonia (NH<sub>3</sub>) in a hydrogen/helium (H<sub>2</sub>/He) atmosphere under Jovian conditions is going very well, and we hope to have the results ready for presentation at the November DPS meeting.

Given the many budgetary uncertainties, along with the cancellation of the Centaur program creating many difficulties for the Galileo program, I'm sure things at SSED are running between "crazy and insane." If I, as a Planetary Atmospheres Investigator, or as a MOWG member, can provide any information or assistance, to make things easier for you, please do not hesitate to ask.

Sincerely,

Paul G. Steffes Assistant Professor

PGS/mjc

Enclosures



GEORGIA INSTITUTE OF TECHNOLOGY SCHOOL OF ELECTRICAL ENGINEERING ATLANTA, GEORGIA 30332 July 10, 1986

PHONE: (404) 894- 3128

NASA Scientific and Technical Information Facility P. O. Box 8757 Baltimore/Washington International Airport, MD 21240

Ladies and Gentlemen:

As specified in the NASA Provisions for Research Grants and Cooperative Agreements, I am enclosing two (2) copies of <u>Semiannual Report #5 for Grant</u> <u>NAGW-533</u> (Laboratory Evaluation and Application of Microwave Absorption Properties under Simulated Conditions for Planetary Atmospheres). I am also forwarding three (3) copies of the report to the NASA technical officers for this grant (K. Fox and H. C. Brinton, Code EL).

Sincerely,

Paul G. Steffes Assistant Professor

PGS/mjc

Enclosures

#### REPORT

#### TO THE

### NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

#### SEMIANNUAL STATUS REPORT #5

for

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### GRANT NAGW-533

#### LABORATORY EVALUATION AND APPLICATION OF

## MICROWAVE ABSORPTION PROPERTIES UNDER SIMULATED

#### CONDITIONS FOR PLANETARY ATMOSPHERES

Paul G. Steffes, Principal Investigator

February 1, 1986 through July 31, 1986

Submitted by

Professor Paul G. Steffes School of Electrical Engineering Georgia Institute of Technology Atlanta, Georgia 30332-0250 (404) 894-3128

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## I. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often lead to significant misinterpretation of available opacity data. Steffes and Eshleman (1981) showed that under environmental conditions corresponding to the middle atmosphere of Venus, the microwave absorption due to atmospheric SO2 was 50 percent greater than that calculated from Van Vleck-Weiskopff theory. Similarly, results obtained for the microwave opacity from gaseous H2SO4 under simulated Venus conditions, during the first two years of Grant NAGW-533 (February 1, 1984 through January 31, 1986), showed that not only was the opacity from  $H_2SO_4$  much greater than theoretically predicted, but that its frequency (wavelength) dependence was far different than that theoretically predicted (Steffes, 1985 and Steffes, 1986a). The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by radio occultation experiments, and over a range of frequencies which correspond to both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements.

In the first two years of Grant NAGW-533 (i.e., February 1, 1984 through January 31, 1986), this facility was developed, and then operated, in order to evaluate the microwave absorbing properties of gaseous sulfuric acid  $(H_2SO_4)$ under Venus atmospheric conditions. The initial results, obtained at 13.4 cm and 3.6 cm wavelengths, were applied to measurements from Mariner 5, Mariner 10, and Pioneer-Venus Radio Occultation experiments, to determine abundances of gaseous sulfuric acid in the Venus atmosphere, with accuracies exceeding those achieved with in-situ instruments (Steffes, 1985). Measurements of the microwave properties of the vapors accompanying liquid  $H_2SO_4$  also resulted in more accurate estimates of the vapor pressure behavior of sulfuric acid, which are critical for modeling the behavior and structure of the Venus atmosphere. Later efforts concentrated on making laboratory measurements of the microwave absorption from gaseous  $H_2SO_4$  at wavelengths from 1.2 to 22 cm under simulated Venus conditions. Additional measurements of the vapor pressure behavior of sulfuric acid were also made. We applied these results to radio astronomical observations of Venus which have been made in the same wavelength range, in order to better model the structure of  $H_2SO_4$  and  $SO_2$  abundance in the Venus atmosphere, and to resolve temporal variations of their abundances on a planet-wide basis. The results of this effort have been especially rewarding in that the unique frequency and pressure dependences measured for the absorption from gaseous  $H_2SO_4$  in these wavelength ranges has finally explained what were thought to be inconsistencies between measurements of the absorption in the Venus atmosphere at 13.3 and 3.6 cm wavelengths and those obtained in the 1 to 3 cm wavelength range. We describe these effects, and the resulting limitations they place on abundances of gaseous  $H_2SO_4$  and  $SO_2$  in the Venus atmosphere, in a paper entitled, "Evaluation of the Microwave Spectrum of

Venus in the 1.2 to 22 Centimeter Wavelength Range Based on Laboratory Measurements of Constituent Gas Opacities," which was submitted in February and has been accepted for publication in <u>The Astrophysical Journal</u> (v. 310, November 1, 1986).

Additional activities during the first half of the current grant year (February 1, 1986 through July 31, 1986) involving application of the laboratory measurements of simulated Venus atmospheres have included assistance to the <u>Magellan</u> program office (J.P.L.) in characterizing effects of the atmosphere on planned microwave radar and radiometric experiments originally planned to characterize surface and geologic parameters. The laboratory measurements have also suggested that a substantial dip in the Venus microwave emission, related to the abundance of gaseous sulfuric acid, should exist near the 2.25 cm wavelength. Since no observations of the Venus emission at this wavelength have ever been published, we hope to use the 140-foot NRAO telescope to not only confirm the presence of the predicted dip, but to use such a dip to determine a planet-wide average for sulfuric acid vapor abundance below the main cloud layer.

The highest priority activity for this grant year has been laboratory measurements of the microwave properties of the simulated atmospheres of the outer planets and their satellites. As described in the most recent <u>Annual</u> <u>Status Report</u> for Grant NAGW-533 (February 1, 1985 through January 31, 1986), our planetary atmospheres simulator underwent a major redesign late last year to permit measurements of the microwave properties of simulated Jovian atmospheres. Further developmental work has been necessary in order to minimize the safety risks involved with high pressure simulations using hydrogen gas at very low temperatures, as required for the outer planets. A

more complete description of this work is given in Section II of this report. However, since many of the low temperature, high pressure tests were initially conducted using the nonexplosive gas nitrogen  $(N_2)$ , which is the primary constituent of the atmosphere of the Saturnian satellite Titan, useful data for microwave refractivity and absorptivity of nitrogen atmospheres under simulated Titan conditions has been obtained at frequencies from 2.2 GHz to 21.7 GHz (1.3 cm to 13.7 cm). Such data is useful in interpretation of both Voyager 1 radio occultation measurements of the Titan atmosphere, as well as radio astronomical observations of Titan.

During the second half of the current grant year (August 1, 1986 through January 31, 1987), we hope to be able to complete measurements of the microwave absorptivity and refractivity of both hydrogen/helium ( $H_2$ /He) atmospheres and of gaseous ammonia ( $NH_3$ ) in an  $H_2$ /He atmosphere under simulated conditions for the outer planets at frequencies from 1.3 GHz to 27 GHz (wavelengths from 1.1 cm to 22 cm). These laboratory results will directly apply to measurements of atmospheric microwave opacity at Jupiter, Saturn, and Uranus made by the Voyager spacecraft, as well as to radio astronomical observations of the outer planets.

Beyond the current grant year, our goals are to continue such laboratory measurements of the microwave absorption and refraction from other potential microwave absorbers contained in the outer planets' atmospheres, including methane ( $CH_4$ ) and phosphine ( $PH_3$ ). We likewise would hope to be able to pursue a program of analysis and application of these results to microwave data for the outer planets such as Voyager Radio Occultation experiments and earth-based radio astronomical observations.

Of equal importance, we feel, would be the further analysis and application of our laboratory results for the microwave absorption from gaseous  $H_2SO_4$  in the Venus atmosphere. Our <u>long term</u> goal would be a detailed analysis of available multi-spectral microwave opacity data from Venus including data from the Pioneer-Venus Radio Occultation experiments and earth-based radio and radar astronomical observations, such as the kinds which have been performed at the NRAO Very Large Array (VLA) and at stations in the Deep Space Network (DSN). The new measurements of Venus microwave emission at 2.25 cm and 1.9 cm made with the NRAO 140-foot telescope will be an especially important contribution to this data set. This would provide a chance to determine both spatial and temporal variations in the abundances of both  $H_2SO_4$  and  $SO_2$  in the Venus atmosphere.

## II. THE GEORGIA TECH RADIO ASTRONOMY AND PROPAGATION (R.A.P.) FACILITY

The basic configuration of the planetary atmospheres simulator developed at Georgia Tech for use in measurement of the microwave absorptivity of gases under simulated conditions for planetary atmospheres is described at length in the first two <u>Annual Status Report(s)</u> for Grant NAGW-533 (February 1, 1984 through January 31, 1985 and February 1, 1985 through January 31, 1986). It is also discussed at length in Steffes (1985 and 1986b). The updated simulator system, shown in Figure 1, is currently configured for simulations of the outer planets. Measurements of the microwave opacity and refractivity of test gas mixtures can be performed at frequencies from 1.3 GHz to 27 GHz (wavelengths from 1.2 cm to 22 cm). While the pressure chamber itself is capable of containing pressures up to 10 atmospheres, and the temperature chamber is capable of achieving temperatures as low as 150 K, it has been

found that the combination of high pressures and low temperatures can create a substantial problem with sealing the pressure chamber. In previous simulations, small leakages from the pressure vessel presented little or no danger to the experimenters. The use of gaseous hydrogen  $(H_2)$  in the outer planets simulations has required the development of new procedures and equipment for conducting such simulations. Such precautions include the addition of a hydrogen leakage sensor which is placed inside the temperature chamber immediately outside the pressure vessel (see Figure 1). This sensor can detect the potentially dangerous build-up of hydrogen gas within the freezer unit. A ventilation pump has also been provided which, if needed, can be used to draw any escaping hydrogen gas out of the freezer compartment. Additional precautions have included the construction of ramps which allow all of the equipment to be moved out-of-doors to a concrete slab immediately adjacent to the laboratory. All experiments which employ gaseous hydrogen can thus be conducted out-of-doors in order to avoid any build-up of hydrogen gas within the laboratory. A covered outdoor storage area for hydrogen and helium gases has also been constructed in order to allow safe storage of these gases, and to expedite the out-of-doors experiments. It is noteworthy that funding for the hydrogen leakage sensor, for construction of the outdoor gas storage and experimental areas, and for special tools required for sealing the pressure vessel (over \$2,000) has been provided by the Georgia Institute of Technology, in support of planetary atmospheres research at Georgia Tech. In addition, two vacuum sensors have been added to the system. These sensors not only allow accurate determination of pressure vessel evacuation, but they can also be used for accurately determining the abundance of test gases, which are typically very small at low temperatures, due to low saturation vapor pressures.

Initial testing of the new simulator configuration for the outer planets was conducted using nitrogen  $(N_2)$  gas both because of its relative safety as compared with hydrogen, and because measurements of both the refractivity and absorptivity of nitrogen under simulated conditions for the Saturnian satellite Titan were considered important for interpreting results of the Voyager 1 radio occultation studies of the Titan atmosphere (Lindal <u>et al.</u>, 1983 and 1985).

The first tests of the system showed that the pressure vessel could easily hold pressures of 7 atmospheres at room temperature with minimal leakage. However, when the system was cooled to 150 K, significant leakage occurred. A quick review of pressure vessel construction can shed some light on this problem: The pressure vessel is essentially a 13-inch diameter stainless steel cylinder (0.375-inch wall thickness) with a 0.375-inch thickness stainless steel plate welded to the bottom of the cylinder. At the top of the cylinder is a 0.5-inch thick cylindrical flange which is welded to the flange using 16 stainless steel bolts, along with a pressure sealant 0-ring, and sealant putty which is placed outside the 0-ring. (See Figure 2.) All electrical connections are made via hermetically sealed feed-through connectors mounted on the top plate.

Two causes for the increased leakage rate at extremely low temperatures have been found. The first was simply due to metal contraction. When the pressure vessel was cooled to 150 K, it was found that most of the fastener bolts had loosened due to contraction of the stainless steel. Thus, removal of the cooled pressure vessel from the freezer was necessary so that the bolts could be tightened while it was still very cold, and it was then immediately

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replaced inside the freezer. The special lifting frame and pulley system which was designed for easily moving the pressure vessel in and out of the freezer has been especially useful for this task. However, even after retightening of the fastener bolts on the cold pressure vessel, substantial leakage still occurred.

The cause of the continued leakage was found to be the sealing materials used in the pressure vessel. As shown in Figure 2, the seal used when placing the top plate on the pressure vessel consists of a 0.385-inch cross-section O-ring accompanied by a ring of putty-type sealing material. As was the case in the recent Space Shuttle accident, seals which normally function well at room temperature can malfunction when placed under extremely cold conditions. This is due to increasing brittleness of the O-ring and putty seals, accompanied by a higher probability of cracks developing due to the brittleness. Initially, as suggested by several seal material vendors, we used an O-ring made from vulcanized rubber and RTV-silicone putty. The relatively poor performance of this seal caused us to experiment with several other types of sealing materials. We found that nearly all materials typically used in such seals became brittle at 150 K. However, we found that O-rings fabricated from the material Viton were less likely to develop cracks at this temperature. It was also found that RTV-silicone became extremely brittle at 150 K, and was very susceptible to cracking. As a result, we now use a more fibrous silicone composite material for the sealing putty.

The overall result has been a pressure vessel capable of maintaining 6 atmospheres of pressure at a temperature of 170 K, with an acceptably small leak ratio. While the resulting range of pressures and temperatures which can be tested are not quite as large as originally hoped, they do represent the

pressure-temperature ranges over which most of the microwave opacity in the Jupiter atmosphere has been observed, and thus will be very useful in interpretation of microwave opacity data from Voyager I and II radio occultation experiments, as well as from earth based radio astronomical observations, and opacity measurements to be made using the Galileo probe.

#### III. RESULTS OF LABORATORY MEASUREMENTS AND THEIR APPLICATION

Measurements made of the microwave refraction and absorption from Nitrogen  $(N_2)$  under simulated conditions for Titan are shown in Table I. While no absorption was measured at any frequency from 2.2 GHz to 21.7 GHz, upper limits for the opacity from  $N_2$  can be set. This is especially important since  $N_2$  may very well be the source of the 3.6 cm (8.4 GHz) opacity detected by Voyager I radio occultation studies of the Titan atmosphere (Lindal <u>et al.</u>, 1985). Also shown in Table I is the refractivity, N, and the refractivity normalized by molecule number density. Since the normalized refractivity is used directly to determine atmospheric pressure from measurements of the atmospheric refractivity at a given altitude, an accurate expression for normalized refractivity at the temperature of that altitude.

When inverting the Voyager I refractivity data obtained at 2.3 GHz at Titan, in order to develop a temperature-pressure profile for Titan's atmosphere, Lindal <u>et al.</u> (1983) assumed an exponential atmosphere with surface temperature 94 K, and a constant value for the density normalized "efractivity of gaseous N<sub>2</sub> (1.093  $\pm$  0.0004 N-units/molecule/cm<sup>3</sup>). This value or normalized refractivity was obtained by Essen and Froome (1951) based on a ingle measurement of the refractivity of nitrogen under standard laboratory onditions at a single frequency (24 GHz). This value is included, for

comparison, in Table I. We have measured the refractivity of nitrogen over a wide range of temperatures (down to 156 K) and over a wide range of frequencies (2.2 GHz to 21.7 GHz) in order to determine whether any temperature or frequency variations of the density-normalized refractivity existed, and to estimate their effects on the interpretation of the refractivity data. It appears that the refractivity of  $N_2$ , at a frequency of 2.2 GHz and at a temperature of 156 K, is slightly lower than the value reported at standard temperature and pressure at a frequency of 24 GHz by Essen and Froome (1951). However, the difference is at the limit of resolution of the present experiment, so that the statistical significance is difficult to determine. These results suggest that the surface atmospheric pressure at Titan may be as much as 10% greater than the 1496  $\pm$  20 mbar pressure given by Lindal <u>et al.</u> (1983).

#### IV. PUBLICATIONS AND INTERACTION WITH OTHER INVESTIGATORS

At the beginning of the current grant year, a paper was completed and accepted for publication in The Astrophysical Journal, describing results and applications of experiments performed during the second year of Grant NAGW-533 (Steffes, 1986a). This paper is described at length in Section I of this report. In May 1985, a paper was presented at the Conference on Jovian Atmospheres, at the Goddard Institute for Space Studies (New York) entitled, \*Laboratory Measurements of Microwave Absorption from Gaseous Atmospheric Constituents under Simulated Conditions for the Outer Planets." This paper described our plans and capabilities for simulating outer planets atmospheres and measuring microwave properties of those atmospheres. At the beginning of this year, a revised manuscript was completed and submitted for inclusion in a NASA Conference Proceedings for this conference (Steffes, 1986b). More

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informal contacts have been maintained with groups at the California Institute of Technology (Dr. Duane O. Muhleman), at the Stanford Center for Radar Astronomy (V. Eshleman, director), and at JPL (Drs. Michael J. Klein, Michael Janssen, A. J. Kliore, and Samuel Gulkis). We have also had discussions with members of the Magellan Program Office at JPL with regards to using results from our laboratory measurements of simulated Venus atmospheres in planning mission experiments.

Another source of close interaction with other planetary atmospheres principal investigators has been Dr. Steffes' membership in the Planetary Atmospheres Management and Operations Working Group (PAMOWG). Travel support for attendance at PAMOWG meetings has been provided by Georgia Tech. Travel to Paris, France in November is also being planned so as to allow presentation of our research results from this current grant year at both the 18th Annual Meeting of the Division of Planetary Sciences of the American Astronomical Society and at an accompanying meeting, entitled "Laboratory Measurements for Planetary Science." Support for this travel from the Georgia Tech Foundation, Inc. has been solicited.

Additional publications which describe our research activities in the Planetary Atmospheres to the public have also been released. The first, entitled "Probing Venus," was a two-page color article describing the results of our work simulating the microwave properties of the Venus atmosphere, and ppeared in the publication <u>Research Horizons</u>, published by the Georgia nstitute of Technology (Winter 1986, Volume 3, Number 4, pp. 8-9). It is :tached as Appendix 1. As Appendix 2, we have attached an article which peared in the Tuesday, June 10 edition of <u>The Atlanta Constitution</u> (pg. 34a) d in the Tuesday, June 10 edition of <u>The Atlanta Journal</u> (pg. 22a), entitled

"Tech Making Down-to-Earth Studies of Outer Space." This article likewise appeals to a large public audience. As Appendix 3, we have attached a short article, entitled "Remote Sensing Reveals Much About Planets," which appeared in the <u>Georgia Tech Whistle</u> (Volume 12, Number 18, May 26, 1986), a newspaper for Georgia Tech employees.

#### V. CONCLUSION

During the first half of the current grant year (February 1, 1986 through July 31, 1986), we have completed construction of the outer planets simulator and conducted measurements of the microwave absorption and refraction from nitrogen  $(N_2)$  under simulated Titan conditions. We have also applied the results of these and previous laboratory measurements to a wide range of microwave opacity measurements, in order to derive constituent densities and distributions in planetary atmospheres such as Venus. In the second half of this grant year, we hope to measure microwave absorption from ammonia  $(NH_3)$  in a hydrogen/helium atmosphere  $(H_2/He)$  under simulated Jovian conditions. We will likewise continue work on the application of our newly derived absorptivity spectra to a wide range of spacecraft and radio astronomical data.

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VII. KEY FIGURES

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#### TABLE I

## LABORATORY MEASUREMENTS OF NITROGEN (N2) REFRACTIVITY AND ABSORPTIVITY

Frequency, GHz	Conditions	Refractivity N = 10 <sup>6</sup> (n-1)	Refractivity Normalized by Number Density (N-units/molecule/cm <sup>3</sup> )	Absorptivity (dB/km)	Source
2,255	$T = 295 \pm 1 K$ $P = 5 \pm .2 atm$	1354 ± 70	$(1.0886 \pm .104) \times 10^{-17}$	< .14	This work
2,255	T = 188 K ± 10 K P = 5.5 ± .2 atm	2156 ± 120	$(.9979 \pm .145) \times 10^{-17}$	< .05	This work
2.255	T = 156 K + 12 K - 5 K	2509 ± 130	$(.9422 + .154) \times 10^{-17}$	< .1	This work
	$P = 5.7 \pm .2 atm$				
9,28	T = 273 K P = 1 atm	293.4 ± 1.4	$(1.0912 \pm .005) \times 10^{-17}$		Birnbaum, Kryder and Lyons (1951)
13.3	$T = 295 \pm 1 K$ P = 6 ± .2 atm	1475 ± 75	$(.988 \pm .086) \times 10^{-17}$	< 9	This work
21.7	T = 295 ± 1 K P = 5.7 ± .2 atm	1447.0 ± 75	$(1.059 \pm .096) \times 10^{-17}$	< 4.5	This work
24.0	T = 273 K P = 1 atm	294.1 ± 0.1	$(1.0938 \pm .0004) \times 10^{-17}$		Essen and Froome (1951)

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properties of gases under simulated conditions for the outer planets.

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Figure 2: Sketch of pressure vessel, as viewed from above, with the top plate removed. Note the welded flange, with groove for the O-ring.

#### VIII. APPENDICES

PROBING VENUS Remote sensing technology gives researchers

detailed new data about the Venusian atmosphere

emote sensing from space has given the human race a new perspective on itself. With vivid satellite portraits of the earth, planners can see pollution in coastal marshlands, insect infestation, fishing beds and a wealth of other important information. Now, the same basic technology is letting scientists map the atmospheres of planets millions of miles away. At Georgia Tech, Dr. Paul Steffes, an assistant professor of electrical engineering, has used remote sensing to "see" the makeup and structure of sulfuric acid in the midatmosphere of Venus.

The raw data for his research were microwave signals transmitted by Mariner and Pioneer spacecraft as they flew behind Venus. Before returning to earth, these transmissions penetrated the Venusian atmosphere, where they were either bent (refracted) or weakened (attenuated). The resulting signal modifications were collected by the Deep Space Network of antennas at stations in California, Spain and Australia.

"Most of the refraction is due to the density of the atmosphere," says Steffes. "It's like a light wave being bent as it passes through the ocean."

Weakened signals indicate an absorption of electromagnetic energy by the atmosphere. Since only certain gases absorb electromagnetic waves, this information becomes a key for accomplishing this on a parts-

Remote sensing technology used by a Georgia Tech professor is helping uncover new knowledge about the planet Venus.

per-million level through a laboratory simulation of Venus' mid-atmosphere. With funding from NASA, he and his students built a small pressure chamber. They first injected gases known to be in the planet's atmosphere and subjected them to pressures and temperatures which exist at certain altitudes above Venus. Next, they propagated signals through the chamber at the same frequencies originally transmitted by NASA spacecraft back to earth. Finally, they manipulated the concentration of gases present until they were able to bring about the same degree of signal attenuation or refraction observed through remote sensing. Through these experiments, Steffes' group found that they could calculate concentrations of a microwaveabsorbing vapor, sulfuric acid, in the mid-atmosphere of Venus.

The accuracy of this method has proven greater than achieved by probes dropped into the Venusian atmosphere by American spacecraft earlier exploring the planet. Simulations using remote sensing data have resolved sulfuric acid vapor abundances down to an order of 10 parts-per-million. The picture revealed by

these experiments has been somewhat surprising. "Approximately 20 parts-permillion of sulfuric acid vapor exist below the clouds of Venus," Steffes says. "In the altitude range of 30 to 48 kilometers there is sulfuric acid vapor which condenses at 48 kilometers. Initially, it wasn't thought that that amount of sulfuric acid might have existed. It was thought that sulfuric acid clouds existed but that immediately below the clouds only sulfur dioxide existed." Steffes' current work grew



This pressure chamber developed by Dr. Paul Steffes simulates atmospheric conditions on Venus and th outer planets.

of these planets, Steffes has

acquired a deep freeze which

can reach - 120 ° C., well above

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temperature of Uranus but

close enough for accurate

sent an entirely different

The outer planets will pre-

challenge than Venus. Accord-

ing to Steffes, some scientists

describe them as "suns that

never quite made it" - that is,

relatively small rocky cores.

Their atmospheres are very

highly pressurized and com-

posed primarily of hydrogen

large gaseous balls with

extrapolations of data.

out of remote sensing experiments he made in the early 1980s as a doctoral degree student at Stanford University. However, his interest in the field began during his underalready have passed lupiter and graduate years at the Massa-Saturn, Last January, Voyager II chusetts Institute of Techstarted sending data back from nology. There, remote sensing Uranus. In another three years, techniques were being used to the ship will make its approach learn about the abundance of to Neptune and its moon certain gases in the upper part Triton. of the earth's atmosphere. To simulate the atmospheres

"At MIT, we were able to infer how much oxygen and water vapor was in the earth's atmosphere," Stelfes says, "so we said: if it works for us here we can apply it to other planets too."

The implications of his findings for Venus are uncertain, but Steffes says that they could provide evidence for volcanic activity on the planet. He also believes that knowledge of how sulfuric acid moves in the atmosphere of Venus possibly can help scientists better understand the acid rain problem on earth.

With the Venus simulations completed, NASA is sponsoring Steffes in a program to interpret remote sensing data on outer planets. Voyagers 1 and III

will till the deep ireeze with these gases, then start adding other gases, such as ammonia and methane, trying to match the attenuation and refraction of the Voyager microwave data. The work on Jupiter, Saturn and Uranus will probably take a year and a half.

What will be the value of his findings? "We'll be learning a lot about the nature of the formation of planets," Steffes says. "That tells us in the long run about how the universe is formed and whether or not there may be planets around other stars. So the applications are long term, obviously. They aren't necessarily something where we're going to be able to turn out a new widget next week, but the implications are pretty significant."[]

Appendix

GEORGIA INSTITUTE OF TECHNOLOGY

GEORGIA INSTITUTE OF TECHNOLOGY



Tuesday, June 10, 1986

THE ATLANTA CONSTITUTION

## Tech making down-to-Earth studies of outer space

#### By Robert Lee Hotz

Science/Medicine Writer

Inside a squat, square freezer at the Georgia Institute of Technology, scientists are seeking clues to the frigid gases that envelop the solar system's outer planets in a project that may yield new insights into the evolution of Earth's atmosphere.

By interpreting subtle distortions in radio signals from the Voyager 2 space probe, Georgia Tech researchers are mapping the exotic atmospheres of Jupiter, Saturn, Uranus and Titan, Saturn's largest moon, then duplicating the volatile blend of gases in a pressurized laboratory freezer.

Analyzing microwave signals from unmanned Mariner and Pioneer probes, they have detected corrosive clouds of sulfuric acid that dot the skies of Venus. Using large Earth-based radiotelescopes, they hope one day to track the daily weather on Venus or detect any active volcances.

"We are doing this research not only so we could better understand these other planets, but also to understand how our own atmosphere might be evolving," says Dr. Paul Steffes, a Georgia Tech professor of electrical engineering who leads the research team.

"We are learning how fragile our own atmospheric balance is — because if the scales the one way, we could end up very hot like Venus or, if the scales tip the other way, we could end up like Mars. It is a very deticate balance that keeps our atmosphere so amenable to life."

None of the probes carries instruments designed to detect atmospheric gases. But when a satellite passes behind a planet or moon, the radio signals being transmitted back to Earth are garbled briefly by interference from the gases surrounding the other planet. Steffes has turned that distortion into a tool for mapping the alien . atmosphere itself.

"Using the U.S. Deep Space Network, we are ahle to measure the amount of weakening in the microwave signal as it passed through the planet's atmosphere." Steffes says. "The amount of microwave radiation absorbed or refracted depends on what gases are in the planet's atmosphere. The question for us is what gases and how much," he says.

Any weakening of the satellite's signal indicates the presence of chemical compounds that absorb microwave energy. By pinpointing the frequency blocked, researchers can decipher the chemical signature of gases such as helium, hydrogen and sulfuric acid. In addition, any bending of the microwaves is an effective barometer of the atmospheric pressure, Steffes says.

"Sulfuric acid vapor, for instance, has a very unique frequency signature," Steffes says. "Using the Pioneer orbiter, we could resolve the amount of sulfuric acid vapor to an accuracy better than 10 parts per million.

"Sulfuric acid vapor is like water in the Venus atmosphere. The clouds are of sulfuric acid vapor," he says.

Like house painters trying to match the proper pastel hue. Steffes and his assistants double-check their interpretations of the radio data by mixing gas samples in a stainless steel pressure chamber and bombarding it with microwaves. They continue to adjust the mix until the microwave distortion matches that actually measured from the orbiting spacecraft.

To simulate the furnace-like surface temperatures of Venus, they cooked the laboratory pressure chamber in an oven at 600 degrees. Now, to simulate the frigid temperatures of the outer planets, Steffes is using a special freezer that can lower the chamber temperature to less than 125 degrees below zero. Probing the planets

Earth station

Atmospheric diffraction:

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As the Voyager 2 spacecraft passed Jupiter, Saturn and Uranus, it sent back to Earth electronic signals that first penetrated the planet's atmosphere. The distortion of the signal indicates the presence of gases that absorb microwave energy.

RON CODDINGTON/Staff

Voveder 2

# THE GEORGIA TECH

## OR THE GEORGIA TECH COMMUNITY - VOLUME 12, NUMBER 18 - MAY 26, 1986

### Remote Sensing Reveals Much About Planets

By Mark Hodges, Research Communications Office

A deep freeze in a Tech laboratory is unlocking secrets about Jupiter, Saturn and Uranus.

Dr. Paul Steffes, an electrical engineering professor, is using the freezer to simulate the frigid atmospheres of these planets.

"Knowing the air pressures and gas mixtures on the outer planets," he says, "is a building block to learning more about how our solar system was formed."

Steffes describes the outer planets as "suns that never quite made it"—large gaseous balls with relatively small rocky cores. Their atmospheres are highly pressurized and composed mainly of hydrogen and helium.

Steffes is accurately simulating them with microwave data from the Voyager II mission. As this spacecraft passed Jupiter, Saturn and Uranus, it sent back to earth electronic signals which first penetrated the atmospheres of these planets.

Atmospheric distortion of the signals is the tool Steffes uses for mapping the atmosphere itself. In the simplest terms, weakening of the signal indicates the presence of gases which absorb microwave energy. Bending of the microwave shows high atmospheric pressure.

Steffes already has used microwave data from the Mariner and Pioneer probes to map the middle atmosphere of Venus. He discovered that the earth's sister planet has even more sulfuric acid vapor in the air than previously suspected: 20 parts per million at 30 to 48 kilometers' altitude.

This finding could provide evidence for volcanic activity on Venus, Steffes believes.

To reach this conclusion, he and his students built a small pressure chamber. They first injected gases into it known to be in the Venusian atmosphere and subjected them to pressures and temperatures which exist above Venus.

They then beamed microwaves through the chamber. Finally, they modified the mixture of gases in the chamber until the microwaves were bent or weakened as in actual measurements of Venus' atmosphere.

Steffes will use the deep freeze in a similar way. With it, he can reach -120°C. This is well above the -190°C. atmospheric temperature of Uranus but close enough for accurate comparisons to be made.

The work on Jupiter, Saturn and Uranus will take approximately a year and a half. Later, he hopes to analyze Voyager microwave signals from Neptune.

"We'll learn a lot about how the planets were formed," Steffes says. "That will tell us in the long run about how the solar system came into being and whether or not there may be planets around other stars."

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Multi-County Star (Covington)

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## Jupiter's secrets unlocked at Tech

A deep freeze in a Georgia Tech laboratory is unlocking secrets about Jupiter, Saturn and Uranus.

Paul Steffes, an electrical engineering professor at Tech, is using this freezer to simulate the frigid atmospheres of these planets.

"Knowing the air pressures and gas mixtures on the outer planets," he says, "is a building block to learning more about how our solar system was formed."

Steffes describes the outer planets as "suns that never quite made it" — large gaseous balls with relatively small rocky cores. Their atmospheres are highly pressurized and composed mainly of hydrogen and helium.

Steffes is accurately simulating them with microwave data from the Voyager II mission. As this spacecraft passed Jupiter, Saturn and Uranus, it sent back to earth electronic signals which first penetrated the atmospheres of these planets.

Atmospheric distortion of the signals is the tool Steffes uses for mapping the atmosphere itself.

In the simplest terms, weakening of the signal indicates the presence of gases which absorb microwave energy. Bending of the microwave shows high atmospheric pressure.

Steffes already has used microwave data from the Mariner and Pioneer probes to map the middle atmosphere of Venus. He discovered that the earth's sister planet has even more sulfuric acid vapor in the air than previously suspected: 20 parts per million at 30 to 48 kilometers' altitude.

This finding could provide evidence for volcanic activity on Venus, Steffes believes.

To reach this conclusion, he and his students built a small pressure chamber. They first injected gases into it known to be in the Venusian atmosphere and subjected them to pressures and temperatures which exist above Venus. Next, they beamed microvaves through the chamber. Finally, they modified the mixture of gases in the chamber until the microwaves were bent or weakened as in actual measurements of Venus' atmosphere.

Steffes will use the deep freeze in a similar way. With it, he can reach -120°C. This is well above the -190°C. atmospheric temperature of Uranus but close enough for accurate comparisons to be made.

The work on Jupiter, Saturn and Uranus will take approximately a year and a half. Later, he hopes to analyze Voyager microwave signals from Neptune.

"We'll learn a lot about how the planets were formed," Steffes says. "That will tell us in the long run about how the solar system came into being and whether or not there may be planets around other stars."

E-21-643

#### REPORT

#### TO THE

#### NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

ANNUAL STATUS REPORT (Includes Semiannual Status Report #6)

for

#### GRANT NAGW-533

## LABORATORY EVALUATION AND APPLICATION OF MICROWAVE ABSORPTION PROPERTIES UNDER SIMULATED

CONDITIONS FOR PLANETARY ATMOSPHERES

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February 1, 1986 through January 31, 1987

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#### I. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often leads to significant misinterpretation of available opacity data. Steffes and Eshleman (1981) showed that under environmental conditions corresponding to the middle atmosphere of Venus, the microwave absorption due to atmospheric  $SO_2$  was 50 percent greater than that calculated from Van Vleck-Weiskopff theory. Similarly, results obtained for the microwave opacity from gaseous  $H_2SO_A$  under simulated Venus conditions, during the first two years of Grant NAGW-533 (February 1, 1984 through January 31, 1986), showed that not only was the opacity from  $H_2SO_A$  much greater than theoretically predicted, but that its frequency (wavelength) dependence was far different than that theoretically predicted (Steffes, 1985 and Steffes, 1986a). The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by both radio occultation experiments and radio astronomical observations, and over a range of frequencies which correspond to those used in both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has

been the goal of this investigation to conduct such measurements and to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profiles of constituents in those planetary atmospheres.

In the first two years of Grant NAGW-533 (i.e., February 1, 1984 through January 31, 1986), this facility was developed, and then operated, in order to evaluate the microwave absorbing properties of gaseous sulfuric acid  $(H_2SO_4)$ under Venus atmospheric conditions. The initial results, obtained at 13.4 cm and 3.6 cm wavelengths, were applied to measurements from Mariner 5, Mariner 10, and Pioneer-Venus Radio Occultation experiments, to determine abundances of gaseous sulfuric acid in the Venus atmosphere, with accuracies exceeding those achieved with in-situ instruments (Steffes, 1985). Measurements of the microwave properties of the vapors accompanying liquid H<sub>2</sub>SO<sub>A</sub> also resulted in more accurate estimates of the vapor pressure behavior of sulfuric acid, which are critical for modeling the behavior and structure of the Venus atmosphere. Later efforts concentrated on making laboratory measurements of the microwave absorption from gaseous  $H_2SO_4$  at wavelengths from 1.2 to 22 cm under simulated Venus conditions. Additional measurements of the vapor pressure behavior of sulfuric acid were also made. We applied these results to radio astronomical observations of Venus which have been made in the same wavelength range, in order to better model the structure of H2SO4 and SO2 abundance in the Venus atmosphere, and to resolve temporal variations of their abundances on a planet-wide basis. The results of this effort have been especially rewarding in that the unique frequency and pressure dependences measured for the absorption from gaseous H2SO4 in these wavelength ranges has finally explained what were thought to be inconsistencies between measurements of the absorption

in the Venus atmosphere at 13.3 and 3.6 cm wavelengths and those obtained in the 1 to 3 cm wavelength range. We describe these effects, and the resulting limitations they place on abundances of gaseous  $H_2SO_4$  and  $SO_2$  in the Venus atmosphere, in a paper entitled, "Evaluation of the Microwave Spectrum of Venus in the 1.2 to 22 Centimeter Wavelength Range Based on Laboratory Measurements of Constituent Gas Opacities," which was submitted in February (1986) and has been published in <u>The Astrophysical Journal</u> (v. 310, pp. 404-414, November 1, 1986--Galley Proofs included as Appendix 6).

Additional activities conducted during this grant year (February 1, 1986 through January 31, 1987) involving application of our laboratory measurements of simulated Venus atmospheres have included assistance to the Magellan program office (J.P.L.) in characterizing effects of the atmosphere on planned microwave radar and radiometric experiments originally planned to characterize surface and geologic parameters. Our laboratory measurements have also suggested that a substantial dip in the Venus microwave emission, related to the abundance of gaseous sulfuric acid, should exist near the 2.25 cm wavelength. Since no observations of the Venus emission at this wavelength have ever been published, we have received permission to use the 140-foot NRAO telescope in May 1987 to not only confirm the presence of the predicted dip, but to use such a dip to determine a planet-wide average for sulfuric acid vapor abundance below the main cloud layer. (Support for travel to NRAO and use of the telescope will be provided by NRAO in response to our proposal, "Observations of the Venus Microwave Spectrum at 1.9 and 2.25 cm" - attached as Appendix 5.)

The highest priority activity for this grant year has been laboratory measurements of the microwave properties of the simulated atmospheres of the

outer planets and their satellites. As described a previous Annual Status Report for Grant NAGW-533 (February 1, 1985 through January 31, 1986), our planetary atmospheres simulator underwent a major redesign late last year to permit measurements of the microwave properties of simulated Jovian atmospheres. Further developmental work has been necessary in order to minimize the safety risks involved with high pressure simulations using hydrogen gas at very low temperatures, as required for the outer planets. A more complete description of this work is given in Section II of this report. However, since many of the low temperature, high pressure tests were initially conducted using the nonexplosive gas nitrogen  $(N_2)$ , which is the primary constituent of the atmosphere of the Saturnian satellite Titan, useful data for microwave refractivity and absorptivity of nitrogen atmospheres under simulated Titan conditions has been obtained at frequencies from 2.2 GHz to 21.7 GHz (1.3 cm to 13.7 cm). Such data is useful in interpretation of both Voyager 1 radio occultation measurements of the Titan atmosphere, as well as radio astronomical observations of Titan.

The most significant accomplishment of this grant year has been the successful completion of laboratory measurements of the microwave opacity of gaseous ammonia  $(NH_3)$  in a hydrogen/helium  $(H_2/He)$  atmosphere, under simulated conditions for the outer planets. These measurements were conducted at frequencies from 1.3 GHz to 22 GHz (wavelengths from 1.3 cm to 22 cm), at temperatures from 178 K to 300 K, and under total pressures reaching as high as 6 atmospheres. Such measurements have long been sought by a number of researchers working on inferring ammonia abundance profiles in Jovian atmospheres. (See, for example, de Pater and Massie, 1985, or West <u>et al.</u>, 1986.) Our measurements represent the first time that measurements of the

microwave absorption of gaseous ammonia under simulated conditions for the outer planets have been conducted. The results of these measurements, and their effect on the interpretation of microwave opacity data obtained both from <u>Voyager</u> radio occultation measurements made at 13 cm and 3.6 cm wavelengths, and from radio astronomical observations in the 1.3 cm to 22 cm wavelength range, are discussed in Section IV of this report.

Beyond this current grant year, our goals are to continue such laboratory measurements of the microwave absorption and refraction from other potential microwave absorbers contained in the outer planets' atmospheres, including methane (CH<sub>4</sub>), water vapor (H<sub>2</sub>O), and phosphine (PH<sub>3</sub>), as well as additional high-sensitivity mesurements of the absorption from gaseous NH<sub>3</sub> at 13.3 and 18.5 cm. We likewise would hope to be able to pursue a program of further analysis and application of these results to microwave data for the outer planets, such as Voyager Radio Occultation experiments and earth-based radio astronomical observations.

Of equal importance, we feel however, would be the further analysis and application of our laboratory results for the microwave absorption from gaseous  $H_2SO_4$  in the Venus atmosphere. Our long term goal would be a detailed analysis of available multi-spectral microwave opacity data from Venus, including data from the Pioneer-Venus Radio Occultation experiments and earth-based radio and radar astronomical observations, such as the kinds which have been performed at the NRAO Very Large Array (VLA) and at stations in the Deep Space Network (DSN). The new measurements of Venus microwave emission at 2.25 cm and 1.9 cm made with the NRAO 140-foot telescope will be an especially important contribution to this data set. This would provide a chance to determine both spatial and temporal variations in the abundances of both  $H_2SO_4$  and  $SO_2$  in the Venus atmosphere.

Further discussion of proposed future activity, both regarding the outer planets and Venus, is included in the accompanying proposal to NASA entitled, "Laboratory Evaluation and Application of the Microwave Properties of Simulated Planetary Atmospheres."

#### II. THE GEORGIA TECH RADIO ASTRONOMY AND PROPAGATION (R.A.P.) FACILITY

The basic configuration of the planetary atmospheres simulator developed at Georgia Tech for use in measurement of the microwave absorptivity of gases under simulated conditions for planetary atmospheres is described at length in the first two Annual Status Report(s) for Grant NAGW-533 (February 1, 1984 through January 31, 1985 and February 1, 1985 through January 31, 1986). It is also discussed at length in Steffes (1985 and 1986b). The updated simulator system, shown in Figure 1, is currently configured for simulations of the outer planets. Measurements of the microwave opacity and refractivity of test gas mixtures can be performed at frequencies from 1.3 GHz to 27 GHz (wavelengths from 1.2 cm to 22 cm). While the pressure chamber itself is capable of containing pressures up to 10 atmospheres, and the temperature chamber is capable of achieving temperatures as low as 150 K, it has been found that the combination of high pressures and low temperatures can create a substantial problem with sealing the pressure chamber. In previous simulations, small leakages from the pressure vessel presented little or no danger to the experimenters. The use of gaseous hydrogen  $(H_2)$  in the outer planets simulations has required the development of new procedures and equipment for conducting such simulations. Such precautions have included the addition of a hydrogen leakage sensor which is placed inside the temperature chamber immediately outside the pressure vessel (see Figure 1). This sensor can

detect the potentially dangerous build-up of hydrogen gas within the freezer unit. A ventilation pump has also been provided which can be used to draw any escaping hydrogen gas out of the freezer compartment. Additional precautions have included the construction of ramps which allow all of the equipment to be moved out-of-doors to a concrete slab immediately adjacent to the laboratory. All experiments which employ gaseous hydrogen can thus be conducted out-ofdoors in order to avoid any build-up of hydrogen gas within the laboratory. A covered outdoor storage area for hydrogen and helium gases has also been constructed in order to allow safe storage of these gases, and to expedite the out-of-doors experiments. It is noteworthy that funding for the hydrogen leakage sensor, for construction of the outdoor gas storage and experimental areas, and for special tools required for sealing the pressure vessel (over \$2,000) has been provided by the Georgia Institute of Technology, in support of planetary atmospheres research at Georgia Tech. In addition, two vacuum sensors have been added to the system. These sensors not only allow accurate determination of pressure vessel evacuation, but they can also be used for accurately determining the abundances of microwave-absorbing test gases, which are typically very small at low temperatures, due to low saturation vapor pressures.

Initial testing of the new simulator configuration for the outer planets was conducted using nitrogen  $(N_2)$  gas both because of its relative safety as compared with hydrogen, and because measurements of both the refractivity and absorptivity of nitrogen under simulated conditions for the Saturnian satellite Titan were considered important for interpreting results of the Voyager 1 radio occultation studies of the Titan atmosphere (Lindal <u>et al.</u>, 1983 and 1985).

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The first tests of the system showed that the pressure vessel could easily hold pressures of 7 atmospheres at room temperature with essentially zero leakage. However, when the system was cooled to 150 K, significant leakage occurred. A quick review of pressure vessel construction can shed some light on this problem. The pressure vessel is essentially a 13-inch diameter stainless steel cylinder (0.375-inch wall thickness) with a 0.375inch thickness stainless steel plate welded to the bottom of the cylinder. At the top of the cylinder is a 0.5-inch thick cylindrical flange which is welded to the cylinder. The top plate (0.5-inch thickness stainless steel) bolts to the flange using 16 stainless steel bolts, along with a pressure sealant O-ring, and sealant putty which is placed outside the 0-ring. (See Figure 2.) All electrical connections are made via hermetically sealed feed-through connectors mounted on the top plate.

Two causes for the increased leakage rate at extremely low temperatures have been found. The first was simply due to metal contraction. When the pressure vessel was cooled to 150 K, it was found that most of the fastener bolts had loosened due to contraction of the stainless steel. Thus, removal of the cooled pressure vessel from the freezer was necessary so that the bolts could be tightened while it was still very cold, and it was then immediately replaced inside the freezer. The special lifting frame and pulley system which was designed for easily moving the pressure vessel in and out of the freezer has been especially useful for this task. However, even after retightening of the fastener bolts on the cold pressure vessel, substantial leakage still occurred.

The cause of the continued leakage was found to be the sealing materials used in the pressure vessel. As shown in Figure 2, the seal used when placing

the top plate on the pressure vessel consists of a 0.385-inch cross-section O-ring accompanied by a ring of putty-type sealing material. As was the case in the recent Space Shuttle accident, seals which normally function well at room temperature can malfunction when placed under extremely cold conditions. This is due to increasing brittleness of the O-ring and putty seals, accompanied by a higher probability of cracks developing due to the brittleness. Initially, as suggested by several seal material vendors, we used an O-ring made from vulcanized rubber and RTV-silicone putty. The relatively poor performance of this seal caused us to experiment with several other types of sealing materials. We found that nearly all materials typically used in such seals became brittle at 150 K. However, we found that O-rings fabricated from the material Viton were less likely to develop cracks at this temperature. It was also found that RTV-silicone became extremely brittle at 150 K, and was very susceptible to cracking. As a result, we now use a more fibrous silicone composite material for the sealing putty.

The overall result has been a pressure vessel capable of maintaining 6 atmospheres of pressure at a temperature of 150 K, with an acceptably small leak ratio. While the range of pressures which can be tested is not quite as large as originally hoped, the resulting range of temperatures and pressures does represent the range over which nearly all of the microwave opacity in the Jupiter atmosphere has been observed, and thus is extremely useful in interpretation of microwave opacity data from Voyager I and II radio occultation experiments, as well as from earth based radio astronomical observations, and, in the future, opacity measurements to be made using the Galileo probe. Likewise, the pressure-temperature ranges measured will be close enough to those over which microwave absorption or refraction has been measured in the

atmospheres of Saturn, Titan, Uranus, and Neptune, so that accurate estimates of abundances of microwave-absorbing constituents in these atmospheres can also be made.

#### **III. EXPERIMENTAL APPROACH**

The two key measurements made in the past grant year with the Georgia Tech planetary atmospheres simulator have been the measurements of the refractivity of gaseous nitrogen  $(N_2)$  under conditions similar to the Titan atmosphere and the measurements of the microwave absorption from gaseous ammonia  $(NH_3)$  under simulated conditions for the outer planets. It should be noted, however, that the techniques described for measuring refraction and absorption can be applied to any gases which are placed in the simulator.

The refractive index, n, of a gas is the ratio of the velocity of an electromagnetic wave in a vacuum, c, to the velocity of the wave in the presence of the gas,  $V_g$ . Since the resonant frequencies of a microwave resonator are directly proportional to the velocity of electromagnetic waves within the resonator, the refractive index of a gas can be determined by comparing the frequency of a particular resonance when a vacuum is present within the resonator,  $f_v$ , to the resonant frequency when the gas is present,  $f_q$ . That is,

$$n = c/V_{g} = f_{v}/f_{g}$$
(1)

Refractivity, N, is defined as being equal to  $(n-1) \ge 10^6$ . Thus, it can be determined simply by measuring the <u>change</u> in center frequency of a given resonance after the gas has been introduced. That is,

$$N = (n-1) \times 10^{6} = \left[ (f_v - f_g) | f_g \right] \times 10^{6}$$
(2)

Since the refractivity of a gas is directly proportional to the molecular density of the gas,  $\rho$ , the refractivity is often expressed in a form which is normalized by molecular density in terms of the temperature and pressure of the gas. That is,  $\rho = P/RT$  where  $\rho$  is density in molecules per cm<sup>3</sup>, P is the pressure in atmosphere, R is the ideal gas constant (1.362344 x 10<sup>-22</sup> cm<sup>3</sup>-atm/molecule/Kelvin), and T is the temperature in Kelvins. Thus, the density-normalized refractivity, N/ $\rho$ , can be expressed as NRT/P. It is often assumed that the density-normalized refractivity for a gas is independent of pressure, temperature, or frequency.

It is this assumption which allowed Lindal <u>et al.</u> (1983) to infer a temperature-pressure profile for Titan's atmosphere from measurements of atmospheric refractivity at 2.3 GHz (13 cm) made with Voyager 1. That is, by assuming an exponential atmosphere with surface temperature 94 K, and by assuming a constant value for the density-normalized refractivity of N<sub>2</sub> (the predominant constituent) to be  $1.093 \times 10^{-17}$  N-units/molecule/cm<sup>3</sup>, the measured refractivity profile for Titan's atmosphere was used to infer a temperature-pressure profile for the atmosphere. This value for normalized refractivity was obtained by Essen and Froome (1951) based on a single measurement of the refractivity of nitrogen (N<sub>2</sub>) under standard laboratory conditions at a single frequency (24 GHz). The need to confirm by laboratory measurement the nature of the refractive properties of nitrogen at 2.3 GHz under conditions similar to those of Titan is considered critical for the proper interpretation of Titan refractivity data.

In order to measure the refractivity of nitrogen, the pressure vessel and its microwave resonators (see Figure 1) must first be cooled to the desired temperature. Because of the large mass of the pressure vessel and microwave resonators (over 400 pounds), nearly 24 hours are required to reach thermal equilibrium at temperatures below 195 K using our current freezer system. Temperatures within the temperature chamber (freezer) are monitored by two temperature sensors: the first being placed within the chamber, but outside the pressure vessel; and the second being placed within the pressure vessel. Another technique used in monitoring the cooling of the resonator within the pressure vessel is to monitor its resonant frequencies. As the resonator is cooled, the resonant frequencies drop due to thermal contraction. Thus, when thermal stability is reached, the resonant frequencies likewise stabilize. After thermal stability is reached, a vacuum is drawn in the pressure vessel containing the two microwave resonators, and the center frequencies of the resonances of interest are then measured. For this experiment (refractivity of N<sub>2</sub>), resonances at 2.26 GHz (13.3 cm), 8.53 GHz (3.52 cm), 13.3 GHz (2.26 cm), and 21.77 GHz (1.38 cm) were used. A valve is then opened which admits the test gas (N2 in this case) to the chamber. The nitrogen is admitted at a sufficiently slow rate so as not to significantly affect the temperature within the chamber. As the gas is added, the shifting of the center frequencies of the various resonances can be observed. Once the desired pressure is reached (pressures between 1 atm and 6 atm were used in our experiments), the total frequency shift can be used to compute the refractivity, N, of the gas under those conditions. It should be noted that while the major source of uncertainty for our refractivity measurements is the frequency measuring capability of our system, two other sources of uncertainty

affect the accuracy of our determination of <u>density-normalized</u> refractivity, and those are our abilities to measure pressure and temperature. For example, in some early measurements a less accurate pressure gauge was used. Thus, the error bars for the measurement of density-normalized refractivity were larger than those for similar measurements made later. The results of these measurements are discussed in Section IV of this report.

The approach used to measure the microwave <u>absorptivity</u> of gaseous  $NH_3$  in an  $H_2/He$  atmosphere is similar to that used previously by Steffes (1985 and 1986a) for simulated Venus atmospheres. As can be seen in Figure 1, the absorptivity is measured by observing the effects of the introduced gas mixture on the Q, or quality factor, of two cavity resonators at particular resonances from 1.34 GHz to 21.8 GHz. The changes in the Q of the resonances which are induced by the introduction of an absorbing gas mixture can be monitored by the high resolution microwave spectrum analyzer, since Q is simply the ratio of the cavity resonant frequency to its half-power bandwidth. For relatively low-loss gas mixtures, the relation between the absorptivity of the gas mixture and its effect on the Q of a resonance is straightforward:

$$\alpha \approx \left(Q_{\rm L}^{-1} - Q_{\rm C}^{-1}\right) \pi / \lambda \tag{3}$$

where  $\alpha$  is absorptivity of the gas mixture in Nepers km<sup>-1</sup>. (Note, for example, that an attenuation constant or absorption coefficient or absorptivity of 1 Neper km<sup>-1</sup> = 2 optical depths per km (or km<sup>-1</sup>) = 8.686 dB km<sup>-1</sup>, where the first notation is the natural form used in electrical engineering, the second is the usual form in physics and astronomy, and the third is the common (logarithmic) form. The third form is often used in order to avoid a

possible factor-of-two ambiguity in meaning.)  $Q_L$  is the quality factor of the cavity resonator when the gas mixture is present,  $Q_C$  is the quality factor of the cavity resonator in a vacuum, and  $\lambda$  is the wavelength (in km) of the test signal in the gas mixture.

In order to obtain a gas mixture with a sufficient amount of gaseous  $NH_3$ so that microwave absorption is detectable using our system, temperatures at or above 170 K must be used. (This limit is set by the saturation vapor pressures for ammonia and by the sensitivity of our measurement system.) While this covers most of the temperature range in the Jupiter atmosphere over which radio occultation and radio astronomical experiments have detected microwave opacity (140-300 K), it is somewhat above the temperature range over which microwave opacity has been detected at Saturn. However, the measured temperature dependencies can be used to extrapolate to those temperatures. In order to conduct the required measurements, the pressure vessel and its microwave resonators must first be cooled to the desired temperature. As with the refractivity measurements, this requires over 24 hours of cooling in the temperature chamber. Before the cooling process begins, the chamber is filled with 6 atm of a neutral gas (either helium or nitrogen). This is because the pressure vessel achieves a much better seal if it is cooled while under pressure.

After thermal stability is reached, which can be monitored using both the temperature sensors and the resonant frequencies of the system, a vacuum is drawn in the pressure vessel containing the resonators, and the bandwidth and center frequency of each of resonances is then measured. For this experiment (absorption from  $NH_3$ ), resonances at 1.34 GHz (22.3 cm), 2.25 GHz (13.3 cm), 8.53 GHz (3.52 cm), 13.3 GHz (2.26 cm), and 21.7 GHz (1.38 cm) were used.

A valve is then opened which allows the ammonia gas to enter the chamber. For the experiments conducted at 178 K, only about 2 torr NH3 pressure was admitted so as not to cause saturation or condensation. For all other experiments (193 K and above), 20 torr NH3 pressure was used. Measurements of the gaseous NH3 pressure were made with the high accuracy thermocouple vacuum gauge tubes which are shown in Figure 1. Next, 5.4 atm of hydrogen (H2) and 0.6 atm of helium (He) are added. These gases are admitted to the chamber at a sufficiently slow rate so as not to significantly affect the temperature The result is an atmosphere with 6 atm total pressure within the chamber. composed of 90 percent hydrogen, 10 percent helium, and approximately 4300 ppm ammonia (except for the measurements conducted at 178 K, where the ammonia abundance was approximately 430 ppm). The bandwidth of each resonance is then measured and compared with its value when the chamber was evacuated in order to determine the absorptivity of the gas mixture at 6 atm total pressure. The total pressure is then reduced by venting to 4 atm, and the bandwidths are again measured. Subsequent measurements are likewise made at lower pressures in order to determine absorptivities at those pressures. The pressure vessel is then evacuated and the bandwidths again measured so as to assure no variation (either due to thermal shift or chemical reaction) of the Q's of the evacuated resonators has occurred. The measured changes of bandwidth (Q) can then be used to compute the absorptivity of the gas mixture under the various pressure conditions.

This approach has the advantage that the same gas mixture is used for the absorptivity measurements at the various pressures. Thus, even though some small uncertainty may exist as to the mixing ratio of the initial mixture, the mixing ratios at all pressures are the same, and thus the uncertainty for any

derived pressure <u>dependence</u> is due only to the accuracy limits of the absorptivity measurements, and not to uncertainty in the mixing ratio. (This assumes that the mixing ratio is small, so that foreign-gas broadening predominates, as is the case for our measurements.) Similarly, measurements of the frequency <u>dependence</u> of the absorptivity from the mixture would likewise be immune to any mixing ratio uncertainty, since foreign-gas broadening predominates.

#### IV. RESULTS OF LABORATORY MEASUREMENTS AND THEIR APPLICATION

Measurements made of the microwave refraction (and absorption) from Nitrogen  $(N_2)$  under simulated conditions for Titan are shown in Table I. While no absorption was measured at any frequency from 2.2 GHz to 21.7 GHz, upper limits for the opacity from  $N_2$  can be set. This is especially important since  $N_2$  may very well be the source of the 3.6 cm (8.4 GHz) opacity detected by Voyager I radio occultation studies of the Titan atmosphere (Lindal <u>et al.</u>, 1985). Also shown in Table I is the refractivity, N, and the refractivity normalized by molecule number density. Since the normalized refractivity is used directly to determine atmospheric pressure from measurements of the atmospheric refractivity at a given altitude, an accurate expression for normalized refractivity at the temperature of that altitude is required.

When inverting the Voyager I refractivity data obtained at 2.3 GHz at Titan, in order to develop a temperature-pressure profile for Titan's atmosphere, Lindal <u>et al</u>. (1983) assumed an exponential atmosphere with surface temperature 94 K, and a constant value for the density normalized refractivity of gaseous N<sub>2</sub> (1.093  $\pm$  0.0004 N-units/molecule/cm<sup>3</sup>). This value for normalized refractivity was obtained by Essen and Froome (1951) based on a

single measurement of the refractivity of nitrogen under standard laboratory conditions at a single frequency (24 GHz). This value is included, for comparison, in Table I. It should be noted that 4 additional data points are presented in this table, beyond those previously presented in Semiannual Status Report #5 (July 1986) for this grant. We have measured the refractivity of nitrogen over a wide range of temperatures (down to 156 K) and over a wide range of frequencies (2.2 GHz to 21.7 GHz) in order to determine whether any temperature or frequency variations of the density-normalized refractivity existed, and to estimate such effects on the interpretation of the refractivity data. While some variations were found between the measurements of the density-normalized refractivity of nitrogen, all were within experimental error of the values determined by Essen and Froome (1951) at 24 GHz and by Birnbaum et al. (1951) at 9.58 GHz. These results confirm that the density-normalized refractivity of nitrogen remains essentially constant, even at the very cold temperatures characteristic of the outer planetary systems. While our measurements did not go as low as the 94 K surface temperature of Titan, they do strongly indicate that the method for inferring the temperature-pressure profile of Titan's atmosphere from Voyager 1 refractivity data used by Lindal et al. (1983) was appropriate, and that the resulting profiles are accurate.

Measurements of the microwave absorption from  $NH_3$  in a 90% hydrogen  $(H_2)/10$ % helium (He) atmosphere have been conducted over a wide range of temperatures, pressures, and frequencies. For measurements conducted at a temperature of 178 K, with an ammonia mixing ratio of only 430 ppm, the error bars are quite large as a percentage of the measured value, since the opacity of the gas mixture is small. For the measurements made at 193 K and 300 K,

where an ammonia mixing ratio of 4300 ppm was used, the error bars are much smaller. Data for absorption from the gas mixtures was obtained at all of five resonant frequencies previously mentioned: 1.34 GHz (22.3 cm), 2.25 GHz (13.3 cm), 8.53 GHz (3.52 cm), 13.3 GHz (2.26 cm), and 21.7 GHz (1.38 cm). However, due to the relatively low opacity exhibited by the gas mixtures at 1.34 GHz, and likewise due to the relatively poor sensitivity of our system at that lowest resonance, the data obtained at 1.34 GHz was fairly noisy, and will require further analysis and calibration. The complete results of all measurements at all other frequencies are presented in tabular form in <u>Appendix 1</u>, by frequency: 2.25 GHz (Table 1), 8.53 GHz (Table 2), 13.3 GHz (Table 3), and 21.7 GHz (Table 4). In addition to the measured values for absorption, and the accompanying error bars, the theoretically-derived values for the opacity for each gas mixture under the conditions of each measurement are also given for comparison.

A more useful presentation of this data is given in Figures 3, 4, and 5. In Figure 3, the measured values of the absorptivity are plotted as a function of frequency (wavelength), for total gas mixture pressures of 6, 4, and 2 atmospheres. All measurements shown in this figure were made at a temperature of 193 K and with an NH<sub>3</sub> mixing ratio of  $4.3 \times 10^{-3}$ . Also shown are plots of the theoretically-derived absorption spectra for such gas mixtures at these three pressures. These theoretically-derived NH<sub>3</sub> absorption spectra were taken from the calculations of De Pater and Massie (1985) and Berge and Gulkis (1976), both of which employ a modified Ben-Reuven line shape. Our theoretical spectra have also been corrected for the temperature, pressure, and mixing ratio conditions of our experiment. Figure 4 presents similar results for data taken at 300 K with an NH<sub>3</sub> mixing ratio of  $4.3 \times 10^{-3}$ . Figure 5

presents results for a temperature of 178 K and an  $NH_3$  mixing ratio of only  $4.3 \times 10^{-4}$ , resulting in fairly large error bars, as previously discussed. It should be noted that the 13.3 GHz data points (2.26 cm) and data at 21.7 GHz taken on July 25, 1986, are not included in Figures 3, 4, and 5, both for the sake of clarity and because some calibration accuracy problems arose due to problems with connectors on that resonator, especially with the August 8 data set. Also, some data taken at 1 atm pressure was not included (even though it agrees well with the theoretical predictions for opacity at that pressure) for the sake of clarity.

Inspection of Figures 3, 4, and 5 shows that the laboratory data agree surprisingly well with the theoretical predictions for NH3 opacity made using the modified Ben-Reuven lineshape. Our results provide what De Pater and Massie (1985) characterized as "information which is desperately needed," in that they provide answers to a number of questions which have been published regarding the NH3 spectrum. The key finding is that the modified Ben-Reuven line shape appears to correctly describe the shape of the absorption spectrum from gaseous ammonia in a hydrogen/helium atmosphere in the 1.3 to 13.3 cm wavelength range even under the temperature-pressure conditions characteristic of the Jovian atmosphere. This finding answers some questions and raises others. For example, both de Pater and Massie (1985) and West et al. (1986) recognized that, based on interpretation of the Jovian emission spectrum in the 10 to 20 cm wavelength range using the theoretically-derived absorption spectrum from ammonia, opacity at pressure levels greater than 2 atm had to exceed the amount which would be caused by the solar abundance of ammonia. As a result, both sets of authors concluded that either the theoretical lineshape was incorrect and understated the opacity of ammonia at these wavelengths by a
factor of 1.5-2.0, or the ammonnia abundance was greater than its solar abundance by a factor of 1.5-2.0 at pressures greater than 2 atm, or that an extra opacity source, possibly  $H_2O$  condensate, was present. Thus, since our measurements of the ammonia opacity at 13.3 cm (2.25 GHz) agree quite closely with the theoretically-derived values for opacity, and since our preliminary work at 22.3 cm (1.34 GHz) is likewise consistent with the theoretical lineshape, it would appear that either the ammonia abundance is greater than solar by a factor of 1.5-2.0 in the deeper layers of the Jovian atmosphere, or that an extra opacity source is present.

Either or both of these scenarios are possible. For example, based on the somewhat localized Voyager 1 radio occultation studies, Lindal et al. (1981) concluded that at a pressure of 1 atm, the ammonia mixing ratio was 2.2 x  $10^{-4}$  (1.5 times the solar abundance). On the other hand, de Pater and Massie (1985) found that in order to best match Jupiter's microwave emission spectrum (planet-wide average, 1.3-20 cm), ammonia abundances of 3 x  $10^{-5}$  for pressures less than 1 atm,  $1.5 \times 10^{-4}$  at 1.5 atm, and 2.5 x  $10^{-4}$  at pressures greater than 2 atm were required. However, they did suggest that a solar abundance of ammonia  $(1.5 \times 10^{-4})$  at pressures greater than 1.5 atm would be consistent with the observed emission spectrum if a water cloud with mass density 35 g/m<sup>3</sup> were present and if the Ben-Reuven lineshape used in their calculation was correct. West et al. (1986) agreed by stating the only direct evidence which suggest that an H<sub>2</sub>O abundance great enough to form a cloud actually exists is the 10 to 20 cm spectrum, and only if it can be shown that WH<sub>3</sub> is incapable of supplying the required opacity. Our work is especially important in that it shows that a solar abundance of ammonia is indeed incapable of supplying the required opacity.

It may be possible to differentiate between the remaining two suggested possibilities (an ammonia abundance which is 1.5-2.0 times the solar abundance or a cloud layer in the 5-6 atm region) if a more accurate estimate of the frequency dependence of the atmosphere opacity in the 10 to 20 cm wavelength range could be obtained. We would then be able to determine which combinations of gaseous  $NH_3$ , liquid  $H_2O$ , and gaseous  $H_2O$  (in the 5 to 6 atm pressure range) could account for such a frequency (wavelength) dependence. In order to support this effort, we are proposing to make additional measurements in the laboratory of the microwave absorption from gaseous  $NH_3$  and gaseous  $H_2O$ under simulated conditions for the 5 to 6 atm pressure range of the Jovian atmosphere. These measurements would be conducted at 1.62 GHz (18.5 cm) and 2.25 GHz (13.3 cm), and would take advantage of special adjustments to the atmospheric simulator so as to maximize sensitivity in the 10 to 20 cm wavelength range. We would also hope to further study the possible contributions to the opacity from the cloud layer itself, or from other unrelated gases such as phosphine (PH3). For further discussion of proposed future activity, see the attached proposal, "Laboratory Evaluation and Application of the Microwave Properties of Simulated Planetary Atmospheres."

Another important aspect of our measurements of the opacity from ammonia under simulated Jovian atmospheric conditions are the results obtained at 21.7 GHz (1.38 cm). These results (shown in Figures 3-5 and listed in Appendix 1, Table 4) indicate that the absorption from ammonia is great enough at the 1.3 cm wavelength so as to require its depletion at pressures less than 0.5 atm in the Jovian atmosphere, as suggested both by Klein and Gulkis (1978) and by de Pater and Massie (1985). This drop in NH<sub>3</sub> abundance at altitudes above the 0.5 atm pressure level was also inferred by Lindal <u>et al.</u> (1981)

from Voyager 1 measurements of 13 cm and 3.6 cm radio opacity. Our laboratory measurements of the  $NH_3$  opacity at these wavelengths confirms this conclusion.

Although the temperatures over which most of our laboratory measurements have been made are somewhat higher than those at Saturn over which microwave opacity has been detected, our results can be extrapolated to allow analysis of the Saturn microwave emission spectrum and radio occultation data, similar to what we have done for Jupiter. In fact, a very similar problem regarding the presence of a potential water cloud, or an overabundance of  $NH_3$  (by a factor of 3 compared to its solar abundance) in the deep Saturn atmosphere could be resolved by applying our current results and future planned measurements. (Reference: Attached proposal).

At present, we are conducting preliminary studies of the microwave refraction and absorption from methane  $CH_4$ . These results for refractivity at 13.3 cm and 3.52 cm will be especially important for correctly inferring the temperature-pressure profiles of the atmospheres of Titan and Uranus from Voyager refractivity profiles near these wavelengths, since both have been shown to have significant quantities of methane. In addition, since the ammonia abundance at Uranus has been shown to be substantially depleted (see Gulkis <u>et al</u>., 1978), any measurement of, or limitation on, the microwave opacity from methane will be especially important for interpretation of any radio opacity data (both radio occulation measurements and radio astronomical observations). Likewise, due to the depleted ammonia abundance, the role of water vapor and condensed water as microwave absorbers in the deep Uranus atmosphere becomes more critical, hence, the further need for the proposed measurements of the microwave absorption from gaseous  $H_2O$  under simulated

conditions for the deep atmospheres of the outer planets. The results for ammonia, methane, and water vapor will likewise be invaluable for the interpretation of data from Neptune.

# V. PUBLICATIONS AND INTERACTION WITH OTHER INVESTIGATORS

At the beginning of the grant year, a paper was completed and accepted for publication in The Astrophysical Journal, describing results and applications of experiments performed during the second year of Grant NAGW-533 (Steffes, 1986a). This paper is described at length in Section I of this In May 1985, a paper was presented at the Conference on Jovian report. Atmospheres, at the Goddard Institute for Space Studies (New York) entitled, "Laboratory Measurements of Microwave Absorption from Gaseous Atmospheric Constituents under Simulated Conditions for the Outer Planets." This paper described our plans and capabilities for simulating outer planets atmospheres and measuring microwave properties of those atmospheres. At the beginning of this year, a revised manuscript was completed and submitted for inclusion in a NASA Conference Proceedings for this conference (Steffes, 1986b).

Our most recent results for the laboratory measurements of the microwave opacity of ammonia under Jovian conditions are being presented at the 18th Annual Meeting of the Division of Planetary Sciences of the American Astronomical Society (AAS/DPS Meeting - Abstract attached as Appendix 2). In a related meeting being held in conjunction with the DPS meeting, entitled, "Laboratory Measurements for Planetary Science," we are presenting a summary of our laboratory activities, and their application to the interpretation of planetary radio absorptivity and refractivity data. Support for travel to Paris for these meetings has been provided by the Georgia Tech Research

Corporation (GTRC) in support of planetary atmospheres research at Georgia Tech.

The results of our laboratory measurements of the microwave opacity of ammonia and their application to the interpretation of microwave opacity data from Jupiter are currently being prepared for submission as a paper to Icarus. The paper will be entitled, "Laboratory Measurements of the Microwave Opacity of Gaseous Ammonia (NH<sub>2</sub>) under Simulated Conditions for Jovian Atmospheres," and will be submitted in December. Similarly, our measurements of the refractive and absorption properties of nitrogen under conditions similar to the Titan atmosphere are being prepared for publication. More informal contacts have been maintained with groups at the California Institute of Technology (Dr. Duane O. Muhleman, regarding radio astronomical measurements of Venus opacity), at the Stanford Center for Radar Astronomy (V. Eshleman, regarding Voyager results for the outer planets, and laboratory measurements), and at JPL (Drs. Michael J. Klein, Michael Janssen, and Samuel Gulkis regarding radio astronomical observations of the outer planets, and A. J. Kliore regarding the Pioneer-Venus Radioscience Program). We have also had discussions with members of the Magellan Program Office at JPL with regard to using results from our laboratory measurements of simulated Venus atmospheres in planning mission experiments.

Another source of close interaction with other planetary atmospheres principal investigators has been Dr. Steffes' membership in the Planetary Atmospheres Management and Operations Working Group (PAMOWG). Travel support for attendance at PAMOWG meetings has been provided by Georgia Tech.

Several publications which describe our research activities in the Planetary Atmospheres to the public have also been released. Two examples are

shown in Appendix 3. The first, entitled "Probing Venus," was a two-page color article describing the results of our work simulating the microwave properties of the Venus atmosphere, and appeared in the publication <u>Research</u> <u>Horizons</u>, published by the Georgia Institute of Technology (Winter 1986, Volume 3, Number 4, pp. 8-9). We have also attached an article which appeared in the Tuesday, June 10 edition of <u>The Atlanta Constitution</u> (pg. 34a) and in the Tuesday, June 10 edition of <u>The Atlanta Journal</u> (pg. 22a), entitled "Tech Making Down-to-Earth Studies of Outer Space." This article likewise appeals to a large public audience. As in the past, we have maintained contact with members of the Georgia congressional delegation, both by telephone and by letter (see Appendix 4), keeping them aware of our work and aware of the need for continued support of the solar system exploration program.

### VI. CONCLUSION

This grant year (February 1, 1986 through January 31, 1987) has been both exciting and productive. Two papers have been published (<u>Astrophys. J</u> and NASA-CP), two conference papers are being presented, and two new papers are being submitted for publication. After long, arduous work with our simulator, measurements of the refractivity and absorptivity of nitrogen under conditions similar to those for Titan were completed. The most significant measurements, however, were those of the microwave absorption from gaseous ammonia (NH<sub>3</sub>) under simulated conditions for the Jovian atmospheres (in a 90% H<sub>2</sub>/10% He atmosphere at temperatures from 178 K to 300 K and pressures from 1 to 6 atm) over wavelengths from 1.3 to 22 cm. The results of these measurements are critical in that they confirm the theoretical calculation of the ammonia opacity using the Ben-Reuven lineshape. The application of both these

results, and results obtained previously, to planetary observations at microwave frequencies has been especially rewarding.

Applications of our results for ammonia to radio astronomical observations of Jupiter in the 1.3 to 20 cm wavelength range show that a strong depletion of ammonia for pressures less than 0.5 atm in the Jovian atmosphere must exist. They also show that a solar abundance of ammonia gas cannot account for the opacity inferred from the 10 to 20 cm emission spectrum in the 5 to 6 atm pressure range of the Jovian atmosphere, suggesting either an overabundance of NH<sub>3</sub> vapor, or the presence of another microwave absorber such as a dense cloud or gases such as  $H_2O$  which would accompany clouds at that altitude. We are proposing to further investigate the differences in the frequency dependence of opacity from gaseous  $H_2O$  and gaseous  $NH_3$  in the laboratory under Jovian conditions so as to better determine which of the two sources of opacity are present and in what relative quantities. (See accompanying proposal.)

Application of our results for gaseous  $H_2SO_4$  under simulated Venus conditions, obtained during the previous grant year, have shown that a 15 ppm abundance of gaseous sulfuric acid (on average) below the main cloud layer exists, and approximately 150 ppm of  $SO_2$  exists in the same region. Of more importance, our results show that certain portions of the Venus emission spectrum (especially around 13 GHz) are especially strongly affected by the sulfuric acid vapor abundance. Since no observations of the Venus emission at this wavelength have ever been published, we will use newly acquired data from NRAO in order to confirm the gaseous  $H_2SO_4$  abundance on a planet-wide basis. It is also important to recognize that due to instrumental limitations of the Venus atmospheric probes, no direct measurements of the abundance of gaseous

 $H_2SO_4$  exist for the subcloud region of Venus. However, Steffes (1985) showed that the major source of the microwave absorptivity measured by the Pioneer-Venus Radio Occultation experiments is gaseous  $H_2SO_4$ . Since the current radio occultation "season" is expected to yield the best measurements since the initial Pioneer-Venus radio measurements, we are hoping to become involved directly with the interpretation of that data in order to develop localized profiles for gaseous  $H_2SO_4$  abundance in the Venus atmosphere. This information would be of key importance in better modeling the Venus atmosphere.

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VIII. KEY FIGURES

Frequency, GH	Conditions	Refractivity N = 10 <sup>6</sup> (n-1)	Refractivity Normalized by Number Density (N-units/molecule/cm <sup>3</sup> )	Absorptivity (dB/km)	Source
2.255	$T = 295 \pm 1 K$ $P = 5 \pm .2 atm$	1354 ± 70	$(1.0886 \pm .104) \times 10^{-17}$	< .14	This work
2.255	$T = 295 \pm 3 K$ $P = 6.1 \pm 0.1 atm$	1665 ± 90	$(1.097 \pm .088) \times 10^{-17}$	< .27	This work
2.255	$T = 188 K \pm 10 K$ $P = 5.5 \pm .2 atm$	2156 ± 120	$(.9979 \pm .145) \times 10^{-17}$	< .05	This work
2.255	$T = 180 \pm 10 K$ P = 6.37 ± 0.1 atm	3015 ± 160	$(1.1609 \pm .083) \times 10^{-17}$	< .19	This work
2.255	T = 173 ± 10 K P = 6.34 ± 0.1 atm	2795 ± 150	$(1.039 \pm .128) \times 10^{-17}$	< .19	This work
2.255	T = 156 K + 12 K - 5 K	2509 ± 130	$(.9422 + .154) \times 10^{-17}$	< .1	This work
8.53	$P = 5.7 \pm .2 \text{ atm}$ $T = 208 \pm 10 \text{ K}$ $P = 5.85 \pm 0.1 \text{ atm}$	2269 ± 36	$(1.099 \pm .088) \times 10^{-17}$		This work
9.28	T = 273 K P = 1 atm	293.4 ± 1.4	$(1.0912 \pm .005) \times 10^{-17}$		Birnbaum, Kryder and Lyons (1951)
13.3	$T = 295 \pm 1 K$ P = 6 ± .2 atm	1475 ± 75	$(.988 \pm .086) \times 10^{-17}$	< 9	This work
21.7	$T = 295 \pm 1 K$ P = 5.7 ± .2 atm	1447.0 ± 75	$(1.059 \pm .096) \times 10^{-17}$	< 4.5	This work
24.0	T = 273 K P = 1 atm	294.1 ± 0.1	$(1.0938 \pm .0004) \times 10^{-17}$		Essen and Froome (1951)

.

# TABLE I

# LABORATORY MEASUREMENTS OF NITROGEN (N2) REFRACTIVITY AND ABSORPTIVITY





Figure 2: Sketch of pressure vessel, as viewed from above, with the top plate removed. Note the welded flange, with groove for the O-ring.



Figure 3 : Measured microwave absorption from ammonia (4.3 X  $10^{-3}$  by volume) in a 90% hydrogen/10% helium atmosphere as a function of frequency (wavelength) at 193 K. Also shown are the theoretically-derived absorption profiles.

Frequency, GHz

Figure 4 : Measured microwave absorption from ammonia (4.3  $\times 10^{-3}$  by volume) in a 90% hydrogen/10% helium atmosphere as a a function of frequency (wavelength) at 300 K. Also shown are the theoretically-derived absorption profiles.



Figure 5 : Measured microwave absorption from ammonia (4.3 X  $10^{-4}$  by volume) in a 90% hydrogen/10% helium atmosphere as a function of frequency (wavelength) <sup>36</sup> at 178 K. Also shown are the theoretically-derived absorption profiles.



Absorption, dB/Km

Frequency, GHz

# IX. APPENDICES

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Table 1: Absorption Summary for 2.25 GHz

Date	Temp.	Mixing Patio	Fress.	a (dB/Km)	a (dB/Km) Theoretical	Notor
	<u>_</u>			Tuggarter	TUGATERTERT.	
8-06-86	27	4.3 X 10 <sup>-3</sup>	6.1	0.91	0.73	measured
				1.18		maximum
			-	0.64		տinimum
			4.1	0.36	0.35	measured
				0.64		maximum
				0.09		minimum
			2.0	0.27	0.09	measured
				0.55		maximum
				0.00		minimum
7-25-86	80	4.3 X 10-3	6.1	1.73	2.01	measured
				2.00		maximum
				1.46		տուուտատ
			4.1	0.91	0.81	measured
				1.09		maximum
				0.73		minimum
			2.1	0.50	0.25	measured
				0.68		maximum
				0.32		minimum
8-13-86			6.1	2.10	2.01	measured
				2.37		maximum
				1.82		minimum
			4.1	0.82	0.81	measured
				1.05		maximum
				0.59		minimum
			2.0	0.37	0.25	measured
				0.55		maximum
				0.18		៣inimum
7-14-86	-95	4.3 X 104	6.1	0.18	0.24	measured
				0.46		maximum
				0.00		տiпiտստ
-			3.2	0.18	0.07	measured
				0.40		maximum
				0.05		minimum

Table 2: Absorption Summary for 8.53 GHz

Date 	Temp.	Mixing <u>Ratio</u>	Press. <u>atm.</u> _	a (dB/Km) _Measured_	a (dB/Km) <u>Theoretical</u>	<u>Notes</u>
8-06-86	27	4.3 X 10 <sup>-3</sup>	6.1	15.3 17.5 13.1	12.5	measured maximum minimum
			4.1	7.1 8.8 5.4	6.5	measured maximum minimum
			2.0	0.18 0.91 0.00	2.01	measured maximum minimum
7-25-86	-80	4.3 X 10 <sup>-3</sup>	6.1	29.5 32.2 26.7	30.3	measured maximum minimum
			4.1	8.5 9.9 7.2	14.1	measured maximum minimum
			2.1	2.2 3.1 1.3	4.9	measured maximum minimum
8-13-86			6.1	49.5 54.4 44.6	30.3	measured maximum minimum
			4.1	16.7 18.5 15.0	14.1	measured maximum minimum
			2.0	3.3 4.4 2.2	4.9	measured maximum minimum
7-14-86	-95	4.3 X 10 <sup>-4</sup>	<u></u> 6.1	0.55 0.91 0.18	3.74	measured maximum minimum
			3.2	0.18 0.54 0.00	1.17	measured maximum minimum

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Table 3: Absorption Summary for 13.3 GHz

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Date	Temp.	Mixing <u>Ratio</u>	Press. <u>atm</u>	a (dB/Km) _ <u>Measured</u>	α (dB/Km) <u>Theoretical</u>	<u>Notes</u>
8-06-86	27	4.3 X 10 <sup>-3</sup>	6.1	4.5 11.8 0.0	31.1	measured maximum minimum
			4.1	21.8 29.1 14.5	17.4	measured maximum minimum
			2.0	0.0 7.3 0.0	6.1	measured maximum minimum
7-25-86	-80	4.3 X 10 <sup>-3</sup>	6.1	46.4 68.9 23.8	71.8	measured maximum minimum
			4.1	18.2 25.4 10.9	34.1	measured maximum minimum
			2.1	0.0	14.3	measured maximum minimum
7-14-86	-75	4.3 X 10 <sup>-4</sup>	<i>٤.</i> 1	9.11 16.38 1.83	8.94	measured maximum minimum
			3.2	7.29 12.75 1.83	3.05	measured maximum minimum

Table 4: Absorption Summary for 21.7 GHz

Date	Temp. C	Mixing Ratio	Press. atm.	a (dB/Km) Measured	α (dB/Km) Theoretical	Notes
						<b>_</b> _
8-06-86	27	4.3 X 10 <sup>-3</sup>	6.1	132	85	measured
				150		max1mum
				114		ጠነበነጠረጠ
			4.1	59.2	65.9	measured
				77.4		maximum
				41.0		տiпiտպտ
			2.0	59.2	41.7	measured
				77.4		maximum
				41.0		minimum
			1.0	36.4	26.8	measured
				63.7		maximum
				9.1		minimum
7-25-86	-80	4.3 X 10-3	6.1	54.8	182.0	measured
				91.1		maximum
				18.4		minimum
			4.1	54.7	144.0	measured
				86.5		ma≺imum
				22.9		minimum
			2.1	54.6	89.2	measured
				84.5		maximum
				22.8		minimum
8-13-86			6.1	187	182	measured
	•			207		maximum
				166		minimum
			4.1	137	144	measured
				164		maximum
				109		minimum
			2.0	114	89	measured
				136		maximum
				91		minimum
7-14-86	-95	4.3 X 10 <sup>-4</sup>	6.1	54.8	20.7	measured
				95.7		maximum
				13.8		minimum
			3.2	0.0	14.0	measured
				22.8		maximum
				0.0		៣ពាំពាលពា

Laboratory Measurements of the Microwave Opacity of Gaseous Ammonia (NH<sub>3</sub>) under Simulated Conditions for Jovian Atmospheres

P. G. Steffes, J. M. Jenkins, M. F. Selman, W. W. Gregory (Georgia Institute of Technology)

We have recently completed laboratory measurements of the microwave opacity of gaseous ammonia (NH<sub>3</sub>) under simulated conditions for Jovian atmospheres (i.e., as a minor constituent in a 90%  $H_2/10$ % He atmosphere, with temperatures ranging from 170 K to 300 K, and total atmospheric pressures ranging from 1 to 6 atmospheres). While the shapes of the absorption spectra, observed at wavelengths from 1.3 cm to 22 cm, seem consistent with theoretically derived expressions for the microwave opacity from NH<sub>3</sub> obtained using the Ben-Reuven line shape, some small variations from the theoretical absorption spectra may exist. These measurements are important for the interpretation of radio absorptivity data obtained from both radio astronomical and radio occultation observations of the outer planets. They are especially important for use in determining the abundance and distribution of NH<sub>3</sub> in the atmosphere of Jupiter.

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American Astronomical Society - Paris DPS meeting Date Submitted **PROBING VENUS** 

Remote sensing technology gives researchers detailed new data about the Venusian atmosphere

emote sensing from space has given the human race a new perspective on itself. With vivid satellite portraits of the earth, planners can see pollution in coastal marshlands. Insect infestation, fishing beds and a wealth of other impor-sant information. Now, the same ball testimology is betting scientists map the atmospheres of planets millions of miles away, At Georgia Tech, Dr. Paul Stelles, an assistant professor of electrical engineering, has used remote sensing to "see" the makeup and structure of sulfuric acid in the midatmosphere of Venus.

The raw data for his research were microwave signals transmitted by Mariner and Pioneer spacecraft as they flew behind Venus. Before returning to earth, these transmissions penetrated the Venusian atmosphere, where they were either bent (refracted) or weakened (attenuated). The resulting signal modifications were collected by the Deep Space Network of antennas at stations in California, Spain and Australia.

"Most of the refraction is due to the density of the atmosphere," says Steffes. "It's like a light wave being bent as it passes through the ocean."

Weakened signals indicate an absorption of electromagnetic energy by the atmosphere. Since only certain gases absorb electromagnetic waves, this information becomes a key for accomplishing this on a parts-



Remote sensing technology used by a Georgia Tech professor is helping uncover new knowledge about the planet Venus.

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per-million level through a laboratory simulation of Venus mid-atmosphere. With funding from NASA, he and his students built a small pressure chamber. They first injected gases known to be in the planet's atmosphere and subjected them to pressures and temperatures which exist at certain altitudes above Venus. Next, they propagated signals through the chamber at the same frequencies originally transmitted by NASA spacecraft back to earth. Finally, they manipulated the concentration of gases present until they were abls to bring about the same degree of signal attenuation or refraction observed through remote sens ing. Through these experi-ments, Stelfes' group found that they could calculate con centrations of a microwaveabsorbing vapor, sulfuric acid, in the mid-atmosphere of Venus.

The accuracy of this method has proven greater than achieved by probes dropped into the Venusian atmosphere by Américan spacecraft earlier exploring the planet. Simulations using remote sensing data have resolved sulfuric acid vapor abundances down to an order of 10 parts-per-million. The picture revealed by these experiments has been somewhat surprising. "Approximately 20 parts-permillion of sulfuric acid vapor exist below the clouds of Venus," Steffes says. "In the altitude range of 30 to 48 kilometers there is sulfuric acid vapor which condenses at 48 kilometers. Initially, it wasn't thought that that amount of sulfuric acid might have existed. It was thought that sulfuric acid clouds existed but that immediately below the clouds only sulfur dioxide existed." Steffes' current work grew



prosente chamber developed by Br. Bull Steffer umulaters atmagherte conditions on Venus and the

outer planets. Voyagers landli

already have passed jupiter and

Saturn, Last January, Voyager II

started sending data back from

Uranus. In another three years,

the ship will make its approach

posed primarily of hydrogen

to Neptune and its moon

Triton

out of remote sensing experiments he made in the early 1980s as a doctoral degree student at Stanford University. However, his interest in the field began during his undergraduate years at the Massachusetts institute of Technology. There, remote sensing techniques were being used to learn about the abundance of certain gases in the upper part of the earth's atmosphere.

"At MIT, we were able to infer how much oxygen and water vapor was in the earth's atmosphere," Steffes says, "so we said: if it works for us here we can apply it to other planets too."

The implications of his findings for Venus are uncertain, but Steffessays that they could provide evidence for volcanic activity on the planet. He also believes that knowledge of how sulfuric acid moves in the atmosphere of Venus possibly can help scientists better understand the acid rain problem on earth. With the Venus simulations and helium, gases which do completed. MASA is sponsoring Stelfes in a program to interrorer remote sensing data on in his experiments, Stelfer

In his experiments, Steffes will full the deep freeze with these gases, then start adding other gases, such as ammonia and methane, trying to match the attenuation and refraction of the Voyager microwave data. The work on Jupiter, Saturn and Uranus will probably

Append

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To simulate the atmospheres take a year and a half. of these planets, Steffes has What will be the value of his findings? "We'll be learning a acquired a deep freeze which canreach 120°C., well above lot about the nature of the forthe -190° C. atmospheric mation of planets," Steffes says. temperature of Uranus but "That tells us in the long run close enough for accurate about how the universe is extrapolations of data. formed and whether or not The outer planets will prethere may be planets around sent an entirely different other stars. So the applications

challenge than Venus. Accordare long term, obviously. They are it necessarily something where we're going to be able to never quite made it'' — that is, large gaseous balls with relatively small rocky cores. Their atmospheres are very highly pressurized and com-

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Tuesday, June 10, 1986

THE ATLANTA CONSTITUTION

# Page 34-A

# Tech making down-to-Earth studies of outer space

#### By Robert Lee Hotz Science/Medicine Writer

Inside a squat, square freezer at the Georgia Institute of Technology, scientists are seeking clues to the frigid gases that envelop the solar system's outer planets in a project that may yield new insights into the evolution of Earth's atmosphere.

By interpreting subtle distortions in radio signais from the Voyager 2 space probe, Georgia Tech researchers are mapping the exotic atmospheres of Jupiter, Saturn, Uranus and Titan, Saturn's largest moon, then duplicating the volatile blend of gases in a pressurized laboratory freezer.

. Analyzing microwave signals from unmanned Mariner and Pioneer probes, they have detected corrosive clouds of sulfuric acid that dot the skies of Venus. Using large Earth-based radiotelescopes, they hope one day to track the daily weather on Venus or detect any active volcances.

"We are doing this research not only so we could better understand these other planets, but also to understand how our own atmosphere might be evolving," says Dr. Paul Steffes. a Georgia Tech professor of electrical engineering who leads the research team.

"We are learning how fragile our own atmospheric balance is — because if the scales tip one way, we could end up very hot like Venus or, if the scales tip the other way, we could end up like Mars. It is a very delicate balance that keeps our atmosphere so amenable to jife."

None of the probes carries instruments designed to detect atmospheric gases. But when a satellite passes behind a planet or moon, the radio signals being transmitted back to Earth are garbled briefly by interference from the gases surrounding the other planet. Steffes has turned that distortion into a tool for mapping the alien . atmosphere itself.

"Using the U.S. Deep Space Network, we are able to measure the amount of weakening in the microwave signal as it passed through the planet's atmosphere," Steffes says. "The amount of microwave radiation absorbed or refracted depends on what gases are in the planet's atmosphere. The question for us is what gases and how much," he says.

Any weakening of the satellite's signal indicates the presence of chemical compounds that absorb microwave energy. By pinpointing the frequency blocked, researchers can decipher the chemical signature of gases such as belium, hydrogen and sulfuric acid. In addition, any bending of the microwaves is an effective barometer of the atmospheric pressure, Steffes says.

"Sulfuric acid vapor, for instance, has a very unique frequency signature." Steffes says. "Using the Pioneer orbiter, we could resolve the amount of sulfuric acid vapor to an accuracy better than 10 parts per million.

"Sulfuric acid vapor is like water in the Venus atmosphere. The clouds are of sulfuric acid vapor," he says.

Like house painters trying to match the proper pastel hue. Steffes and his assistants double-check their interpretations of the radio databy mixing gas samples in a stainless steel pressure chamber and bombarding it with microwaves. They continue to adjust the mix until the microwave distortion matches that actually measured from the orbiting spacecraft.

To simulate the furnace-like surface temperatures of Venus, they cooked the laboratory pressure chamber in an oven at 600 degrees. Now, to simulate the frigid temperatures of the outer planets, Steffes is using a special freezer that can lower the chamber temperature to less than 125 degrees below zero.



RON CODDINGTON/Staff



# GEORGIA INSTITUTE OF TECHNOLOGY SCHOOL OF ELECTRICAL ENGINEERING ATLANTA, GEORGIA 30332

HONE: (404) 894- 3128

July 24, 1986

Congressman Wyche Fowler,Jr. 1210 Longworth House Office Building Washington, DC 20515

Dear Congressman Fowler:

As a strong supporter of the Space Sciences and Solar System Exploration over the past years, I thought I would take this opportunity to alert you to our concerns regarding the current appropriations debate regarding the NASA budget. As was the case with the FY 1985 budget, when I previously contacted you (March 7, 1984), attempts are currently being made to substantially cut the Research and Analysis Program within the planetary exploration budget. These attempts are probably related to the "crowding out" effect which you referred to in your March 9, 1984 letter to Congressman Michael A. Andrews, when you expressed a fear that programs like the Space Station might place such strong funding constraints on the agency that the small, potentially vulnerable, yet highly valuable, space science programs might disappear. A large portion of such funding is provided to university scientists to support research at the highest levels of excellence.

While we all are aware of the national requirements for budget restraint, the future of U.S. leadership in this field, which you have characterized as "prestigious and technology-expanding", lies in the elements of basic research and analysis, which involve students and faculty at American universities. Your support of such research in the past has been greatly appreciated, as has been your support of such issues as "peer review". We hope that it will continue both now, and in your future career, be it in the House or the Senate.

> Sincerely, Paul G. Steffes Assistant Professor



GREEN BANK OBSERVING APPLICATION COVER SHEET

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Received:

SEND TO: Director NRAO Edgemont Road, Charlottesville, Va. 22901

information (supporting documents are for refereeing only).

Date: April 29, 1986

Title of Proposal: Observations of the Venus Microwave Emission at 1.9 and 2.25 cm.

Anathors	Institution	Who Will Observe?	Grad Student?	Observations for PhD Thesis?	Anticipated PhD Year
I G. Steffes	School of Electrical Engr. Georgia Institute of Tech.	Steffeså Gurski	S. (no) G. (yes)	possible	1990
Contact Author for Scheduling: Paul G.	Steffes 5	Telephon or TW	e: (404) X:	) 894-3128	
Address: School ( Georgia Atlanta F <del>elescope</del> (check one):	of Electrical Engineering Institute of Technology GA30332-025- 300'				
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Receiver: (please consul Cassegrain - Two Receiver Frequency (or range of fr I3.3 GHz and 15.8 GHz (or ressions/Days Requested: 4-6 bstract (do not write ou ervations of the planet 25 cm and 1.9cm) are prop the continuum emission s isphere. The measurement t wapowr abundance in the properly modelling and	t Front End Box Status Sheet rs (2 cm - upconverting Maser requencies) (include test lin ne channel for each) (12) LST Ran Local tside this space): Venus (and reference sources) posed in order to confirm the pectrum at 2.25 cm due to sul t would also provide a planet sub-cloud region of the Venu understanding the Venus atmos	): r) es): Transit ) at 13.3 e presence lfuric ac t-wide av us atmosp sphere.	±2 hours GHz and e of a pu id vapor erage of here, a c	15.8 GHz redicted "di in the Venu the sulfuri critical fac	p" Is C Stor
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After your proposal is	scheduled, the contents of	this cove	r sheet	become publ:	ic

# Observations of the Venus Microwave Brightness at 1.9 and 2.25 cm.

# Proposed by

# Paul G. Steffes, Assistant Professor School of Electrical Engineering Georgia Institute of Technology Atlanta, GA 30332-0250 (404) 894-3128

As viewed from Earth, the planet Venus is the third strongest radio source in the sky, behind the sun and the moon. As a result, it is often used as a test target for new microwave antennas. It is surprising, however, that a large amount of uncertainty still remains as to the nature of the Venus microwave emission spectrum between 1 and 22 cm. Even more surprising is the fact that no results of observations of the microwave spectrum between 2.07 and 3 cm have ever been published.

As part of a NASA-sponsored program for making laboratory measurements of the microwave properties of simulated planetary atmospheres, it has been found that the microwave absorption from gaseous sulfuric acid exhibits a peak at the 2.25 cm wavelength. This unexpected absorption behavior from a key constituent in the Venus atmosphere is quite surprising in that previously it had been believed that all microwave absorbers would exhibit absorption with a frequency-squared dependence under the high atmospheric pressures which characterize the Venus atmosphere. (For further discussion, see Steffes, 1986.) As a result, we have developed a model for the microwave emission spectrum of Venus (see attached figure), using our newly-obtained laboratory results for gaseous  $H_2SO_4$ , and previous laboratory results for  $SO_2$  and  $CO_2$ , which provides a better fit to the ensemble of Venus microwave observations than any model yet proposed. Inspection of the model shown in Figure 1 shows a very good correlation with previously observed brightness temperatures in the 1 to 10 cm wavelength range, when a sulfuric acid vapor abundance of 15 ppm is assumed.

The most interesting aspect of our model for the Venus microwave emission spectrum lies in the 2 to 3 cm wavelength range. As can be seen from Figure 1, a noticeable dip in brightness temperature should exist between 2 and 3 cm. Unfortunately, few measurements of the Venus microwave emission have been made in this wavelength range. Two measurements have been made of the emission near 2 cm. The first, by McCullough and Boland (1964), was 500  $\pm$  75 K at a wavelength of 2.07 cm. The second, by Pollack and Morrision (1970), was 495  $\pm$  35 K at the 1.94 cm wavelength. Both are in good agreement with the model, assuming a gaseous H<sub>2</sub>SO<sub>4</sub> abundance of 15 ppm. The most interesting wavelengths to observe would be in the 2.2 to 2.3 cm range, where a strong dip in the microwave emission, related to the abundance of gaseous H<sub>2</sub>SO, should occur. Unfortunately, no observations have been published for these wavelengths. Since a high sensitivity receiver system capable of operation in this wavelength range current exists at NRAO, we are proposing such an observation. The proposed observation would require the use of the NRAO 140-foot telescope, using the "Cassegrain, two-receiver" configuration. The two receivers used would be the 12.0-16.2 GHz upconverter/maser systems which are currently being completed. We propose to observe simultaneously at two frequencies: 13.3 GHz (2.25 cm) and 15.8 GHz (1.9 cm) in order to confirm the existence of the predicted "dip" in the continuum spectrum by observing the "differential" emission at the two frequencies, and would then attempt to make measurements of the absolute brightness temperatures by comparison with between 5 to 10 references sources in the area. In order to minimize difficulties with beam size correction and source size correction, we propose to observe Venus when it is fairly close to superior conjunction, yet still clear of the sun (either May/June 1987 or November/December 1987). Similarly, we hope to use weaker point sources as our Venus calibrators in order to minimize any beam-size correction difficulties. We propose to observe over a 4 to 6 day period with the daily observations of Venus and the references consuming approximately 4 to 6 hours.

The successful completion of the proposed observation will have several benefits:

- 1. Two high-accuracy data points will be added to our current knowledge of the Venus continuum microwave spectrum.
- It will either confirm or refute the new model for the Venus microwave emission developed by Steffes (1986) (also, see Figure 1) in which sulfuric acid vapor and CO<sub>2</sub> are most responsible for the spectral shape at wavelengths longward of 1.8 cm.
- 3. Assuming the laboratory-based model by Steffes (1986) is confirmed, the resulting 2.25 cm brightness measurement should provide a planet-wide average of the sulfuric acid vapor abundance in the sub-cloud region of the Venus atmosphere, a critical factor for properly understanding and modelling the Venus atmosphere. It should likewise be noted that because of the instrumental limitations of <u>in situ</u> probes, this may become the major technique for determing gaseous  $H_2SO_4$  abundance in the Venus atmosphere.

# Reference:

Steffes, P. G. (1986). Evaluation of the Microwave Spectrum of Venus in the 1.2 to 22 cm Wavelength Range Based on Laboratory Measurements of Constituent Opacities. <u>Astrophysical Journal</u>, in press.

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Figure 1: Existing measurements of the microwave emission from Venus. The solid line represents the model when a 15 ppm abundance of gaseous H<sub>2</sub>SO<sub>4</sub> is assumed below the main cloud layer. The dashed line represents the model when a 30 ppm abundance is assumed. Both models assume abundances for CO<sub>2</sub>, H<sub>2</sub>O, and SO<sub>2</sub> of 96 percent, 100 ppm, and 150 ppm, repectively.

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#### EVALUATION OF THE MICROWAVE SPECTRUM OF VENUS IN THE 1.2-22 CENTIMETER WAVELENGTH RANGE BASED ON LABORATORY MEASUREMENTS OF CONSTITUENT GAS OPACITIES

PAUL G. STEFFES

School of Electrical Engineering, Georgia Institute of Technology Received 1986 February 10; accepted 1986 April 16

#### ABSTRACT

The microwave spectrum of Venus is reviewed in light of new laboratory measurements of the microwave-absorbing properties of its gaseous atmospheric constituents under simulated conditions for the Venus atmosphere. In particular, our laboratory measurements in the wavelength range 1.2–22.3 cm show that gaseous  $H_2SO_4$  and  $CO_2$  are the predominant microwave absorbers at wavelengths longer than 1.8 cm (frequencies below 17 GHz), and that SO<sub>2</sub> and CO<sub>2</sub> are the predominant absorbers at wavelengths from 1.2 to 1.8 cm. As a result, it is concluded that measurements of Venus emission/opacity at longer wavelengths (frequencies below 18 GHz) are only loosely correlated with measurements at shorter wavelengths. We also use these measurements in the development of a new model for the Venus microwave emission spectrum. The new model correlates quite well with the ensemble of microwave observations of Venus when abundances of  $H_2O$ ,  $SO_2$ , and gaseous  $H_2SO_4$  are assumed to be 100 ppm, 150 ppm, and 15 ppm, respectively. We also discuss how variations in SO<sub>2</sub> abundance from 80 ppm to 200 ppm, and variations in gaseous H<sub>2</sub>SO<sub>4</sub> abundance from 15 ppm to 30 ppm can explain the observed variations in 1.35 cm and 13 cm opacity, respectively. We conclude by suggesting additional observations of Venus, especially in the 2-3 cm wavelength range, in order to better evaluate the role played by gaseous sulfuric acid in the Venus atmosphere.

Subject headings: planets: abundances - planets: atmospheres - planets: radio radiation - planets: Venus

#### L INTRODUCTION

The microwave emission spectrum of Venus in the 1.2-22 cm wavelength range has been measured and studied for nearly 30 yr. A number of works (e.g., Barrett and Staelin 1965; Muhleman, Orton, and Berge 1979; Kuz'min 1983) have summarized observations and have developed model spectra based on such observations. The microwave opacity profiles which have been inferred from these models have likewise been studied carefully in order to set limits on the abundances of microwave absorbing constituents (see, for example, Ho, Kaufman, and Thaddeus 1966 or Janssen and Klein 1981). The accuracy of such abundance estimates for microwave absorbing constituents depends critically on the availability of reliable information regarding the microwave absorbing properties of potential constituents under the conditions of the atmosphere being studied. To this end, laboratory measurements were made of the microwave absorption from potential constituents under simulated conditions for the middle atmosphere of Venus (pressures up to 6 atm): from SO2 and SO3 (Steffes and Eshleman 1981), and from gaseous H<sub>2</sub>SO<sub>4</sub> (Steffes and Eshleman 1982 and Steffes 1985). These measurements were conducted at the 3.6 cm and 13.4 cm wavelengths in order to be directly applicable to opacity profiles obtained from radio occultation measurements. Previous to these measurements, it was assumed that the frequency dependence of the opacity from microwave-absorbing constituents, under the pressure conditions of the subcloud region of the Venus atmosphere, was simply  $f^2$ . (Most profiles of the radio opacity at Venus, derived from either radio occultation or radio astronomical observations, place the bulk of the opacity at altitudes below the cloud base altitude, about 48 km.) Such an assumption would preclude the need for laboratory measurements of absorption properties over a wide spectral range. While this

Note Corrections: Page 408 (Splaces) Page 411 (1 place) Page 413 (1 place)

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assumption seemed to be correct for SO2 in a simulated Venus atmosphere (see Steffes and Eshleman 1981), it was shown not to be valid for gaseous H2SO4 in a simulated Venus atmosphere at wavelengths between 13.4 and 3.6 cm (Steffes 1985). As a result, we have performed further measurements of the microwave absorption spectrum of gaseous H2SO4 in a CO, atmosphere at selected frequencies between 1.34 GHz (22.3 cm wavelength) and 23.6 GHz (1.2 cm wavelength) under simulated conditions for the middle atmosphere of Venus (total pressures from 1 to 6 atmospheres). The results of these measurements significantly change the ways in which the Venus microwave emission spectrum is interpreted. We have used these results to develop a model for the microwave emission spectrum of Venus, which correlates quite well with the ensemble of microwave observations of Venus. We conclude by suggesting further observations and analysis of the Venus microwave emission spectrum, especially in the less frequently observed 2-3 cm wavelength range.

#### II. EXPERIMENTAL APPROACH

The approach used to measure the microwave absorptivity of gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere is similar to that used previously by Steffes (1985). As can be seen in Figure I, the absorptivity is measured by observing the effects of the introduced gas mixture on the Q, or quality factor, of two cavity resonators at particular resonances from 1.34 GHz to 23.6 GHz. The changes in the Q of the resonances which are induced by the introduction of an absorbing gas mixture can be monitored by the high-resolution microwave spectrum analyzer, since Q is simply the ratio of the cavity resonant frequency to its half-power bandwidth. For relatively low loss gas mixtures, the relation between the absorptivity of the gas mixture and its effect on the Q of a resonance is straightforward:

$$\alpha \approx (Q_L^{-1} - Q_C^{-1})\pi/\lambda, \qquad (1)$$

where  $\alpha$  is absorptivity of the gas mixture in nepers km<sup>-1</sup>. (Note, for example, that an attenuation constant or absorption coefficient or absorptivity of 1 neper km<sup>-1</sup> = 2 optical depths per km [or km<sup>-1</sup>] = 8.686 dB km<sup>-1</sup>, where the first notation is the natural form used in electrical engineering, the second is the usual form in physics and astronomy, and the third is the common [logarithmic] form. The third form is often used in order to avoid a possible factor-of-two ambiguity in meaning.)  $Q_L$  is the quality factor of the cavity resonator when the **gas** mixture is present,  $Q_C$  is the quality factor of the cavity resonator in a vacuum, and  $\lambda$  is the wavelength (in km) of the test signal in the gas mixture.

In order to obtain a gas mixture with a sufficient amount of H<sub>2</sub>SO<sub>4</sub> vapor so that the microwave absorption is detectable, the system must be operated at temperatures exceeding 450 K. While this is suboptimal in that the temperatures at altitudes from 35 to 50 km (where both radio occultation and radio astronomical experiments have detected microwave opacity) range from 350 to 450 K, temperature dependencies measured for similar gases (such as SO2) can be used to estimate temperature effects in that range. Two different approaches are used to infer H2SO4 vapor pressure. With the first, the volume of liquid sulfuric acid which is vaporized to generate the gaseous H<sub>2</sub>SO<sub>4</sub> is determined to a high accuracy (up to  $\pm 0.005$  ml). It is then possible to compute an upper limit for the partial pressure of gaseous H2SO4 using the ideal gas equation, the measured change in liquid volume, and published densities for  $H_1SO_4$  liquid. The second method for accurately determining amounts of liquid-derived vapor measures the refractivity of those vapors. Since the index of

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refraction (relative to unity) is proportional to the vapor abundance, the system's ability to accurately measure the refractivity of such vapors can also be used to infer the relative vapor abundance and/or pressure. Note that it is not yet possible to use this approach for accurate determination of the absolute vapor pressure from gaseous  $H_2SO_4$ , since accurate refractivity data for gaseous  $H_2SO_4$  is not currently available. While neither method is able to accurately resolve dissociation of  $H_2SO_4$  into  $H_2O$  and  $SO_3$ , an upper limit of  $H_2SO_4$  vapor abundance can be inferred, using the first method, which is accurate to  $\pm 2\%$  at a temperature of 500 K.

As shown in Figure 1, a flask is filled with a precisely known volume of liquid sulfuric acid (99% by weight) at room temperature. For nearly all of the experiments, the initial volume used was 2.5 ml (measured at room temperature). Smaller quantities were also tried, with no significant difference in results. The volume measurements are made using a 1 ml syringe with 0.01 ml gradiations. For volumes less than 1 ml, repeatable accuracies of better than 0.005 ml have been obtained. The entire system is then heated and allowed to thermally stabilize at the chosen experimental temperature (approximately 575 K), which requires approximately 6 hr. The temperature is monitored using a thermocouple which is placed in the center of the pressure vessel, near the microwave resonator, which is one of the coldest places in the system. Thermal equilibrium within the system is achieved by preheating the system for 6-8 hr before beginning the experiments. Since the thermal time constant is approximately 2 hr 15 minutes, this assures a relatively constant temperature within the chamber after heating. Another technique used in monitoring the heating of the resonator within the chamber is to monitor the resonant frequencies of the resonator. As the resonator is heated, the resonant frequenceis drop due to thermal expansion. Thus, when thermal stability is reached, the resonant frequencies likewise stabilize. After thermal stability is reached, a vacuum is drawn in the pressure vessel containing the microwave cavity resonator, and the bandwidth and center frequency of the resonances are then measured. For this experiment, resonances at 1.34 GHz (22.3 cm), 2.24 GHz (13.4 cm), 8.42 GHz (3.6 cm), 13.23 GHz (2.26 cm), 21.63 GHz (1.38 cm), and 23.64 GHz (1.27 cm) were used. A valve is then opened which allows the sulfuric acid vapor eluting from the flask to fill the pressure vessel (0.031 m<sup>3</sup> of open volume with resonator in place) and reach vapor pressure equilibrium with the liquid H<sub>2</sub>SO<sub>4</sub>. Note that all components which contact the gaseous sulfuric acid are maintained at the same temperature as the flask, so as to avoid condensation. In addition, because of the high temperatures and corrosive vapor involved, all tubing, valves, and the pressure chamber itself are fabricated from stainless steel. Gaskets and cables are fabricated from either viton or PTFE. These steps not only ensure the survival of components under the test conditions but also reduce possible reactions of the components with the sulfuric acid vapor and possible outgassing of vapors related to the materials from which the components are fabricated.

As  $H_2SO_4$  vapor fills the chamber, changes in the resonance center frequency are observed. These changes, which reach over 400 kHz at the 13.4 cm resonance, are related to the  $H_2SO_4$  vapor abundance. After approximately 10 minutes, the frequency shift ceases, as vapor pressure equilibrium is reached. The valve to the reservoir flask is then closed, and  $CO_2$  is admitted to the chamber containing the  $H_2SO_4$  vapor. For this experiment, a total pressure of 6 atm was used. The  $CO_2$  gas is admitted to the chamber at a sufficiently slow rate so as not to significantly affect the temperature within the chamber. The bandwidth of each response is then measured 406

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and compared with its value when the chamber was evacuated in order to determine the absorptivity of the CO<sub>2</sub>/H<sub>2</sub>SO<sub>4</sub> gas mixture at 6 atm total pressure. The total pressure is then reduced by venting, and the bandwidths are again measured. Subsequent measurements are likewise made at lower pressures in order to determine absorptivities at those pressures. The pressure vessel is then evacuated and the bandwidths again measured so as to assure no variation (either due to thermal shift or chemical reaction) of the Q's of the evacuated resonators has occurred. After the system has been allowed to cool, the volume of the remaining sulfuric acid liquid (at room temperature) is measured and compared with the initial volume measured (at room temperature) in order to set an upper limit on H<sub>2</sub>SO<sub>4</sub> vapor present in the gas mixture tested. This approach has the advantage that the same gas mixture is used for the absorptivity measurements at the various pressures. Thus, even though some uncertainty may exist as to the mixing ratio of the initial mixture, the mixing ratios at all pressures are the same, and thus the uncertainty for any derived pressure dependence is due only to the accuracy limits of the absorptivity measurements, and not to uncertainty in the mixing ratio. (This assumes that the mixing ratio is small, so that foreign-gas broadening predominates, as is the case for our measurements.) Similarly, measurements of the frequency dependence of the absorptivity from a CO<sub>2</sub>/H<sub>2</sub>SO<sub>4</sub> mixture are likewise immune to mixing ratio uncertainty, as long as foreign-gas broadening predominates.

Since the overall equipment configuration and experimental approach for these measurements is similar to that used by Steffes (1985) for measurement of the microwave (3.6 cm and 13.4 cm) absorptivity and vapor pressure behavior of gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere, it is not surprising that the results from these experiments for the 3.6 cm and 13.4 cm opacity, and for the vapor pressure from  $H_2SO_4$ , agree quite closely with those previously obtained. However, the extended spectral coverage of these measurements (1.2-22.3 cm) presents much needed results, especially for wavelengths shortward of 3 cm. In fact, the results indicate that simple extrapolation of longer wavelength results can lead to significant errors, even with total pressure as high as 6 atm.

#### III. MEASUREMENT RESULTS

Measurements of the microwave absorption from gaseous H<sub>2</sub>SO<sub>4</sub> in a CO<sub>2</sub> atmosphere were conducted at a temperature of 575 K. This resulted in the evaporation of a volume of 99.0% concentration (by weight) liquid sulfuric acid which corresponded to an  $H_2SO_4$  vapor pressure of  $2.2 \times 10^{-2}$  atmospheres in our pressure vessel. (We assumed that 47% of the vaporized H<sub>2</sub>SO<sub>4</sub> dissociated to form SO<sub>3</sub> and H<sub>2</sub>O, as computed by Gmitro and Vermeulen 1964 for 99% solutions.) This value for H<sub>2</sub>SO<sub>4</sub> vapor pressure agreed quite closely with the value obtained in earlier measurements (see Steffes 1985). Since the same earlier measurements indicated that microwave opacity from gaseous H<sub>2</sub>SO<sub>4</sub> in a CO<sub>2</sub> atmosphere increased with frequency, we made our initial high-frequency measurements of absorptivity at wavelengths around 1.3 cm, expecting large opacities. Much to our surprise, we were unable to detect opacity in this wavelength range from the  $H_2SO_4/CO_2$  mixture. The experiment was repeated several times, with equipment being rechecked, but the results were still the same. Since the refractivity of the gaseous H<sub>2</sub>SO<sub>4</sub> and CO<sub>2</sub> were easily measured at all wavelengths as they entered the pressure chamber, it was clear that the gases were indeed present in the quantities expected. In fact, the absorptivity measured at the 13.4 cm and 3.6 cm wavelengths agreed quite well with previous measurements by Steffes (1985). It was

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therefore clear that even with total pressures as high as 6 atm, that mixtures of gaseous H2SO4 and CO2 had a unique microwave absorption spectrum which differed significantly from a simple  $f^2$  dependence. It should also be noted that the maximum value of the collision-induced absorption from pure  $CO_2$  as determined by Ho, Kaufman, and Thaddeus (1966) could not exceed 0.03 dB km<sup>-1</sup> under the conditions of these experiments. Since this is far below the threshold sensitivity of our system, it had no affect on the measurements.

Our measured results for the microwave absorption from gaseous  $H_2SO_4$  in a CO<sub>2</sub> atmosphere at 575 K are shown in Figure 2. The data points presented are for an  $H_2$  mixing ratio of 0.4% and were obtained for total pressures of 6 atm and 1 atm. Best-fit curves for the absorption spectra from 1.3 GHz (22.5 cm) to 25 GHz (1.2 cm) at pressures of 6 atm and 1 atm are also shown. Inspection of these results reveals three major results:

1. The first major result is the extremely low absorptivity observed in the 1.2-1.8 cm wavelength range. For example, measurements made at 21.63 GHz (1.38 cm wavelength) show an opacity for 0.4% mixture of gaseous H<sub>2</sub>SO<sub>4</sub> in a CO<sub>2</sub> atmosphere (with a total pressure of 6 atm and temperature of 575 K) of less than 9 dB km<sup>-1</sup>. This is far below the opacity predicted by using previous measurements at 2.2 GHz and 8.4 GHz and assuming a simple  $f^2$  dependence such as exhibited by SO<sub>2</sub> and CO<sub>2</sub>. In fact, this even implies that using the measured frequency dependencies for the microwave absorption from H<sub>2</sub>SO<sub>4</sub> in a CO<sub>2</sub> atmosphere in the 2.2-8.4 GHz range to predict absorption at 21.6 GHz (1.38 cm wavelength) would result in overstating the absorption at 1.38 cm by at least a factor of 2.

Such behavior implies that the contribution of gaseous H<sub>2</sub>CO/to the overall non-CO<sub>2</sub> opacity inferred from radio  $= \frac{1}{2} \underbrace{504}$ astronomical observations at the 1.35 cm wavelength is minimal. Such a result is not surprising in light of the calculations done by Janssen and Klein (1981) which attribute nearly all of the non-CO2 opacity at 1.35 cm in the Venus atmosphere to  $SO_2$ . This behavior is likewise consistent with the calculated  $H_2SO_4$  resonance frequencies computed by Poynter (R. Poynter, personal communication. Note that the results of the resonance calculations are shown in Cimino 1982.). These calculations are based on rotational constants computed using measurements of H<sub>2</sub>SO<sub>4</sub> resonances in the 60-120 GHz range by Kuczkowski, Suenram, and Lovas (1981). It is significant that there is a notable absence of  $H_2SO_4$  resonances in the 1560 GHz frequency range, which would be  $\leftarrow$ consistent with our measurements.

2. The second major result is the discovery of a peak in the absorptivity from gaseous H2SO4 at a wavelength of approximately 2.2 cm, even at pressures as high as 6 atm. This does a lot to explain why observations of the Venus brightness temperature at 2 cm (495 K-from Pollack and Morrison 1970), which were initially felt to be inconsistent with measurements at 1.35 cm (474 K-from Muhleman, Orton, and Berge 1979), can indeed be consistent, since there is at least one absorber whose opacity decreases with increasing frequency, in that wavelength range. In fact, we have developed a model for the microwave emission spectrum of Venus, using our laboratory results for gaseous H<sub>2</sub>SO<sub>4</sub>, and previous results for SO<sub>2</sub> and CO<sub>2</sub> which provides a better fit to the ensemble of Venus microwave observations than any model yet proposed (see § IV). A computation of the microwave absorption spectrum from H<sub>2</sub>SO<sub>4</sub>, made by Allen, which is presented in Cimino (1982), shows significantly less absorption from H2SO4 in this same frequency range than we have actually measured This can be explained by the fact that a pressure hardened

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linewidth parameter of 7.2 MHz was assumed for gaseous  $H_2SO_{\bullet}$  in a  $CO_2$  atmosphere. It appears that using a smaller broadening parameter would result in a spectrum which would be more consistent with our measurements.

3. The third major result is the low absorptivity measured at 22.3 cm. This is important since the well-known problem of the fall-off of brightness temperatures at wavelengths longward of 20 cm (see, for example, Muhleman, Orton, and Berge 1979) is still unsolved, and might have been explained by a large opacity from a gaseous constituent. Our negative result indicates that no atmospheric constituent can be responsible for this effect.

In Figure 3, we compare the microwave absorptivity from 15 ppm gaseous H2SO4 in a CO2 atmosphere with that from 150 ppm SO<sub>2</sub> in a CO<sub>2</sub> atmosphere, under pressure and temperature conditions corresponding to an altitude of 35 km in the Venus atmosphere. The opacity from  $SO_2$  is computed from the results of Steffes and Eshleman (1981). The opacity from gaseous H<sub>2</sub>SO<sub>4</sub> is as presented in Figure 2 for a pressure of 6 atm, however, an estimated temperature dependence of  $T^{-3}$  was used in order to determine the effects of the colder temperature at the 35 km altitude of the Venus atmosphere (450 K vs. 575 K used in the laboratory). Likewise, since the microwave absorption from a minor constituent (number mixing ratio less than 10%) is generally directly proportional to its abundance (either by number or by volume), we assume a linear dependence with mixing ratio. The SO<sub>2</sub> mixing ratio (150 ppm) was picked based on in situ measurements (see, for example, Oyama et al. 1980 or Gel'man et al. 1979). The H<sub>2</sub>SO<sub>4</sub> mixing ratio (15 ppm) corresponds to the minimum abundance necessary so as to reach saturation under Venus atmospheric conditions at altitudes at or above 48 km, the highest measured cloud base altitude (see Knollenberg et al. 1980). Note we assume H<sub>2</sub>SO<sub>4</sub> saturation vapor pressure characteristics as described by Steffes (1985). Inspection of Figure 3 shows that while radio occultation and radio astronomical measurements at wavelengths longer than 1.8 cm detect absorption at the 35 km altitude which is predominated by gaseous H2SO4, opacity at this altitude measured by radio astronomical observations in the 1.2-1.8 cm wavelength range is predominated by SO2.

At other altitudes, the frequency above which opacity from  $SO_2$  (150 ppm) predominates over that from gaseous  $H_2SO_4$  (15 ppm) is essentially the same. It should be noted that this applies only to altitudes from 30 km to 50 km, since most chemical models of the Venus atmosphere require the dissociation of gaseous  $H_2SO_4$  at altitudes below 30 km (see, for example, vom Zahn *et al.* 1983), and since the sulfuric acid vapor condenses out at altitudes above 48 km. It should also be noted that at altitudes below 30 km in the Venus atmosphere, the collision-induced microwave absorption from  $CO_2$  (Ho, Kaufman, and Thaddeus 1966) exceeds that from expected abundances of  $SO_2$  (150 ppm) or  $H_2SO_4$  vapor (below 10 ppm), regardless of wavelength.

Thus, our measurements suggest that at wavelengths longer than 1.8 cm, the opacity from the expected quantities of gaseous  $H_2SO_4$  (15-30 ppm) would predominate in the 30-50 km altitude range in the Venus atmosphere, while the opacity from expected quantities of SO<sub>2</sub> predominates at wavelengths from 1.2 to 1.8 cm. At wavelengths shortward of 1 cm, the relative predominance of the opacity from either gaseous  $H_2SO_4$  or SO<sub>2</sub> is not as clear, since both have large numbers of rotational resonances throughout the millimeter-wavelength range. Laboratory measurements of the millimeter-wave absorption properties of constituent gases under simulated conditions for the Venus atmosphere could be a useful tool for

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interpreting the millimeter-wave spectrum of Venus.

IV. MODELS FOR THE MICROWAVE EMISSION SPECTRUM OF VENUS

Several models for the microwave emission spectrum of Venus have been developed, based on numerous observations, in attempts to infer constituent abundances and atmospheric structure. However, the lack of reliable data on both the identities and microwave absorbing properties of many of the atmospheric constituents cast an aura of uncertainty over the resulting inferred atmospheric models. When Pollack and Morrison (1970) attempted to explain the microwave emission spectrum of Venus, they attributed all microwave atmospheric opacity to CO2, N2, gaseous H2O, and dust. It was found that in order to match the observed brightness temperature at the 1.94 cm wavelength, an H<sub>2</sub>O vapor abundance ranging between 3500 ppm and 10.000 ppm was required. This value far exceeds any in situ measurement of water vapor abundance made by any of the subsequent atmospheric probes. In fact, current models of the Venus atmosphere place water vapor abundances below the cloud layers in the 100 ppm range (see von Zahn et al. 1983). In addition, the accompanying opacity at 1.35 cm from such quantities of water vapor would result in a 1.35 cm brightness temperature which would be notably below those measured (see Janssen and Klein 1981).

Muhleman, Orton, and Berge (1979) used an alternative approach for modeling the opacity from atmospheric constituents. Like Pollack and Morrison (1970), they found that opacity from CO<sub>2</sub> alone could not explain radio observations of the planet; that is, additional sources of radio absorption were required. However, instead of attributing this absorption to a specific constituent with known absorption properties, it was assumed that all sources of additional opacity would exhibit microwave opacity whose pressure, temperature, and frequency dependencies would be the same as those for  $CO_2$ . It should be emphasized that this was a perfectly reasonable approach for modeling microwave opacity in the Venus atmosphere previous to the Pioneer-Venus mission, since a large number of potential absorbing constituents might have existed, the microwave absorption properties for which, under Venus atmospheric conditions, were unknown. Similarly, since those constituents whose opacity had been measured under simulated laboratory conditions for the Venus atmosphere (CO2, H2O, and N2-see Ho, Kaufman, and Thaddeus 1966) had all exhibited the same basic pressure, temperature, and frequency dependence, this assumption seemed to be reasonable; yet the authors recognized and stated the possibility for significant error.

As with previous models for atmospheric opacity in the Venus atmosphere, the model used by Muhleman, Orton, and Berge (1979) resulted in a theoretical microwave emission spectrum which agreed quite well with the actual observations at certain wavelengths, but poorly at others. While some of these discrepancies may have been due to calibration errors in the observed data, an opacity frequency dependence other than  $f^2$  would be required in order to better match the observed brightness temperatures at other wavelengths, especially near 2 cm. It was also pointed out that such a difference in frequency dependence was also required in order for the opacity models in the 1.35 cm wavelength range to be able to match observed opacity at 3.6 cm and 13 cm as measured by radio occultation experiments. This was likewise recognized by Janssen and Klein (1981).

As shown in Figure 4, we have developed a model for the microwave emission spectrum from Venus based on the laboratory measurements of opacities of the potential constituents. Computations of the modeled brightness temperatures were

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made by CSP Gurski (private communication) and assume that the primary absorbing constituents of the Venus atmosphere are CO<sub>2</sub>, SO<sub>2</sub>, and gaseous H<sub>2</sub>SO<sub>4</sub>. While some microwave opacity can also be attributed to gaseous H<sub>2</sub>O and the cloud layers themselves, the contribution from the expected abundance of gaseous H<sub>2</sub>O (in the 100 ppm range below the clouds-see von Zahn et al. 1983), and from the cloud layers as measured by Knollenberg and Hunten (1979) and by Ragent and Blamont (1979), would be very small. For our model, we assume an atmosphere with a 96% abundance of CO<sub>2</sub>, and a temperature/pressure structure as measured by the Pioneer-Venus probes (Seiff et al. 1980). The opacity from CO, is computed using the laboratory-based expression for CO2 microwave opacity from Ho, Kaufman, and Thaddeus (1966) and assuming a CO<sub>2</sub> abundance of 96%. We assume a SO<sub>2</sub> abundance which is essentially zero above the main cloud layer (48-50 km) and 150 ppm below the main cloud layer down to the surface. The opacity from SO<sub>2</sub> is computed from the results of Steffes and Eshleman (1981). For gaseous  $H_2SO_4$ , we use the results from § III and assume a uniform abundance between the 30 km altitude and the cloud base (approximately 48 km). Below 30 km, we assume dissociation of gaseous H<sub>2</sub>SO<sub>4</sub> into SO<sub>3</sub> and H<sub>2</sub>O (which are both relatively transparent in the abundances expected) and above 48 km, we assume condensation begins. In Figure 4, we present microwave emission spectrum for two different abundances of gaseous H<sub>2</sub>SO<sub>4</sub>: 30 ppm and 15 ppm. These abundances are near the upper and lower limits for the gaseous H2SO4 abundance required to account for the 13 cm opacity measured by radio occultation experiments (see Steffes 1985). Also plotted are actual observed brightness temperatures, as given by Pollack and Morrison (1970), Kuz'min (1983), and Muhleman, Onton, and Berge (1979). It should be noted that a constant surface microwave brightness of 640 K is assumed, hence the apparent variation between the model and observed emission at longer wavelengths.

Inspection of our model shows a very good correlation with observed brightness temperatures in the 1-10 cm wavelength range, when a sulfuric acid vapor abundance of 15 ppm is assumed. However, certain aspects of the spectra bear special consideration. The first is the 1.2-1.5 cm wavelength range. A great number of brightness measurements have been made in this wavelength range, especially at the 1.35 cm wavelength (the center of the  $H_2O$  absorption band). These measurements range from  $550 \pm 26$  K (see Janssen 1972), down to  $436 \pm 39$  K (Law and Staelin 1968). While some of the resulting variations in the 1.35 cm atmospheric opacity required to explain these brightness temperature variations are probably due to interpretive or calibration inaccuracies, since the observations were made with relatively primitive receivers and since there is substantial uncertainty in the 1.35 cm flux calibration scale, they may suggest significant temporal changes in atmospheric opacity, such as might be induced by volcanism (see, for example, Esposito 1985, or Prinn 1984). A question then arises as to which constituents could be responsible for such opacity variations. Inspection of Figure 4 shows that assuming abundances of 150 ppm SO<sub>2</sub> and 15 ppm H<sub>2</sub>SO<sub>4</sub>, our model predicts a 1.35 cm brightness temperature of 490 K. Even if the H,SO, abundance were doubled to 30 ppm, the resulting reduction in the 1.35 cm brightness temperature would be only 25 K. This is not surprising in light of the relatively low opacity exhibited by H<sub>2</sub>SO<sub>4</sub> at these wavelengths. Thus, we conclude that the variations in 1.35 cm brightness cannot be attributed to variations in the abundance of gaseous H<sub>2</sub>SO<sub>4</sub> since the required abundance variations would far exceed allowable limits for H<sub>2</sub>SO<sub>4</sub> abundance set by sulfuric acid vapor satura-

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tion or 13 cm opacity measurements from radio occultation experiments (see Steffes 1985).

An alternative explanation for opacity variations would be variations in the abundance of SO<sub>2</sub>. If the SO<sub>2</sub> abundance of our model were increased from 150 to 200 ppm (a 33% increase), the 1.35 cm brightness (assuming a constant H2SO4 abundance of 15 ppm) would go from 490 K down to 460 K, well within the error bars of the Law and Staelin (1968) measurement, while not seriously affecting the 13 cm opacity or the emission brightness at wavelengths longer than 1.8 cm. Similarly, reducing the SO<sub>2</sub> abundance to 80 ppm results in a brightness temperature of approximately 525 K which is within the error bars of the Janssen (1972) measurement. It should be emphasized that these apparent long-term variations in opacity may be partially due to calibration inaccuracies; however, the required long-term variations in SO<sub>2</sub> abundance required to explain these opacity variations would be of the same order as the long-term variations in upper atmospheric SO2 abundances as observed by Esposito (1985).

The second interesting aspect of our model for the microwave emission spectrum lies in the 2-3 cm wavelength range. As can be seen from Figure 4, a noticeable dip in brightness temperature should exist between 2 and 3 cm. Unfortunately, few measurements of the Venus microwave emission have been made in this wavelength range. Two measurements have been made of the emission near 2 cm. The first, by McCullough and Boland (1964), was 500  $\pm$  75 K at a wavelength of 2.07 cm. The second, by Pollack and Morrison (1970), was  $495 \pm 35$  K at the 1.94 cm wavelength. Both are in good agreement with the model, assuming a gaseous H<sub>2</sub>SO<sub>4</sub> abundance of 15 ppm. The most interesting wavelengths to observe would be in the 2.2-2.3 cm range, where a strong dip in the microwave emission, related to the abundance of gaseous H2SO4, should occur. Unfortunately, no observations have been published for these wavelengths. Since high-resolution, high-sensitivity receiver systems capable of operation in both this wavelength range and at 1.35 cm do exist, we suggest that long-term observation of Venus microwave emission could be made so as to characterize variations of the SO<sub>2</sub> abundance (1.35 cm) and gaseous H<sub>2</sub>SO<sub>4</sub> abundance (2.2 cm), both on a planet-wide basis, and on a localized basis, as limited by instrument spatial resolution.

As mentioned, our model assumes a constant surface brightness temperature of 640 K. This simplification could be substantially improved. Muhleman, Orton, and Berge (1979) used radar reflectivity measurements in developing a better model for surface brightness but still could not match observations of emission at wavelengths longward of 20 cm. It is likely that laboratory measurements of dielectric properties of simulated surfaces, under simulated Venus atmospheric conditions, may be able to provide an understanding of this phenemonon, and should therefore be pursued. It should also be noted that the expression used for opacity from SO2 was based on laboratory measuremnets in the 1-6 atm pressure range. This expression may understate the opacity from SO2 in the 50-100 atm (0-10 km) range, which would result in our model understating the Venus brightness temperature, especially in the 4-6 cm wavelength range.

#### V. CONCLUSIONS AND SUGGESTIONS FOR FUTURE WORK

Laboratory measurements have been made of the microwave opacity from gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere, under simulated conditions for the middle atmosphere of Venus, in the 1.2-22.3 cm wavelength range. Our laboratory results have shown that the predominant microwave absorbers in the Venus atmosphere at wavelengths longer than about 1.8 cm are gaseous  $H_2SO_4$  and  $CO_2$ , whereas  $SO_2$  and  $CO_2$  pre-

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dominate at shorter wavelengths. Thus, in using these measurements to model the Venus microwave emission spectrum, we have found that gaseous  $H_2SO_4$  and  $CO_2$  are the constituents most responsible for the Venus microwave emission spectrum at wavelengths longer than about 1.8 cm, and that SO<sub>2</sub> and CO, most affect the emission spectrum between 1.2 and 1.8 cm. Our model of the Venus microwave emission spectrum correlates well with the ensemble of microwave observations of Venus when abundances of  $H_2O$ ,  $SO_2$ , and gaseous  $H_2SO_4$ , below the main cloud layer, are assumed to be 100 ppm, 150 ppm, and 15 ppm, respectively. Analysis of our model has shown that the significant variations observed in the 1.35 cm emission cannot be explained by allowable variations in the abundance of gaseous  $H_2SO_4$  abundance (15-30 ppm, as limited by 13 cm opacity measurements), but could be explained by variations in the subcloud SO<sub>2</sub> abundance from approximately 80 ppm to 200 ppm.

Additional microwave observations of Venus should be conducted both at 1.35 cm and near the 2.2 cm wavelength in order to better characterize both spatial and temporal variations in abundances of atmospheric SO<sub>2</sub> and gaseous H<sub>2</sub>SO<sub>4</sub>. We also intend to conduct further analysis of existing observational data, in light of the laboratory results. A final critical issue to the understanding of the Venus microwave spectrum involves the surface microwave emission. Actual laboratory measurement of the microwave dielectric properties of simulated surfaces under Venus atmospheric conditions would be an important key to a better understanding of the microwave emission of the surface as well to characterizing variations in surface structure from existing and future radar measurements.

The author wishes to thank graduate students Gregory (\$) Gurski, for his assistance in modeling the Venus microwave spectrum, and David H. Watson, for his assistance in conducting laboratory measurements. The author also thanks Ms. Peggy Knight and Ms. Mary Ann Tripp for assistance in typing and providing graphic arts for this manuscript. This work was supported by the Planetary Atmospheres Program of the Solar System Exploration Division (Office of Space Science and Applications) of the National Aeronautics and Space Administration under grant NAGW-533.

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IG. 1.—Block diagram of the atmospheric simulator as configured for measurements of the microwave absorption of gaseous  $H_2SO_4$  under Venus atmospheric tions over the 1.2–22.3 cm wavelength range.

G. 2.—Laboratory measurements of absorptivity (dB km<sup>-1</sup>) of gaseous  $H_2SO_4$  in a  $CO_2$  atmosphere (T = 575 K;  $H_2SO_4$  mixing ratio = 0.4%). Solid curves est-fit expressions for mixtures with total pressures of 6 atm and 1 atm.

G. 3.—Comparative microwave absorption from gaseous  $H_2SO_4$  (15 ppm, solid line) and  $SO_2$  (150 ppm, dashed line) at the 35 km altitude in the Venus sphere (P = 6 atm; T = 450 K).

16. 4.—Measurements of the microwave emission from Venus. The solid represents the model when a 15 ppm abundance of gaseous  $H_2SO_4$  is med below the main cloud layer. The dashed line represents the model a a 30 ppm  $H_2SO_4$  abundance is assumed. Both models assume abunres for  $CO_2$ ,  $H_2O_4$  and  $SO_2$  of 96%, 100 ppm, and 150 ppm, respectively.

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Dr. Kenneth Fox Discipline Scientist Solar System Exploration Division Code EL National Aeronautics and Space Administration Washington, DC 20546

Dear Dr. Fox:

As specified in the NASA Provisions for Research Grants and Cooperative Agreements, I am enclosing three (3) copies of <u>Semiannual Status Report #7 for</u> <u>Grant NAGW-533</u> (Laboratory Evaluation and Application of Microwave Absorption Properties under Simulated Conditions for Planetary Atmospheres). As required, I am also forwarding two (2) copies of the report to the NASA Scientific and Technical Information Facility.

I apologize for the lengthiness of the report, but we are working on several projects "in parallel", and I felt it appropriate to include background information and current status on all of the activities. With the 10% reduction in our budget for this grant year, I was fearful that our research productivity would drop. However, I was able to obtain (for 1 year, non-renewable) support for an additional graduate student from the Office of the Vice President of Research at Georgia Tech. As a result, I am looking forward to completing a productive grant year, with (hopefully) a goodly increase in journal publications.

We are looking forward to the DPS meeting this fall, and hope to see you there. If I can provide any additional information or assistance, feel free to contact me by telphone or letter.

Sincerely,

Paul G. Steffes Assistant Professor

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Ladies and Gentlemen:

As specified in the NASA Provisions for Research Grants and Cooperative Agreements, I am enclosing two (2) copies of <u>Semiannual Report #7 for Grant</u> <u>NAGW-533</u> (Laboratory Evaluation and Application of Microwave Absorption Properties under Simulated Conditions for Planetary Atmospheres). I am also forwarding three (3) copies of the report to the NASA technical officers for this grant (K. Fox and H. C. Brinton, Code EL), as required.

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### REPORT

# TO THE

# NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

### SEMIANNUAL STATUS REPORT #7

for

GRANT NAGW-533

# LABORATORY EVALUATION AND APPLICATION OF MICROWAVE ABSORPTION PROPERTIES UNDER SIMULATED CONDITIONS FOR PLANETARY ATMOSPHERES

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Submitted by

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### I. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often leads to significant misinterpretation of available opacity data. Steffes and Eshleman (1981) showed that under environmental conditions corresponding to the middle atmosphere of Venus, the microwave absorption due to atmospheric  $SO_2$  was 50 percent greater than that calculated from Van Vleck-Weiskopff theory. Similarly, results obtained for the microwave opacity from gaseous  $H_2SO_4$  under simulated Venus conditions, during the first two years of Grant NAGW-533, showed that not only was the opacity from  $H_2SO_4$  much greater than theoretically predicted, but that its frequency (wavelength) dependence was far different than that theoretically predicted (Steffes, 1985 and Steffes, 1986). More recently, measurements made by Steffes and Jenkins (1987), during the third year of Grant NAGW-533, have shown that simulated Jovian conditions do indeed agree with theoretical predictions, but only at wavelengths longward of 1.3 cm. The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by both radio occultation experiments and radio astronomical observations, and over a range of frequencies which correspond to those used

in both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has been the goal of this investigation to conduct such measurements and to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profiles of constituents in those planetary atmospheres.

Initially, this facility was developed, and then operated, in order to evaluate the microwave absorbing properties of gaseous sulfuric acid  $(H_2SO_A)$ under Venus atmospheric conditions. The results, obtained at 13.4 cm and 3.6 cm wavelengths, were applied to measurements from Mariner 5, Mariner 10, and Pioneer-Venus Radio Occultation experiments, to determine abundances of gaseous sulfuric acid in the Venus atmosphere, with accuracies exceeding those achieved with in-situ instruments (Steffes, 1985). Later efforts concentrated on making laboratory measurements of the microwave absorption from gaseous  $H_2SO_4$  at wavelengths from 1.2 to 22 cm under simulated Venus conditions. We applied these results to radio astronomical observations of Venus which have been made in the same wavelength range, in order to better model the structure of  $H_2SO_4$  and  $SO_2$  abundance in the Venus atmosphere, and to resolve temporal variations of their abundances on a planet-wide basis. The results of this effort were especially rewarding in that the unique frequency and pressure dependences measured for the absorption from gaseous  $H_2SO_4$  in these wavelength ranges has finally explained what were thought to be inconsistencies between measurements of the absorption in the Venus atmosphere at 13.3 and 3.6 cm wavelengths and those obtained in the 1 to 2 cm wavelength range. Our laboratory measurements also suggested that a substantial variation in the

Venus microwave emission, related to the abundance of gaseous sulfuric acid, might exist near the 2.2 cm wavelength. Since no observations of the Venus emission at this wavelength have ever been published, we conducted observations of Venus using the 140-foot NRAO telescope in April 1987 to not only search for the presence of the predicted feature, but to use such a feature to determine a planet-wide average for sulfuric acid vapor abundance below the main cloud layer. This observation and initial results are discussed in Section V of this report.

The highest priority activity for the first half of this grant year has continued to be laboratory measurements of the microwave properties of the simulated atmospheres of the outer planets and their satellites. As described in the previous Annual Status Report for Grant NAGW-533 (February 1, 1986 through January 31, 1987), initial laboratory measurements of the microwave opacity of gaseous ammonia (NH<sub>3</sub>) in a hydrogen/helium (H<sub>2</sub>/He) atmosphere, under simulated conditions for the outer planets were completed in September 1986. These measurements were conducted at frequencies from 1.3 GHz to 22 GHz (wavelengths from 1.3 cm to 22 cm), at temperatures from 178 K to 300 K, and under total pressures reaching as high as 6 atmospheres. Such measurements have long been sought by a number of researchers working on inferring ammonia abundance profiles in Jovian atmospheres. Our measurements represented the first time that measurements of the microwave absorption of gaseous ammonia under simulated conditions for the outer planets had been conducted. The results of these measurements, and their effect on the interpretation of microwave opacity data from Jupiter, obtained both from Voyager radio occultation measurements made at 13 cm and 3.6 cm wavelengths and from radio astronomical observations in the 1.3 cm to 20 cm wavelength range, are

discussed in a paper entitled "Laboratory Measurements of the Microwave Opacity of Gaseous Ammonia (NH<sub>3</sub>) Under Simulated Conditions for the Jovian Atmosphere," by Steffes and Jenkins, which was submitted to Icarus in December 1986, and has since been revised and accepted for publication (accepted April 20, 1987). These results showed that in the 1.38 to 18.5 cm wavelength range, the absorption from gaseous ammonia was correctly expressed by the modified Ben-Reuven lineshape as per Berge and Gulkis (1976). As a result, it became clear that the opacity which would be exhibited by a solar abundance of ammonia would be a factor of 1.5-2.0 below the opacity at the 2 to 6 Bar levels of the atmosphere as inferred from radio emission studies in the 6 to 20 cm wavelength range (see de Pater and Massie, 1985 and de Pater, 1986), and the opacity at the 1 Bar level measured by the Voyager 13 cm wavelength radio occultation experiment (see Lindal et al., 1981). This additional opacity, while most likely due to an overabundance of gaseous ammonia beyond the solar abundance, may also be partially due to other gases or condensates, especially in the 2 to 6 Bar levels of the atmosphere.

As a result, in the first half of this current grant year, we have continued such laboratory measurements of the microwave absorption and refraction from other potential microwave absorbers contained in the outer planets' atmospheres, including methane ( $CH_4$ ) and water vapor ( $H_2O$ ), as well as additional high-sensitivity mesurements of the absorption from gaseous  $NH_3$ at 13.3 and 18.5 cm. It has also been brought to our attention by several researchers that significant uncertainties exist as to the actual absorption spectrum of gaseous ammonia at wavelengths shortward of 1 cm. It has been found by some (e.g., de Pater and Massie, 1985) that the observed millimeterwave emission from Jupiter is inconsistent with the millimeter-wave absorption

spectrum predicted using the modified Ben-Reuven line shape for ammonia. In order to confirm this, we have recently developed a Fabry-Perot spectrometer system capable of operation from 30 to 41 GHz (wavelengths from 7.3 to 10 mm). This system can be used at pressures up to 2 Bars and temperatures as low as 150 K, which corresponds closely to the conditions at altitudes in the Jovian atmosphere most responsible for the observed millimeter-wave absorption. Measurements are currently being conducted, and preliminary results are presented in Section IV. A complete description of the millimeter-wave spectrometer is given in Section II.

In the second half of this grant year, we hope to complete laboratory measurements of the 7.3-8.3 mm absorption spectrum of ammonia, as well as additional measurements of  $H_2O$  and  $CH_{A*}$ . We are also investigating the feasibility of making laboratory measurements of the microwave properties of gaseous phosphine (PH<sub>2</sub>). Like ammonia, phosphine is a symmetric-top molecule with rotational resonances at millimeter or submillimeter wavelengths. However, also like ammonia, an inversion spectrum should exist much lower in frequency (see Townes and Schawlow, 1955). Unlike the ammonia inversion spectrum, which is centered around 23 GHz, the phosphine inversion spectrum is expected below 10 MHz. However, under the proper conditions of temperature and high pressure, phosphine may exhibit measurable absorption in the 10 to 20 cm wavelength range. Laboratory measurements of the microwave properties of phosphine in a hydrogen/helium atmosphere present even greater hazards than those for the previous measurements of ammonia in a like atmosphere. Because of the high toxicity of phosphine (more than 30 times more toxic than ammonia), self-contained breathing apparatus must be available in case of accident. Likewise, special ventilation procedures will be required since

phosphine can spontaneously combust. We hope to measure the microwave absorption properties of phosphine, under simulated Jovian conditions (i.e., temperatures down to 153 K and pressures to 6 atm in a hydrogen/helium atmosphere), and over the full wavelength range (1.37 cm to 18.5 cm), but only after completion of the measurements for gaseous NH3, H20, and CH4. We likewise hope to pursue a program of further analysis and application of our laboratory results to microwave data for the outer planets, such as Voyager Radio Occultation experiments and earth-based radio astronomical observations. Of equal importance, we feel however, would be the further analysis and application of our laboratory results for the microwave absorption from gaseous SO2 and gaseous  $H_2SO_4$  in the Venus atmosphere. Our long term goal would be a detailed analysis of available multi-spectral microwave opacity data from Venus, including data from the Pioneer-Venus Radio Occultation experiments and earth-based radio and radar astronomical observations, such as the kinds which have been performed at the NRAO Very Large Array (VLA) and at stations in the Deep Space Network (DSN). The new measurements of Venus microwave emission at 2.25 cm and 1.9 cm made with the NRAO 140-foot telescope are an especially important contribution to this data set. This would provide a chance to determine both spatial and temporal variations in the abundances of both  $H_2SO_4$ and SO<sub>2</sub> in the Venus atmosphere. We intend to pursue discussions with the Pioneer-Venus Radioscience Team Leader, in order to obtain additional absorptivity data from the 3.6 and 13 cm radio occultation experiments.

# II. THE GEORGIA TECH RADIO ASTRONOMY AND PROPAGATION (R.A.P.) FACILITY

The basic configuration of the planetary atmospheres simulator developed at Georgia Tech for use in measurement of the microwave absorptivity of gases under simulated conditions for planetary atmospheres is described at length in the previous Annual Status Report(s) for Grant NAGW-533. It is also discussed at length in Steffes (1986) and Steffes and Jenkins (1987). The updated simulator system, shown in Figure 1, is currently configured for simulations of the outer planets. Measurements of the microwave opacity and refractivity of test gas mixtures can be performed at frequencies from 1.6 GHz to 27 GHz (wavelengths from 1.2 cm to 18.5 cm) using two microwave resonators contained within a pressure chamber. While the pressure chamber itself is capable of containing pressures up to 10 atmospheres, and the temperature chamber is capable of achieving temperatures as low as 150 K, it has been found that the combination of high pressures and low temperatures can create a substantial problem with sealing the pressure chamber. The use of gaseous hydrogen  $(H_2)$ in the outer planets simulations has required the development of procedures and equipment for conducting such simulations. Such precautions have included the use of a hydrogen leakage sensor which is placed inside the temperature chamber immediately outside the pressure vessel (see Figure 1). This sensor can detect the potentially dangerous build-up of hydrogen gas within the freezer unit. A ventilation pump has also been provided which can be used to draw any escaping hydrogen gas out of the freezer compartment. Additional precautions have included ramps which allow all of the equipment to be moved out-of-doors to a concrete slab immediately adjacent to the laboratory. All experiments which employ gaseous hydrogen can thus be conducted out-of-doors

in order to avoid any build-up of hydrogen gas within the laboratory. A covered outdoor storage area for hydrogen and helium gases has also been constructed in order to allow safe storage of these gases, and to expedite the out-of-doors experiments. In addition, two vacuum sensors are included in the system. These sensors not only allow accurate determination of pressure vessel evacuation, but they can also be used for accurately determining the abundances of microwave-absorbing test gases, which are typically very small at low temperatures, due to low saturation vapor pressures.

The overall result has been a pressure vessel large enough to contain two microwave resonators, which is capable of maintaining 6 atmospheres of pressure at a temperature of 150 K, with an acceptably small leak ratio. While the range of pressures which can be tested was not quite as large as originally hoped, the resulting range of temperatures and pressures does represent the range over which nearly all of the microwave opacity in the Jupiter atmosphere has been observed, and thus is extremely useful in interpretation of microwave opacity data from Voyager I and II radio occultation experiments, as well as from earth based radio astronomical observations, and, in the future, opacity measurements to be made using the Galileo probe. Likewise, the pressure-temperature ranges measured are close enough to those over which microwave absorption or refraction has been measured in the atmospheres of Saturn, Titan, Uranus, and Neptune, so that accurate estimates of abundances of microwave-absorbing constituents in these atmospheres can also be made.

The most recent addition to the Georgia Tech Radio Astronomy and Propagation Facility has been a Fabry-Perot type resonator capable of operation between 30 and 41 GHz. As shown in Figure 2, the resonator consists of two

gold plated mirrors separated by a distance of about 20 cm. The mirrors are contained in a T-shaped glass pipe which serves as a pressure vessel capable of withstanding over 2 atm of pressure. Each of the three open ends of the pipe is sealed with an O-ring sandwiched between the lip of the glass and a flat brass plate which is bolted to an inner flange. Electromagnetic energy is coupled both to and from the resonator by twin irises located on the surface of one of the two mirrors. Two sections of WR-28 waveguide which are attached to the irises pass through the brass plate to the exterior of the pressure vessel. The end of each waveguide section is pressure-sealed by a rectangular piece of mica which is held in place by a mixture of rosin and beeswax. As shown in Figure 3, one of these ends is connected to the sweep oscillator through a waveguide section. A Ka-band mixer is attached to the other section of waveguide and is coupled to the high resolution spectrum analyzer through a calibrated section of coaxial cable. The entire resonator, including its glass pressure envelope, is placed in the temperature chamber, which is a low-temperature freezer capable of operation down to 150 K. A network of stainless steel tubing and valves connects other components such as gas storage tanks, vacuum gauges, the pressure gauge, and the vacuum pump to the resonator assembly, so that each component may be isolated from the system as necessary. When properly secured, the system is capable of containing up to two atmospheres of pressure without detectable leakage.

### III. EXPERIMENTAL APPROACH

The approach used to measure the microwave absorptivity of test gases in an  $H_2/He$  atmosphere is similar to that used previously by Steffes and Jenkins (1987) for simulated Jovian atmospheres. At frequencies below 22 GHz (see

Figure 1), the absorptivity is measured by observing the effects of the introduced gas mixture on the Q, or quality factor, of two cavity resonators at particular resonances from 1.34 GHz to 21.8 GHz. At frequencies between 35 and 41 GHz, the changes in the Q of several resonances of the Fabry-Perot resonator (see Figure 3) are related to the absorptivity of the test gas mixture at these frequencies. The changes in the Q of the resonances which are induced by the introduction of an absorbing gas mixture can be monitored by the high resolution microwave spectrum analyzer, since Q is simply the ratio of the cavity resonant frequency to its half-power bandwidth. For relatively low-loss gas mixtures, the relation between the absorptivity of the gas mixture and its effect on the Q of a resonance is straightforward:

$$\alpha \simeq \left(Q_{\rm L}^{-1} - Q_{\rm C}^{-1}\right) \pi / \lambda \tag{3}$$

where  $\alpha$  is absorptivity of the gas mixture in Nepers km<sup>-1</sup>. (Note, for example, that an attenuation constant or absorption coefficient or absorptivity of 1 Neper km<sup>-1</sup> = 2 optical depths per km (or km<sup>-1</sup>) = 8.686 dB km<sup>-1</sup>, where the first notation is the natural form used in electrical engineering, the second is the usual form in physics and astronomy, and the third is the common (logarithmic) form. The third form is often used in order to avoid a possible factor-of-two ambiguity in meaning.)  $Q_L$  is the quality factor of the cavity resonator when the gas mixture is present,  $Q_C$  is the quality factor of the resonance in a vacuum, and  $\lambda$  is the wavelength (in km) of the test signal in the gas mixture.

The first experiments involved high-sensitivity measurements of the microwave absorption from gaseous  $NH_3$  under simulated conditions for the 2 to

6 Bar pressure range of the Jovian atmosphere in the 10 to 20 cm wavelength range. These measurements were undertaken in order to help better explain the source of the microwave opacity at altitudes below the 2 atm pressure level in the Jovian atmosphere. (See Steffes and Jenkins, 1987, or the previous <u>Annual Status Report</u> for Grant NAGW-533, for further discussion.) These measurements were conducted at 1.62 GHz (18.5 cm) and 2.25 GHz (13.3 cm), and took advantage of special adjustments to the atmospheric simulator so as to maximize sensitivity in the 10 to 20 cm wavelength range.

Temperatures from 193 K to 300 K were used for the experiments, since lower temperatures would risk condensation of the gaseous NH3. When the pressure vessel reached thermal stability at the desired temperature, which was monitored using both the temperature sensors and the resonant frequencies of the system, a vacuum was drawn in the pressure vessel containing the resonators, and the bandwidth and center frequency of each of resonances was then measured. A valve was then opened which allowed the ammonia gas to enter the chamber, where 17 torr NH<sub>3</sub> pressure was used. Measurements of the gaseous NH<sub>3</sub> pressure were made with the high accuracy thermocouple vacuum gauge tubes which are shown in Figure 1. Next, 5.4 atm of hydrogen  $(H_2)$  and 0.6 atm of These gases were admitted to the chamber at a helium (He) were added. sufficiently slow rate so as not to significantly affect the temperature within the chamber. The result was an atmosphere with 6 atm total pressure composed of 90 percent hydrogen, 10 percent helium, and approximately 3730 ppm ammonia. The bandwidth of each resonance was then measured and compared with its value when the chamber was evacuated in order to determine the absorptivity of the gas mixture at 6 atm total pressure. The total pressure was then reduced by venting to 4 atm, and the bandwidths were again measured.

Subsequent measurements were likewise made at 2 atm pressure. The pressure vessel was then evacuated and the bandwidths again measured so as to assure no variation (either due to thermal shift or chemical reaction) of the Q's of the evacuated resonators had occurred. The measured changes of bandwidth (Q) were then used to compute the absorptivity of the gas mixture under the various pressure conditions.

This approach has the advantage that the same gas mixture is used for the absorptivity measurements at the various pressures. Thus, even though some small uncertainty may exist as to the mixing ratio of the initial mixture, the mixing ratios at all pressures are the same, and thus the uncertainty for any derived pressure <u>dependence</u> is due only to the accuracy limits of the absorptivity measurements, and not to uncertainty in the mixing ratio. (This assumes that the mixing ratio is small, so that foreign-gas broadening predominates, as is the case for our measurements.) Similarly, measurements of the frequency <u>dependence</u> of the absorptivity from the mixture would likewise be immune to any mixing ratio uncertainty, since foreign-gas broadening predominates.

The second major set of experiments involved measurements of the microwave absorption from methane ( $CH_4$ ) under simulated Jovian conditions at frequencies from 2.2 to 21.7 GHz. These experiments employed the same dualcavity atmospheric simulator as was used for the earlier ammonia experiments, shown in Figure 1. However, since methane absorption is very low, much larger methane mixing ratios were required to achieve detectable levels of absorption at these wavelengths. Likewise, since the methane absorption is inversely proportional to temperature, the lowest possible temperatures were used so as

to maximize measurable absorption. In contrast to ammonia, the saturation vapor pressure of methane is quite large, thus risk of condensation was not a consideration. As a result, absorption from pure methane at pressure up to 6 Bars and at a temperature of 153 K was measured, as well as absorption from a 40% methane, 52% hydrogen, and 8% helium atmosphere under the same conditions of temperature and pressure. These measurements were made using resonances at 2.25 GHz (13.3 cm), 8.52 GHz (3.5 cm), and 21.7 GHz (1.38 cm).

The third set of measurements were made at the same frequencies, but involved measurements of the microwave absorption of water vapor under simulated Jovian conditions. Because of the relatively low saturation vapor pressure of water, experiments were conducted at 300 K, in order to obtain sufficient water vapor for the experiment. As with the previous experiments, the dual-cavity atmospheric simulator, shown in Figure 1, was used. A major difference, however, was the source of the test gas, in this case, H<sub>2</sub>O. Since pressurized cannisters of water vapor are not possible at room temperature or lower, a flask of liquid  $H_2O$  was used as the source of water vapor. Using techniques similar to those used by Steffes (1985 and 1986) for measurement of the microwave absorption from gaseous  $H_2SO_4$ , a precise quantity of water vapor was obtained. That is, the flask is filled with a precisely known volume of distilled water. A vacuum is then drawn in the pressure vessel containing the microwave resonantors, and the bandwidth and center frequencies of the resonances are then measured. A valve is then opened which allows the water vapor eluting from the flask to fill the evacuated pressure vessel (0.031  $m^3$  of open volume with the resonators in place) and reach vapor pressure equilibrium with the liquid  $H_0O$ . Note that all components which contact the water vapor are maintained at the same temperature as the flask, so as to avoid condensation.

As H<sub>2</sub>O vapor fills the chamber, changes in the resonance center frequencies are observed. These changes are related to the H2O vapor abundance. After approximately 10 minutes, the frequency shift ceases, as vapor pressure equilibrium is reached. The valve to the reservoir flask is then closed, and 5.4 atm of  $H_2$  and 0.6 atm of He are admitted to the chamber containing the  $H_2^0$ vapor. The bandwidth of each response is then measured and compared with its value when the chamber was evacuated in order to determine the absorptivity of the gas mixture at 6 atm total pressure. The total pressure is then reduced by venting, and the bandwidths are again measured. Subsequent measurements are likewise made at lower pressures in order to determine absorptivities at those pressures. The pressure vessel is then evacuated and the bandwidths again measured so as to assure no variation (either due to thermal shift or chemical reaction) of the Q's of the evacuated resonators has occurred. After the experiment is complete, the volume of the remaining liquid is measured and compared with the initial volume measured in order to determine the amount of H<sub>2</sub>O vapor present in the gas mixture tested. As with the ammonia experiments, this approach has the advantage that the same gas mixture is used for the absorptivity measurements at the various pressures.

The fourth set of measurements, which are currently being conducted, involve the millimeter-wave absorption from gaseous ammonia  $(NH_3)$  in a 90% H<sub>2</sub>/10% He atmosphere. At the present time, the Fabry-Perot resonator (shown in Figure 2) is being modified so as to allow operation at temperatures down to 150 K. Therefore, the initial measurements were conducted at room temperature. As in the previous experiments at lower frequencies, the bandwidth and center frequencies of each of four resonances (36.29 GHz, 38.43 GHz, 39.15 GHz, and 40.62 GHz) were measured in a vacuum. Next, 20 torr of gaseous

ammonia is added to the system. The pressure of the ammonia gas is measured with the high-accuracy thermocouple vacuum gauge, as shown in Figure 3. Next, 1.8 atm of hydrogen  $(H_2)$  and 0.2 atm of helium (He) are added, bringing the total pressure to 2 atm. The bandwidth of each resonance is then measured and compared with its value when the chamber was evacuated in order to determine the absorptivity of the gas mixture at 2 atm total pressure. The total pressure is then reduced, by venting, to 1 atm, and the bandwidths are again measured. Finally, the pressure vessel is again evacuated and the bandwidths again measured so as to assure no variation of the Q's of the evacuated resonator has occurred. As with the previous measurements, the measured changes of bandwidths (Q's) can then be used to compute the absorptivity of the gas mixtures at each of the resonant frequencies.

In all four of the measurements described, the amount of absorption being measured is extremely small. Thus, any errors in measurements of (or other changes in) the apparent bandwidth of the resonances, not caused by the absorbing gases, could lead to significant errors in the absorption measurement. The contribution of instrumental errors and noise-induced errors on such absorptivity measurements have been discussed at length in Steffes and Jenkins (1987). However, because our latest measurements represent such small percentage changes in bandwidth, another instrumental source of error which we refer to as dielectric loading becomes a concern.

As can be seen in Figures 1 and 3, the resonators, which operate as bandpass filters, are connected to a signal source (the microwave sweep oscillator) and to a signal receiver (the high-resolution spectrum analyzer). The "Q" of the resonator, which is defined as the ratio of energy stored in the resonator to the energy lost per cycle, equals the ratio of resonant

center frequency to resonance half-power bandwidth. It is not surprising, therefore, that the stronger the coupling between the resonator and the spectrum analyzer or sweep oscillator, the lower will be the Q of the resonance, since more energy will be lost per cycle through the cables or waveguide connecting the resonator to the spectrum analyzer and sweep oscillator. For this reason, we have always designed our resonators (both the coaxially-coupled cylindrical cavity resonator used below 25 GHz and the waveguide-coupled Fabry-Perot resonator used above 30 GHz) with minimal coupling, so as to maximize Q and to minimize the changes in Q that might result from changes in coupling that occur when gases are introduced into the resonators. It should be noted that these changes in coupling, which are due to the presence of the test gas mixtures, are not related to the absorptivity of the gases, but rather to the dielectric constant or permittivity of the test gas mixtures. (Hence, the term "dielectric loading.")

We have always strived to design the coupling elements of the resonators so that the changes in lossless test gas abundances (and resulting changes in dielectric constant) had little or no effect on the Q of the resonator as measured in the system. This has been no small feat in that slight imperfections in resonators, cables, coupling loops, or waveguides can make the apparent Q of the resonator appear to vary with the abundance of such lossless gases. It has now become a standard part of our experimental procedure to repeat absorption measurements for gas mixtures in which the absorbing gas is a minor constituent, without the absorbing gas present. For example, after measurements were made of the microwave and millimeter-wave absorption from ammonia as a minor constituent in an  $H_2/He$  atmosphere, measurements of the apparent absorption of the  $H_2/He$  atmosphere without the ammonia gas were made.

Since, for the pressures and wavelengths involved, the  $H_2/He$  atmosphere is essentially transparent, no absorption was expected. If any apparent absorption was detected, "dielectric loading," or a change in coupling due to the dielectric properties of the gases, was indicated.

Initially, if any evidence of dielectric loading existed, the experiments were terminated and the apparatus disassembled, including pressure seals. The cables and coupling loops were then readjusted, and the system reassembled and tested again. The entire procedure was repeated until the dielectric loading effect was eliminated or minimized. If some small variation in the resonant Q or bandwidth due to the presence of the non-absorbing gases still remained, it was added to the uncertainty or error bars for each experiment. More recently however, we have found that the effects of dielectric loading are additive, in that they add to the apparent changes of resonator bandwidth caused by the absorbing gases. Thus, as long as the effects of dielectric loading are not time variable, they can be removed by using the measured value of the Q of a resonance with the non-absorbing gases present (instead of the Q of the resonance in a vacuum) for the quantity  $Q_c$  in equation (3).

### IV. RESULTS OF LABORATORY MEASUREMENTS AND THEIR APPLICATION

As described in Section III, the first experimental sequence involved measurements of the microwave absorption from gaseous ammonia  $(NH_3)$  in a 90% hydrogen/10% helium atmosphere at the frequencies 1.62 GHz (18.5 cm) and 2.25 GHz (13.3 cm). These experiments were conducted with an ammonia mixing ratio of 3730 ppm, at temperatures as low as 193 K, with total pressures up to 6 atm. The results of these measurements are shown in tabular form for 1.62 GHz (Table I) and 2.25 GHz (Table II). Note that in Table II we also

include a listing of all our previous measurements of the 2.25 GHz absorption from gaseous ammonia. Figure 4 presents the results of all of our absorptivity measurements for such an ammonia mixture at 193 K. Also shown are plots of the theoretically-derived absorption spectra for such gas mixtures at these three pressures. These theoretically-derived NH3 absorption spectra were calculated as per de Pater and Massie (1985) and Berge and Gulkis (1976), both of which employ a Ben-Reuven line shape which has been modified so as to be consistent with the laboratory results of Morris and Parsons (1970), in which the absorption from  $NH_3$  in a high-pressure  $H_2/He$  atmosphere at room temperature and at 9.58 GHz was measured. Our theoretical spectra have been adjusted for the temperature, pressure, and mixing ratio conditions of our experiment. This mathematical expression for the absorption of ammonia was implemented in a BASIC program for which partial pressures from  $H_2$ , He, and NH3, as well as frequency and temperature, were adjustable variables. This program was used to calculate the theoretical values for absorption given in Tables I, II, and IV and in Figures 4 and 5.

Inspection of Figure 4 shows that the laboratory data agree surprisingly well with the theoretical predictions for  $NH_3$  opacity made using the Ben-Reuven lineshape, and are, likewise, consistent with the results of Morris and Parsons (1970). The key result of our work has been the finding that the modified Ben-Reuven lineshape appears to correctly describe the shape of the absorption spectrum from gaseous ammonia in a hydrogen/helium atmosphere in the 1.3 to 18.5 cm wavelength range even under the temperature-pressure conditions characteristic of the Jovian atmosphere. This finding answers some questions and raises others. For example, both de Pater and Massie (1985) and West <u>et al.</u> (1986) recognized that, based on interpretation of the Jovian

emission spectrum in the 10 to 20 cm wavelength range using the theoreticallyderived absorption spectrum from ammonia, opacity at pressure levels greater than 2 atm had to exceed the amount which would be caused by the solar abundance of ammonia. As a result, both sets of authors concluded that either the theoretical lineshape was incorrect and understated the opacity of ammonia at these wavelengths by a factor of 1.5-2.0, or the ammonia abundance was greater than its solar abundance by a factor of 1.5-2.0 at pressures greater than 2 atm, or that an extra opacity source, possibly  $H_2O$  condensate, was present. Thus, since our measurements of the ammonia opacity at 13.3 cm (2.25 GHz) agree quite closely with the theoretically-derived values for opacity, and since our newest results at 18.5 cm (1.62 GHz) are likewise consistent with the theoretical lineshape, it would appear that either the ammonia abundance is greater than solar by a factor of 1.5-2.0 in the deeper layers of the Jovian atmosphere, or that an extra opacity source is present.

In an attempt to resolve this question, we have made measurements of the microwave opacity of two other Jovian atmospheric constituents: methane and water vapor. The methane experiment was conducted as described in Section III, and not surprisingly, the microwave absorption was very small. Measurements were made at 153 K of the microwave absorption from a pure methane atmosphere at 6 atm pressure, and from a 40% methane, 52% hydrogen, and 8% helium atmosphere at 6 atm pressure. No absorption was measured at the 2.25 GHz (13.3 cm) or 8.52 GHz (3.5 cm) resonances. Some absorption was measured at the 21.7 GHz (1.38 cm) resonance, but its statistical significance is uncertain since the levels measured were below the error bars for the system. (For the pure methane, the measured absorption was  $11.4 \pm 17$  dB/km. For the 40% methane mixture, the measured absorption was  $9.4 \pm 18$  dB/km.) We

intend to further study the theoretically-derived methane absorption spectrum for purposes of comparison and to determine whether further laboratory measurements at higher frequencies might be useful. However, the results strongly indicate that methane absorption in the 3-13 cm wavelength range is negligible in the 2-6 Bar pressure range of Jupiter's atmosphere, and cannot account for the additional opacity required in that altitude range.

Measurements of the microwave absorption from water vapor were conducted at 2.25 GHz, 8.52 GHz, and 21.7 GHz at 297 K in a 90%  $H_2/10$ % He atmosphere as described in Section III. A water vapor mixing ratio of 3509 ppm was used, and total pressures of 6, 4, and 2 atm were employed. None of the measurements detected statistically significant opacity. (System thresholds were 0.45 dB/km, 0.73 dB/km, and 36 dB/km at 2.25 GHz, 8.52 GHz, and 21.7 GHz, respectively.) Thus our results for the microwave opacity from water vapor (1.38 to 13.3 cm) under simulated Jovian conditions are consistent with the theoretically-based expression for opacity derived by Goodman (1969). Furthermore, our results indicate that, like methane, water vapor cannot account for the additional opacity required in the 2-6 Bar pressure range of Jupiter's atmosphere. Thus, all three of these measurements further strengthen the case for an ammonia abundance between 1.5 and 2.0 times larger than solar abundance.

The measurements of the 7.3-8.3 mm absorptivity from  $NH_3$  in a hydrogen/ helium atmosphere were conducted at 298 K as described in Section III, with an ammonia mixing ratio of 1.32 x  $10^{-2}$ , at pressures of 1 and 2 Bars. An examination of the experimental results summarized in Table III and displayed in Figure 5 reveals that because of large error bars, we cannot determine whether the modified Ben-Reuven lineshape best describes the absorption profile of

gaseous ammonia shortward of 1 cm. However, the consistency of our results at 2 atm suggests that the modified Ben-Reuven lineshape understates the actual absorptivity of ammonia in this range. With the exception of one measurement at 40.62 GHz, all measurements were greater than those predicted by the Ben-Reuven theory (see Table III and Figure 5). At 1 atm, however, the observed absorption is so low that no absorption was measured at several frequencies. In its present configuration, our planetary atmospheres simulator is not sensitive enough to make reliable measurements using a total pressure of 1 atm.

There are several ways that we may improve the sensitivity of the system and increase the reliability and accuracy of the measurements. The most useful is to increase the signal to noise ratio, either by increasing the quality factor of the resonator, or by increasing the amount of absorption observed. Both of these can be achieved by performing the experiment at lower As the physical temperature of the system decreases, the temperatures. surface conductivity of the gold on the resonator's mirrors increases, which improves the quality factor of the resonances. Also, as the temperature decreases, the absorption per molecule of ammonia increases rapidly since  $\alpha \sim T^{-7/2}$ . At 183 K, the saturation pressure of ammonia is slightly above 20 Torr. Therefore, at 183 K, the same mixing ratio at which our previous experiments were performed  $(1.32 \times 10^{-2})$  can be achieved. At this temperature the Ben-Reuven model predicts that the absorption will increase by a factor of 3.5. We suggest that further experiments be performed at temperatures as low as 183 K. We feel that such experiments will make it possible to better evaluate the Ben-Reuven theory in the millimeter-wave range.

# V. OBSERVATIONS OF THE MICROWAVE EMISSION OF VENUS FROM 1,3 TO 3.6 CR

As discussed in Steffes (1986), our previous laboratory measurements of the microwave absorption of gaseous  $H_2SO_4$  under simulated conditions for the Venus atmosphere suggested that the 2-3 cm Venus emission spectrum would be especially sensitive to the subcloud abundance of gaseous  $H_2SO_4$ . Since no observations of the Venus emission in this wavelength range have ever been published, we conducted observations using the NRAO 140-foot radio telescope in order to search for spectral features related to gaseous  $H_2SO_4$  abundance. Also, since apparent temporal variations in the Venus microwave emission spectrum from 1.3 to 3.6 cm have occurred, it was felt that simultaneous measurements over that entire wavelength range would best serve our need to characterize both the magnitude and the shape of the microwave emission spectrum of Venus, with the ultimate goal of inferring abundances and distribution of the microwave absorbing constituents (predominantly  $SO_2$  and gaseous  $H_2SO_4$ ).

The observations were conducted by P. G. Steffes and J. M. Jenkins of Georgia Tech and by M. J. Klein of JPL. Observations of Venus and several calibration sources in the same approximate regions of the sky (DR21, P-2134 + 004, Jupiter, and 3C123) were made at 8.42 GHz (3.6 cm), 13.3 GHz (2.25 cm), 18.46 GHz (1.63 cm), and 22.2 GHz (1.35 cm) over a four day period from April 25 through April 28, 1987. Observations at 1.63 cm and 2.25 cm were made using the 140-foot diameter NRAO radio telescope at Greenbank, West Virginia. (The National Radio Astronomy Observatory is operated by Associated Universities, Inc. under contract with the National Science Foundation.) Simultaneous observations at 1.35 cm and 3.6 cm were made with the 64-meter

diameter DSN antenna at Goldstone, California. It should also be noted that over the entire month of April 1987, daily observations of the 3.6 cm Venus emission were also made with the 34-meter diameter DSN antenna, also located at Goldstone.

The dates of the observations were selected so that Venus would be close enough to superior conjunction so that it would appear as a very small source (approximately 12 arcseconds), but still far enough from the sun to avoid interference. This was done in order to minimize difficulties with beam size correction and source size correction. The times of observation were selected so as to maximize the elevation angle of the observing antennas, and thus minimize the effects of the earth's atmosphere on the observation. Since the microwave emission spectrum of Venus is a continuum, with little narrow line structure (due to the high atmospheric pressure), wide receiver bandwidths were used, which also served to increase the accuracy of the radiometric measurement. The bandwidths used were 20 MHz (8.42 GHz), 200 MHz (13.3 GHz), 250 MHz (18.46 GHz), and 20 MHz (22.2 GHz). Receiver system noise temperatures of 81 K or less were achieved at all frequencies further increasing the sensitivity of the measurement.

Calibration and study of the observed data is ongoing and will continue during the second half of the grant year. Effects such as the variation of receiving antenna aperture efficiency with hour angle, declination, and frequency have presented challenging calibration problems, but we believe our frequent observation of reference sources will allow correction for these effects. Preliminary calibration of the data has resulted in somewhat surprising findings. "First-look" results show a relatively high Venus brightness of 655 K at 3.6 cm, which monotonically decreases with decreasing

wavelength down to 520 K at 1.35 cm. This included an emission measurement of 565 K at 2.2 cm. There are two very noticeable aspects of these results. The first is the relatively high values measured for the brightness temperatures at all wavelengths. Such high values, especially at the shortest wavelengths, suggest a reduced SO2 abundance in the atmospheric region near the clouds. At longer wavelengths (3.6 cm), the higher brightness temperatures suggest increased opacity (possibly due to SO<sub>2</sub>) very low in the atmosphere. The second noticeable feature is the lack of the hoped-for dip in emission around A dip on the order of 50 K below the expected value was thought 2.2 cm. possible. However, since the magnitude of such a dip is directly related to the absorption coefficient of gaseous  $H_2SO_4$  at 2.2 cm, and since the error bars on our laboratory measurement at 2.2 cm were so large (a factor of two), it is not surprising that this spectral feature was not measured. However, this will now allow us to set an upper limit on average H2SO4 abundance in the Venus atmosphere.

A large amount of calibration and interpretive study of these observations has still yet to be completed, but we are hoping to soon travel to JPL and meet with Dr. Klein in order to complete initial analysis and begin preparing results for publication.

# VI. PUBLICATIONS AND INTERACTION WITH OTHER INVESTIGATORS

At the beginning of the current grant year, a paper was completed and accepted for publication in <u>Icarus</u>, describing results and applications of experiments performed during the previous year of Grant NAGW-533 (Steffes and Jenkins, 1987). This paper is described at length in Section I of this

report. More recently, we have submitted summaries of our laboratory measurements for inclusion in the twentieth issue of the Newsletter of Laboratory Spectroscopy for Planetary Science. Later this year, we hope to present our most recent results for the laboratory measurements of the millimeter-wave opacity of ammonia under Jovian conditions at the 19th Annual Meeting of the Division of Planetary Sciences of the American Astronomical Society (AAS/DPS Meeting). In a related meeting being held in conjunction with the DPS meeting, entitled, "Laboratory Measurements for Planetary Science," we will present a summary of our millimeter-wave laboratory activities, and their application to the interpretation of radio absorptivity data entitled, "Laboratory Measurements of the Millimeter-Wave Absorption from Gaseous Constituents under Simulated Conditions for the Outer Planets." Also at the AAS/DPS meeting, we hope to present results of our microwave measurements of the absorptivity of  $H_2O$  and  $CH_A$ , as well as initial results from the 1.3-3.6 cm observation of Venus. Support for travel to Pasadena for these meetings will be provided by Georgia Tech in support of planetary atmospheres (It should also be noted that partial support for travel to NRAO research. for the April 1987 observation was provided by Georgia Tech.) By the end of the current grant year, we hope to be able to submit papers on these same subjects to refereed journals for publication.

In addition to the observations of Venus conducted jointly with Dr. Michael J. Klein of JPL, more informal contacts have been maintained with groups at the California Institute of Technology (Dr. Duane O. Muhleman, regarding radio astronomical measurements of Venus opacity), at the Stanford Center for Radar Astronomy (V. Eshleman, regarding Voyager results for the outer planets, and laboratory measurements), and at JPL (Drs. Michael J.
Klein, Michael Janssen, and Samuel Gulkis, regarding radio astronomical observations of the outer planets, and A. J. Kliore, regarding the Pioneer-Venus Radioscience Program). We have also been active in the review of proposals submitted to the Planetary Atmospheres Program at NASA and as a reviewer of manuscripts submitted to <u>Icarus</u> and the <u>Journal of Geophysical</u> <u>Research</u>, for which Dr. Steffes is an Associate Editor. Another source of close interaction with other planetary atmospheres principal investigators has been Dr. Steffes' membership in the Planetary Atmospheres Management and Operations Working Group (PAMOWG). Travel support for attendance at PAMOWG meetings has been provided by Georgia Tech.

#### VII. CONCLUSION

During the first half of this current grant year, we have continued to conduct laboratory measurements of the microwave properties of atmospheric gases under simulated conditions for the outer planets. A most significant addition to this effort has been our capability to make such measurements at longer millimeter-wavelengths (7-10 mm). In the second half of this grant year, we hope to complete measurements of the millimeter-wave absorption from ammonia under simulated Jovian conditions. We will also further study the feasibility of measuring the microwave and millimeter-wave properties of phosphine (PH<sub>3</sub>) under simulated Jovian conditions, and will proceed with such measurements, if feasible. We will likewise continue to pursue further analysis and application of our laboratory results to microwave and millimeter-wave absorption data for the outer planets, such as Voyager Radio Occultation experiments and earth-based radio astronomical observations. We also intend to pursue the analysis of available multi-spectral microwave

opacity data from Venus, including data from our most recent radio astronomical observations in the 1.3-3.6 cm wavelength range and Pioneer-Venus Radio Occultation measurements at 13 and 3.6 cm, using our previous laboratory measurements as an interpretive tool. The timely publication of all of these results will be a high priority.

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IX. KEY FIGURES

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# Table I

Absorption Summary for 1.6 (	GHz
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Date	Temperature K	Mixing Ratio	Pressure (atm.)	α (dB/km) Measured	α (dB/km) Theoretical
9/25/86	206	$3.7 \times 10^{-3}$	6.1	1.273 +1.0	1.35
			4.1	0.0 +1.0	0.66
			2.0	0.0 <sup>±</sup> 1.0	0.18
9/17/86	193	$3.7 \times 10^{-3}$	6.1	1.83 +1.8	1.58
			4.1	$0.92 \begin{array}{c} +1.3 \\ -0.9 \end{array}$	0.78
			2.0	$0.46 \begin{array}{c} +1.4 \\ -0.5 \end{array}$	0.21

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## TABLE II

# Absorption Summary for 2.2 GHz

Date	Temperature K	Mixing Ratio	Pressure (atm.)	α (dB/km) Measured	α (dB/km) Theoretical
8/6/86	300	$3.7 \times 10^{-3}$	6.1	0.91 ±0.27	0.89
			4.1	0.36 +0.28 -0.27	0.42
			2.0	0.27 +0.28 -0.27	0.11
7/25/86	193	$3.7 \times 10^{-3}$	6.1	1.73 ±0.27	2.58
			4.1	0.91 ±0.18	1.23
			2.1	0.50 ±0.18	0.34
8/13/86	193	$3.7 \times 10^{-3}$	6.1	2.10 +0.27 -0.28	2.58
			4.1	0.82 ±0.23	1.23
			2.0	$0.37 \begin{array}{c} +0.18 \\ -0.19 \end{array}$	0.34
9/17/86	193	$3.7 \times 10^{-3}$	6.1	2.19 +0.22 -0.23	2.58
			4.1	1.28 +0.22 -0.23	1.23
			2.0	0.37 +0.22 -0.23	0.34
7/14/86	178	4.3 x 10 <sup>-4</sup>	6.1	0.18 +0.28 -0.18	0.36
			3.2	$0.18 \begin{array}{c} +0.22 \\ -0.13 \end{array}$	0.11

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# Table III

# Absorption Summary for Ammonia at 298K

-2 Mixing Ratio = 1.32 X 10

Date	Frequency GHz	Pressure (atm)	(dB/km) Measured	(db/km) Theoretical
5/19/87	36.27	2.0	107.3 <u>+</u> 73	66.72
5/19/87	36.29	1.0	54.6 <u>+</u> 73	20.13
5/20/87	36.29	2.0	127.5 <u>+</u> 73	66.72
5/20/87	36.29	1.0	0.0±55	20.13
5/19/87	38.43	2.0	73.0 <u>+</u> 73	53.68
5/19/87	38.43	1.0	36.6 <u>+</u> 73	15.43
5/20/87	38.43	2.0	54.6 <u>+</u> 73	53.68
5/20/87	38.43	1.0	54.6 <u>+</u> 73	15.43
5/19/87	39.15	2.0	72.9 <u>+</u> 82	50.21
5/19/87	39.15	1.0	0.0 <u>+</u> 82	14.22
5/20/87	39.15	2.0	72.9 <u>+</u> 36	50.21
5/20/87	39.15	1.0	0.1 <u>+</u> 36	14.22
5/19/87	40.62	2.0	54.8 <u>+</u> 73	44.27
5/19/87	40.62	1.0	27.4 <u>+</u> 73	12.24
5/20/87	40.62	2.0	18.3 <u>+</u> 36	44.27
5/20/87	40.62	1.0	0.0 <u>+</u> 18	12.24



Figure 1: Block diagram of atmospheric simulator, as configured for measurements of the microwave absorption from test gases under simulated conditions for the outer planets.



Figure 2: Diagram of Fabry-Perot Resonator



Figure 3: Block Diagram of Atmospheric Simulator



# Frequency, GHz



-36-



gure 5: Graph of Theoretical and Experimental Absorption for NH3 at Room Temperature

#### REPORT

#### TO THE

#### NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

## ANNUAL STATUS REPORT (Includes Semiannual Status Report #8)

for

#### GRANT NAGW-533

#### LABORATORY EVALUATION AND APPLICATION OF

#### MICROWAVE ABSORPTION PROPERTIES UNDER SIMULATED

#### CONDITIONS FOR PLANETARY ATMOSPHERES

Paul G. Steffes, Principal Investigator

February 1, 1987 through January 31, 1988

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#### I. INTRODUCTION AND SUPPARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often leads to significant misinterpretation of available opacity data. Steffes and Eshleman (1981) showed that under environmental conditions corresponding to the middle atmosphere of Venus, the microwave absorption due to atmospheric  $SO_2$  was 50 percent greater than that calculated from Van Vleck-Weiskopff theory. Similarly, results obtained for the microwave opacity from gaseous  $H_2SO_4$  under simulated Venus conditions, during the first two years of Grant NAGW-533, showed that not only was the opacity from  $H_2SO_4$  much greater than theoretically predicted, but that its frequency (wavelength) dependence was far different than that theoretically predicted (Steffes, 1985 and Steffes, 1986). More recently, measurements made by Steffes and Jenkins (1987), during the third year of Grant NAGW-533, have shown that the microwave opacity of gaseous ammonia (NH2) under simulated Jovian conditions does indeed agree with theoretical predictions at wavelengths longward of 1.3 cm. The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by both radio occultation experiments and radio astronomical observations, and over a

range of frequencies which correspond to those used in both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has been the goal of this investigation to conduct such measurements and to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profiles of constituents in those planetary atmospheres.

In some cases, new observations or experiments have been suggested by the results of the laboratory measurements. For example, this facility was initially developed, and then operated, in order to evaluate the microwave absorbing properties of gaseous sulfuric acid  $(H_2SO_4)$  under Venus atmospheric conditions. The results, obtained at 13.4 cm and 3.6 cm wavelengths, were applied to measurements from Mariner 5, Mariner 10, and Pioneer-Venus Radio Occultation experiments, to determine abundances of gaseous sulfuric acid in the Venus atmosphere, with accuracies exceeding those achieved with in-situ instruments (Steffes, 1985). Later efforts concentrated on making laboratory measurements of the microwave absorption from gaseous  $H_2SO_4$  at wavelengths from 1.3 to 22 cm under simulated Venus conditions. We applied these results to radio astronomical observations of Venus which have been made in the same wavelength range, in order to better model the structure of  $H_2SO_4$  and  $SO_2$ abundance in the Venus atmosphere, and to resolve temporal variations of their abundances on a planet-wide basis. The results of this effort were especially rewarding in that the unique frequency and pressure dependences measured for the absorption from gaseous  $H_2SO_4$  in these wavelength ranges has finally explained what were thought to be inconsistencies between measurements of the absorption in the Venus atmosphere at 13.3 and 3.6 cm wavelengths and those

obtained in the 1 to 2 cm wavelength range. Our laboratory measurements also suggested that a substantial variation in the Venus microwaye emission, related to the abundance of gaseous sulfuric acid, might exist near the 2.2 cm wavelength. Since no observations of the Venus emission at this wavelength have ever been published, we conducted observations of Venus using the 140foot NRAO telescope and the 64-meter DSN/Goldstone antenna in April 1987 to not only search for the presence of the predicted feature, but to use such a feature to determine a planet-wide average for sulfuric acid vapor abundance below the main cloud layer. The results of this observation are substantial in that they not only place limits on the abundance and spatial distribution of gaseous  $H_2SO_4$  and  $SO_2$ , but they also suggest some long term temporal variations for the abundance of these two gases. We hope to obtain further confirmation of these variations through reduction and analysis of recently obtained Pioneer-Venus radio occultation measurements. This observation and its results are discussed in Section V of this report. It should be noted that results of our laboratory measurements of the microwave absorption from gaseous  $H_2SO_4$  have also stimulated new studies of the theoretical basis for  $H_2SO_4$  opacity (see Section V).

An equally important activity for this grant year has continued to be laboratory measurements of the microwave and millimeter-wave properties of the simulated atmospheres of the outer planets and their satellites. As described in the previous <u>Annual Status Report</u> for Grant NAGW-533 (February 1, 1986 through January 31, 1987), initial laboratory measurements of the microwave opacity of gaseous ammonia (NH<sub>3</sub>) in a hydrogen/helium (H<sub>2</sub>/He) atmosphere, under simulated conditions for the outer planets were completed in September 1986. These measurements were conducted at frequencies from 1.3 GHz to 22 GHz

(wavelengths from 1.3 cm to 22 cm), at temperatures from 178 K to 300 K, and under total pressures reaching as high as 6 atmospheres. Such measurements have long been sought by a number of researchers working on inferring ammonia abundance profiles in Jovian atmospheres. Our measurements represented the first time that measurements of the microwave absorption of gaseous ammonia under simulated conditions for the outer planets had been conducted. The results of these measurements, and their effect on the interpretation of microwave opacity data from Jupiter, obtained both from Voyager radio occultation measurements made at 13 cm and 3.6 cm wavelengths and from radio astronomical observations in the 1.3 cm to 20 cm wavelength range, are discussed in a paper entitled "Laboratory Measurements of the Microwave Opacity of Gaseous Ammonia (NH<sub>2</sub>) Under Simulated Conditions for the Jovian Atmosphere," by Steffes and Jenkins (1987), which was recently published in Icarus (September 1987). These results showed that in the 1.38 to 18.5 cm wavelength range, the absorption from gaseous ammonia was correctly expressed by the modified Ben-Reuven lineshape as per Berge and Gulkis (1976). As a result, it became clear that the opacity which would be exhibited by a solar abundance of ammonia would be a factor of 1.5-2.0 below the opacity at the 2 to 6 Bar levels of the atmosphere as inferred from radio emission studies in the 6 to 20 cm wavelength range (see de Pater and Massie, 1985 and de Pater, 1986), and the opacity at the 1 Bar level measured by the Voyager 13 cm wavelength radio occultation experiment (see Lindal et al., 1981). This additional opacity, while most likely due to an overabundance of gaseous ammonia beyond the solar abundance, might also have been due to other gases or condensates, especially in the 2 to 6 Bar levels of the atmosphere.

As a result, in this current grant year, we have continued laboratory measurements of the microwave absorption and refraction from other potential microwave absorbers contained in the outer planets' atmospheres, including methane (CH<sub>4</sub>) and water vapor ( $H_2O$ ), as well as additional high-sensitivity measurements of the absorption from gaseous  $NH_3$  at 13.3 and 18.5 cm (results are presented in Section IV). It has also been brought to our attention by several researchers that significant uncertainties exist as to the actual absorption spectrum of gaseous ammonia at wavelengths shortward of 1 cm. It has been found by some (e.g., de Pater and Massie, 1985) that the observed millimeter-wave emission from Jupiter is inconsistent with the millimeter-wave absorption spectrum predicted using the modified Ben-Reuven line shape for In order to confirm this, we have developed a Fabry-Perot specammonia. trometer system capable of operation from 30 to 41 GHz (wavelengths from 7.3 to 10 mm). This system can be used at pressures up to 2 Bars and temperatures as low as 150 K, which corresponds closely to the conditions at altitudes in the Jovian atmosphere most responsible for the observed millimeter-wave absorption. A complete description of the millimeter-wave spectrometer is given in Section II.

We have used this spectrometer to complete laboratory measurements of the 7.5-9.3 mm absorption spectrum of ammonia, as well as additional measurements of  $CH_4$ . The results of these measurements (presented in Section IV) are substantive in that they show that neither the modified Ben-Reuven lineshape nor the Van Vleck-Weisskopf lineshape best describe the 7.5-9.3 mm (32-40 GHz) absorption from gaseous  $NH_3$  under simulated Jovian conditions. Since even larger variations from theoretically-derived opacity values can be expected at shorter millimeter-wavelengths (see de Pater and Massie, 1985), we hope to

pursue further laboratory measurements at short millimeter-wavelengths, especially near 3.2 mm (94 GHz), where a large number of observations of the emission from the outer planets have been made. A better knowledge of the millimeter-wave absorption properties of NH<sub>3</sub> is essential, not only to help better characterize the distribution and abundance of ammonia at high levels in Jovian atmospheres, but to make it possible to resolve the contributions from other absorbing constituents such as  $H_2S$  (see Bezard <u>et al.</u>, 1983). We also intend to investigate the further improvement of the sensitivity/accuracy of our existing systems (1.3 to 18.5 cm and 7.5 to 10 mm) through the integration of high temperature superconducting materials into the resonators. A more complete discussion of this proposed future activity is included in the accompanying proposal to NASA entitled, "Laboratory Evaluation and Application of Microwave Absorption Properties under Simulated Conditions for Planetary Atmospheres."

Of equal importance, however, will be the further analysis of microwave absorption data from Venus and the application of our laboratory results for the microwave absorption from gaseous  $SO_2$  and gaseous  $H_2SO_4$  in the Venus atmosphere to that data. As discussed in Section V, our recent measurements of the Venus microwave emission at 2.25 cm and 1.9 cm made with the NRAO 140foot telescope, and at 3.6 cm and 1.35 cm made with the DSN (Goldstone) 64meter antenna have shown that substantial spatial or temporal variations in the abundance and distribution of gaseous  $H_2SO_4$  and  $SO_2$  are likely to be occurring in the Venus atmosphere. An effective tool for evaluating spatial variations in the abundance of these microwave absorbing gases is the radio occultation studies which are conducted with the Pioneer-Venus orbiter. It is our hope to be active in the reduction and interpretation of the Pioneer-Venus

radio occultation data obtained in the Winter 1986/87 time period, both for determining spatial variations in sulfur-bearing gas abundances, and for determining temporal variations by comparison with previously obtained data. A more complete description of our proposed involvement in this activity is included in the accompanying proposal.

### II. THE GEORGIA TECH RADIO ASTRONOMY AND PROPAGATION (R.A.P.) FACILITY

The basic configuration of the planetary atmospheres simulator developed at Georgia Tech for use in measurement of the microwave absorptivity of gases under simulated conditions for planetary atmospheres is described at length in the previous Annual Status Report(s) for Grant NAGW-533. It is also discussed at length in Steffes (1986) and Steffes and Jenkins (1987). The updated simulator system, shown in Figure 1, is currently configured for simulations of the outer planets. Measurements of the microwave opacity and refractivity of test gas mixtures can be performed at frequencies from 1.6 GHz to 27 GHz (wavelengths from 1.2 cm to 18.5 cm) using two microwave resonators contained within a pressure chamber. While the pressure chamber itself is capable of containing pressures up to 10 atmospheres, and the temperature chamber is capable of achieving temperatures as low as 150 K, it has been found that the combination of high pressures and low temperatures can create a substantial problem with sealing the pressure chamber. The use of gaseous hydrogen  $(H_2)$ in the outer planets simulations has required the development of procedures and equipment for conducting such simulations. In addition, a thermocouple vacuum sensor is included in the system. This sensor not only allows accurate determination of pressure vessel evacuation, but it can also be used for accurately determining the abundances of microwave-absorbing test gases, which

are typically very small at low temperatures, due to low saturation vapor pressures.

The overall result has been a pressure vessel large enough to contain two microwave resonators, which is capable of maintaining 6 atmospheres of pressure at a temperature of 150 K, with an acceptably small leak ratio. While the range of pressures which can be tested was not quite as large as originally hoped, the resulting range of temperatures and pressures does represent the range over which nearly all of the microwave opacity in the Jupiter atmosphere has been observed, and thus is extremely useful in interpretation of microwave opacity data from Voyager I and II radio occultation experiments, as well as from earth based radio astronomical observations, and, in the future, opacity measurements to be made using the Galileo probe. Likewise, the pressure-temperature ranges measured are close enough to those over which microwave absorption or refraction has been measured in the atmospheres of Saturn, Titan, Uranus, and Neptune, so that accurate estimates of abundances of microwave-absorbing constituents in these atmospheres can also be made.

The most recent addition to the Georgia Tech Radio Astronomy and Propagation Facility has been a Fabry-Perot type resonator capable of operation between 30 and 41 GHz. As shown in Figure 2, the resonator consists of two gold plated mirrors separated by a distance of about 20 cm. The mirrors are contained in a T-shaped glass pipe which serves as a pressure vessel capable of withstanding over 2 atm of pressure. Each of the three open ends of the pipe is sealed with an O-ring sandwiched between the lip of the glass and a flat brass plate which is bolted to an inner flange. Electromagnetic energy is coupled both to and from the resonator by twin irises located on the

surface of one of the two mirrors. Two sections of WR-28 wavequide which are attached to the irises pass through the brass plate to the exterior of the pressure vessel. The end of each waveguide section is pressure-sealed by a rectangular piece of mica which is held in place by a mixture of rosin and As shown in Figure 3, one of these ends is connected to the sweep beeswax. oscillator through a waveguide section. A Ka-band (26-40 GHz) mixer is attached to the other section of wavequide and is coupled to the high resolution spectrum analyzer through a calibrated section of coaxial cable. The entire resonator, including its glass pressure envelope, is placed in the temperature chamber, which is a low-temperature freezer capable of operation down to 150 K. A network of stainless steel tubing and valves connects other components such as gas storage tanks, vacuum gauges, the pressure gauge, and the vacuum pump to the resonator assembly, so that each component may be isolated from the system as necessary. When properly secured, the system is capable of containing up to two atmospheres of pressure without detectable The sensitivities (minimum detectable opacities) achievable with leakage. this system are shown in Figure 4. It should be noted, however, that improvements of several orders of magnitude could be achieved if superconducting materials could be placed on the surfaces of the mirrors in the resonator.

## III. EXPERIMENTAL APPROACH

The approach used to measure the microwave absorptivity of test gases in an  $H_2/He$  atmosphere is similar to that used previously by Steffes and Jenkins (1987) for simulated Jovian atmospheres. At frequencies below 22 GHz (see Figure 1), the absorptivity is measured by observing the effects of the introduced gas mixture on the Q, or guality factor, of two cavity resonators

at particular resonances from 1.62 GHz to 21.8 GHz. At frequencies between 30 and 41 GHz, the changes in the Q of the numerous resonances of the Fabry-Perot resonator (see Figure 3) are related to the absorptivity of the test gas mixture at these frequencies. The changes in the Q of the resonances which are induced by the introduction of an absorbing gas mixture can be monitored by the high resolution microwave spectrum analyzer, since Q is simply the ratio of the cavity resonant frequency to its half-power bandwidth. For relatively low-loss gas mixtures, the relation between the absorptivity of the gas mixture and its effect on the Q of a resonance is straightforward:

$$\alpha \approx \left( \varrho_{\rm L}^{-1} - \varrho_{\rm C}^{-1} \right) \pi / \lambda \tag{3}$$

where  $\alpha$  is absorptivity of the gas mixture in Nepers km<sup>-1</sup>. (Note, for example, that an attenuation constant or absorption coefficient or absorptivity of 1 Neper km<sup>-1</sup> = 2 optical depths per km (or km<sup>-1</sup>) = 8.686 dB km<sup>-1</sup>, where the first notation is the natural form used in electrical engineering, the second is the usual form in physics and astronomy, and the third is the common (logarithmic) form. The third form is often used in order to avoid a possible factor-of-two ambiguity in meaning.)  $Q_{\rm L}$  is the quality factor of the cavity resonator when the gas mixture is present,  $Q_{\rm C}$  is the quality factor of the resonance in a vacuum, and  $\lambda$  is the wavelength (in km) of the test signal in the gas mixture.

The first experiments involved high-sensitivity measurements of the microwave absorption from gaseous  $NH_3$  under simulated conditions for the 2 to 6 Bar pressure range of the Jovian atmosphere in the 10 to 20 cm wavelength range. These measurements were undertaken in order to help better explain the

source of the microwave opacity at altitudes below the 2 atm pressure level in the Jovian atmosphere. (See Steffes and Jenkins, 1987, or the previous <u>Annual</u> <u>Status Report</u> for Grant NAGW-533, for further discussion.) These measurements were conducted at 1.62 GHz (18.5 cm) and 2.25 GHz (13.3 cm), and took advantage of special adjustments to the atmospheric simulator so as to maximize sensitivity in the 10 to 20 cm wavelength range.

Temperatures from 193 K to 300 K were used for the experiments, since lower temperatures would risk condensation of the gaseous NH3. When the pressure vessel reached thermal stability at the desired temperature, which was monitored using both the temperature sensors and the resonant frequencies of the system, a vacuum was drawn in the pressure vessel containing the resonators, and the bandwidth and center frequency of each of resonances was then measured. A valve was then opened which allowed the ammonia gas to enter the chamber, where 17 torr  $NH_3$  pressure was used. Measurements of the gaseous NH3 pressure were made with the high accuracy thermocouple vacuum gauge tubes which are shown in Figure 1. Next, 5.4 atm of hydrogen  $(H_2)$  and 0.6 atm of These gases were admitted to the chamber at a helium (He) were added. sufficiently slow rate so as not to significantly affect the temperature within the chamber. The result was an atmosphere with 6 atm total pressure composed of 90 percent hydrogen, 10 percent helium, and approximately 3730 ppm ammonia. The bandwidth of each resonance was then measured and compared with its value when the chamber was evacuated in order to determine the absorptivity of the gas mixture at 6 atm total pressure. The total pressure was then reduced by venting to 4 atm, and the bandwidths were again measured. Subsequent measurements were likewise made at 2 atm pressure. The pressure vessel was then evacuated and the bandwidths again measured so as to assure no

variation (either due to thermal shift or chemical reaction) of the Q's of the evacuated resonators had occurred. The measured changes of bandwidth (Q) were then used to compute the absorptivity of the gas mixture under the various pressure conditions.

This approach has the advantage that the same gas mixture is used for the absorptivity measurements at the various pressures. Thus, even though some small uncertainty may exist as to the mixing ratio of the initial mixture, the mixing ratios at all pressures are the same, and thus the uncertainty for any derived pressure <u>dependence</u> is due only to the accuracy limits of the absorptivity measurements, and not to uncertainty in the mixing ratio. (This assumes that the mixing ratio is small, so that foreign-gas broadening predominates, as is the case for our measurements.) Similarly, measurements of the frequency <u>dependence</u> of the absorptivity from the mixture would likewise be immune to any mixing ratio uncertainty, since foreign-gas broadening predominates.

The second major set of experiments involved measurements of the microwave absorption from methane (CH<sub>4</sub>) under simulated Jovian conditions at frequencies from 2.2 to 21.7 GHz. These experiments employed the same dualcavity atmospheric simulator as was used for the earlier ammonia experiments, shown in Figure 1. However, since methane absorption is very low, much larger methane mixing ratios were required to achieve detectable levels of absorption at these wavelengths. Likewise, since the methane absorption is inversely proportional to temperature, the lowest possible temperatures were used so as to maximize measurable absorption. In contrast to ammonia, the saturation vapor pressure of methane is quite large, thus risk of condensation was not a consideration. As a result, absorption from pure methane at pressure up to

6 Bars and at a temperature of 153 K was measured, as well as absorption from a 40% methane, 52% hydrogen, and 8% helium atmosphere under the same conditions of temperature and pressure. These measurements were made using resonances at 2.25 GHz (13.3 cm), 8.52 GHz (3.5 cm), and 21.7 GHz (1.38 cm).

The third set of measurements were made at the same frequencies, but involved measurements of the microwave absorption of water vapor under simulated Jovian conditions. Because of the relatively low saturation vapor pressure of water, experiments were conducted at 300 K, in order to obtain sufficient water vapor for the experiment. As with the previous experiments, the dual-cavity atmospheric simulator, shown in Figure 1, was used. A major difference, however, was the source of the test gas, in this case,  $H_2O$ . Since pressurized cannisters of water vapor are not possible at room temperature or lower, a flask of liquid H<sub>2</sub>O was used as the source of water vapor. Using techniques similar to those used by Steffes (1985 and 1986) for measurement of the microwave absorption from gaseous  $H_2SO_A$ , a precise quantity of water vapor was obtained. That is, the flask is filled with a precisely known volume of distilled water. A vacuum is then drawn in the pressure vessel containing the microwave resonantors, and the bandwidth and center frequencies of the resonances are then measured. A valve is then opened which allows the water vapor eluting from the flask to fill the evacuated pressure vessel (0.031  $m^3$  of open volume with the resonators in place) and reach vapor pressure equilibrium with the liquid  $H_2O$ . Note that all components which contact the water vapor are maintained at the same temperature as the flask, so as to avoid condensation.

As  $H_2O$  vapor fills the chamber, changes in the resonance center frequencies are observed. These changes are related to the  $H_2O$  vapor abundance. After approximately 10 minutes, the frequency shift ceases, as vapor pressure

equilibrium is reached. The valve to the reservoir flask is then closed, and 5.4 atm of  $H_2$  and 0.6 atm of He are admitted to the chamber containing the  $H_2O$ vapor. The bandwidth of each response is then measured and compared with its value when the chamber was evacuated in order to determine the absorptivity of the gas mixture at 6 atm total pressure. The total pressure is then reduced by venting, and the bandwidths are again measured. Subsequent measurements are likewise made at lower pressures in order to determine absorptivities at those pressures. The pressure vessel is then evacuated and the bandwidths again measured so as to assure no variation (either due to thermal shift or chemical reaction) of the Q's of the evacuated resonators has occurred. After the experiment is complete, the volume of the remaining liquid is measured and compared with the initial volume measured in order to determine the amount of  $H_2O$  vapor present in the gas mixture tested. As with the anmonia experiments, this approach has the advantage that the same gas mixture is used for the absorptivity measurements at the various pressures.

The fourth set of measurements has involved the millimeter-wave absorption from gaseous ammonia  $(NH_3)$  in a 90%  $H_2/10$ % He atmosphere. Initial measurements were conducted at room temperature, but the majority of measurements have been conducted at a temperature of 203 K. While even lower temperatures could be achieved, the need to avoid the risk of ammonia condensation kept our operating temperatures relatively high. As in the previous experiments at lower frequencies, the bandwidth and center frequencies of each of several resonances between 32 and 41 GHz were measured in a vacuum. Next, 28 torr of gaseous ammonia is added to the system. The pressure of the ammonia gas is measured with the high-accuracy thermocouple vacuum gauge, as shown in Figure 3.

In addition, the ammonia abundance can be monitored by measuring refractivity of the introduced gas. Since the index of refraction (relative to unity) is proportional to the ammonia gas abundance, the ability of the system to accurately measure refractivity (through measurement of the frequency shift of resonances) can be used to infer the relative vapor abundance or pressure. Note that it is not yet possible to use this approach for the accurate determination of absolute NH<sub>3</sub> pressure since accurate refractivity data for the 7.3 to 10 mm wavelength range is not available. In fact, by using our thermocouple vacuum gauge, we have made measurements of the density-normalized refractivity of gaseous ammonia at 39 GHz, and found it to be 8.8 x  $10^{-17}$ N-units/molecule/cm, which is nearly 8 times the value at optical wavelengths.

Next, 1.8 atm of hydrogen  $(H_2)$  and 0.2 atm of helium (He) are added to the chamber, bringing the total pressure to 2 atm. The bandwidth of each resonance is then measured and compared with its value when the chamber was evacuated in order to determine the absorptivity of the gas mixture at 2 atm total pressure. The total pressure is then reduced, by venting, to 1 atm, and the bandwidths are again measured. Finally, the pressure vessel is again evacuated and the bandwidths again measured so as to assure no variation of the Q's of the evacuated resonator has occurred. As with the previous measurements, the measured changes of bandwidths (Q's) can then be used to compute the absorptivity of the gas mixtures at each of the resonant frequencies.

In all four types of the measurements described, the amount of absorption being measured is extremely small. Thus, any errors in measurements of (or other changes in) the apparent bandwidth of the resonances, not caused by the absorbing gases, could lead to significant errors in the absorption

measurement. The contribution of instrumental errors and noise-induced errors on such absorptivity measurements have been discussed at length in Steffes and Jenkins (1987). However, because our latest measurements represent such small percentage changes in bandwidth, another instrumental source of error which we refer to as <u>dielectric</u> loading becomes a concern.

As can be seen in Figures 1 and 3, the resonators, which operate as bandpass filters, are connected to a signal source (the microwave sweep oscillator) and to a signal receiver (the high-resolution spectrum analyzer). The "Q" of the resonator, which is defined as the ratio of energy stored in the resonator to the energy lost per cycle, equals the ratio of resonant center frequency to resonance half-power bandwidth. It is not surprising, therefore, that the stronger the coupling between the resonator and the spectrum analyzer or sweep oscillator, the lower will be the Q of the resonance, since more energy will be lost per cycle through the cables or waveguide connecting the resonator to the spectrum analyzer and sweep oscillator. For this reason, we have always designed our resonators (both the coaxially-coupled cylindrical cavity resonators used below 25 GHz and the waveguide-coupled Fabry-Perot resonator used above 30 GHz) with minimal coupling, so as to maximize Q and to minimize the changes in Q that might result from changes in coupling that occur when gases are introduced into the resonators. It should be noted that these changes in coupling, which are due to the presence of the test gas mixtures, are not related to the absorptivity of the gases, but rather to the dielectric constant or permittivity of the test gas mixtures. (Bence, the term "dielectric loading.")

We have always strived to design the coupling elements of the resonators so that the changes in lossless test gas abundances (and resulting changes in

dielectric constant) had little or no effect on the Q of the resonator as measured in the system. This has been no small feat in that alight imperfections in resonators, cables, coupling loops, or waveguides can make the apparent Q of the resonator appear to vary with the abundance of such lossless gases. It has now become a standard part of our experimental procedure to repeat absorption measurements for gas mixtures in which the absorbing gas is a minor constituent, without the absorbing gas present. For example, after measurements were made of the microwave and millimeter-wave absorption from ammonia as a minor constituent in an H<sub>2</sub>/He atmosphere, measurements of the apparent absorption of the H<sub>2</sub>/He atmosphere without the ammonia gas were made. Since, for the pressures and wavelengths involved, the H<sub>2</sub>/He atmosphere is essentially transparent, no absorption was expected. If any apparent absorption was detected, "dielectric loading," or a change in coupling due to the dielectric properties of the gases, was indicated.

Initially, if any evidence of dielectric loading existed, the experiments were terminated and the apparatus disassembled, including pressure seals. The cables and coupling loops were then readjusted, and the system reassembled and tested again. The entire procedure was repeated until the dielectric loading effect was eliminated or minimized. If some small variation in the resonant Q or bandwidth due to the presence of the non-absorbing gases still remained, it was added to the uncertainty or error bars for each experiment. More recently however, we have found that the effects of dielectric loading are additive, in that they add to the apparent changes of resonator bandwidth caused by the absorbing gases. Thus, as long as the effects of dielectric loading are not time variable, they can be removed by using the measured value of the Q of a resonance with the non-absorbing gases present (instead of the Q of the resonance in a vacuum) for the quantity  $Q_c$  in equation (3).

## IV. RESULTS OF LABORATORY MEASUREMENTS AND THEIR APPLICATION

As described in Section III, the first experimental sequence involved measurements of the microwave absorption from gaseous ammonia  $(NH_3)$  in a 90% hydrogen/10% helium atmosphere at the frequencies 1.62 GHz (18.5 cm) and 2.25 GHz (13.3 cm). These experiments were conducted with an ammonia mixing ratio of 3730 ppm, at temperatures as low as 193 K, with total pressures up to 6 atm. The results of these measurements are included in Figure 5 along with all of our previous absorptivity measurements for such an ammonia mixture at 193 K. Also shown are plots of the theoretically-derived absorption spectra for such gas mixtures at these three pressures. These theoretically-derived NH3 absorption spectra were calculated as per de Pater and Massie (1985) and Berge and Gulkis (1976), both of which employ a Ben-Reuven line shape which has been modified so as to be consistent with the laboratory results of Morris and Parsons (1970), in which the absorption from  $NH_3$  in a high-pressure  $H_2/He$ atmosphere at room temperature and at 9.58 GHz was measured. Our theoretical spectra have been adjusted for the temperature, pressure, and mixing ratio conditions of our experiment. This mathematical expression for the absorption of ammonia was implemented in a BASIC program for which partial pressures from  $H_2$ , He, and  $NH_3$ , as well as frequency and temperature, were adjustable This program was used to calculate the theoretical values for variables. absorption given in Tables I and II and in Figures 5 and 6.

Inspection of Figure 5 shows that the laboratory data agree surprisingly well with the theoretical predictions for  $NH_3$  opacity made using the Ben-Reuven lineshape, and are, likewise, consistent with the results of Morris and Parsons (1970). The key result of our work has been the finding that the

modified Ben-Reuven lineshape appears to correctly describe the shape of the absorption spectrum from gaseous ammonia in a hydrogen/helium atmosphere in the 1.3 to 18.5 cm wavelength range even under the temperature-pressure conditions characteristic of the Jovian atmosphere. This finding answers some questions and raises others. For example, both de Pater and Massie (1985) and West et al. (1986) recognized that, based on interpretation of the Jovian emission spectrum in the 10 to 20 cm wavelength range using the theoreticallyderived absorption spectrum from ammonia, opacity at pressure levels greater than 2 atm had to exceed the amount which would be caused by the solar abundance of ammonia. As a result, both sets of authors concluded that either the theoretical lineshape was incorrect and understated the opacity of ammonia at these wavelengths by a factor of 1.5-2.0, or the ammonia abundance was greater than its solar abundance by a factor of 1.5-2.0 at pressures greater than 2 atm, or that an extra opacity source, possibly  $H_2O$  condensate, was Thus, since our measurements of the ammonia opacity at 13.3 cm present. (2.25 GHz) agree quite closely with the theoretically-derived values for opacity, and since our newest results at 18.5 cm (1.62 GHz) are likewise consistent with the theoretical lineshape, it would appear that either the ammonia abundance is greater than solar by a factor of 1.5-2.0 in the deeper layers of the Jovian atmosphere, or that an extra opacity source is present.

In an attempt to resolve this question, we have made measurements of the microwave opacity of two other Jovian atmospheric constituents: methane and water vapor. The methane experiment was conducted as described in Section III, and not surprisingly, the microwave absorption was very small. Measurements were made at 153 K of the microwave absorption from a pure methane atmosphere at 6 atm pressure, and from a 40% methane, 52% hydrogen,

and 8% helium atmosphere at 6 atm pressure. No absorption was measured at the 2.25 GHz (13.3 cm) or 8.52 GHz (3.5 cm) resonances. Some absorption was measured at the 21.7 GHz (1.38 cm) resonance, but its statistical significance is uncertain since the levels measured were below the error bars for the system. (For the pure methane, the measured absorption was  $11.4 \pm 17$  dB/km.) For the 40% methane mixture, the measured absorption was  $9.4 \pm 18$  dB/km.) We intend to further study the theoretically-derived methane absorption spectrum for purposes of comparison and to determine whether further laboratory measurements at higher frequencies might be useful. However, the results strongly indicate that methane absorption in the 3-13 cm wavelength range is negligible in the 2-6 Bar pressure range of Jupiter's atmosphere, and cannot account for the additional opacity required in that altitude range.

Measurements of the microwave absorption from water vapor were conducted at 2.25 GHz, 8.52 GHz, and 21.7 GHz at 297 K in a 90%  $H_2/10$ % He atmosphere as described in Section III. A water vapor mixing ratio of 3509 ppm was used, and total pressures of 6, 4, and 2 atm were employed. None of the measurements detected statistically significant opacity. (System thresholds were 0.45 dB/km, 0.73 dB/km, and 36 dB/km at 2.25 GHz, 8.52 GHz, and 21.7 GHz, respectively.) Thus our results for the microwave opacity from water vapor (1.38 to 13.3 cm) under simulated Jovian conditions are consistent with the theoretically-based expression for opacity derived by Goodman (1969). Furthermore, our results indicate that, like methane, water vapor cannot account for the additional opacity required in the 2-6 Bar pressure range of Jupiter's atmosphere. Thus, all three of these measurements further strengthen the case for an ammonia abundance between 1.5 and 2.0 times larger than solar abundance.

Initial measurements of the 7.3-8.3 mm absorptivity from  $NH_3$  in a hydrogen/helium atmosphere were conducted at 298 K as described in Section III, with an ammonia mixing ratio of 1.32 x  $10^{-2}$ , at pressures of 1 and 2 Bars. An examination of the experimental results revealed that because of large error bars, we cannot determine whether the modified Ben-Reuven lineshape best describes the absorption profile of gaseous ammonia shortward of 1 cm. However, the consistency of our results at 2 atm suggested that the modified Ben-Reuven lineshape understates the actual absorptivity of ammonia in this range. With the exception of one measurement at 40.62 GHz, all measurements were greater than those predicted by the Ben-Reuven theory.

Results of measurements of the 32 to 40 GHz (7.5 to 9.2 mm) absorptivity of gaseous ammonia under simulated Jovian conditions (203 K) are shown in Table III and Figure 6. These measurements were made using a 90% hydrogen/ 10% helium atmosphere with a total pressure of 2 Bars. The ammonia mixing ratio was .018. With this mixing ratio, temperatures as low as 190 K could have been used before saturation would have become a problem, but difficulties with one of the compressors in our ultra-cold freezer system required our operating at a slightly higher temperature. (Funding is currently being procurred from within Georgia Tech to provide needed repairs to the freezer, in support of planetary atmospheres research at Georgia Tech.) Also, shown in Figure 6 are solid lines which represent the theoretically-computed opacity using the Van Vleck-Weisskopf lineshape (upper line) and the modified Ben-Reuven lineshape (lower line). The Van Vleck-Weisskopf calculation was performed using linewidths and line intensities as per Wrixon et al. (1971). The Ben-Reuven calculation was made as per Berge and Gulkis (1976), as earlier described.

Inspection of the results in Figure 6 shows that neither lineshape correlates well with all of the observed points. Except for one data point, the opacity measurements between 38 and 40 GHz appear to be in best agreement with the modified Ben-Reuven lineshape, whereas between 35 and 37 GHz, the opacity measurements appear to agree best with the Van Vleck-Weisskopf lineshape. Such results are not surprising in that other researchers, such as de Pater and Massie (1985), have found that in order to best explain the 1-10 mm Jupiter emission spectrum, a different sort of lineshape was needed to characterize the ammonia opacity. For this purpose, de Pater and Massie developed two modified Van Vleck-Weisskopf lineshape characterizations for the ammonia opacity in a hydrogen/helium atmosphere. Since even larger variations from either the modified Ben Reuven formulation or the Van Vleck-Weisskopf formulation for ammonia opacity are expected at shorter millimeter-wavelengths, we hope to pursue further laboratory measurements, especially near 3.2 mm (94 GHz), where a larger number of observations of the emission from the outer planets have been made.

A better knowledge of the millimeter-wave absorption properties of  $NH_3$  is essential, not only to help better characterize the distribution and abundance of ammonia at high levels in Jovian atmospheres, but to make it possible to resolve the contributions from other absorbing constituents such as  $H_2S$  (see Bezard <u>et al.</u>, 1983). Our goal to better characterize the millimeter-wave absorption spectrum of ammonia will not only involve increasing the range of frequencies over which measurements are made, but to increase the sensitivity of the measuring systems. There are several ways that we may improve the sensitivity of the system and increase the reliability and accuracy of the measurements. The most useful is to increase the signal to noise ratio,
either by increasing the quality factor of the resonator, or by increasing the amount of absorption observed. Both of these can be achieved by performing the experiment at lower temperatures. As the physical temperature of the system decreases, the surface conductivity of the gold on the resonator's mirrors increases, which improves the quality factor of the resonances. However, lowering the operating temperature below 190 K could cause the gaseous ammonia to condense. The only way to further improve the quality factor would be to place superconducting materials on the mirror surfaces. We intend to pursue the possible integration of superconducting materials into our existing systems (microwave: 1.3-18.5 cm and millimeter-wave: 7.5-10 mm). A more complete discussion of this is included in the accompanying proposal, "Laboratory Evaluation and Application of Microwave Absorption Properties under Simulated Conditions for Planetary Atmospheres."

### V. OBSERVATIONS OF THE MICROWAVE EMISSION OF VENUS FROM 1.3 TO 3.6 CR

As discussed in Steffes (1986), our previous laboratory measurements of the microwave absorption of gaseous  $H_2SO_4$  under simulated conditions for the Venus atmosphere suggested that the 2-3 cm Venus emission spectrum would be especially sensitive to the subcloud abundance of gaseous  $H_2SO_4$ . Since no observations of the Venus emission in this wavelength range have ever been published, we conducted observations using the NRAO 140-foot radio telescope in order to search for spectral features related to gaseous  $H_2SO_4$  abundance. Also, since apparent temporal variations in the Venus microwave emission spectrum from 1.3 to 3.6 cm have occurred, it was felt that simultaneous, or nearly simultaneous, measurements over that entire wavelength range would best

serve our need to characterize both the magnitude and the shape of the microwave emission spectrum of Venus, with the ultimate goal of inferring abundances and distribution of the microwave absorbing constituents (predominantly  $SO_2$  and gaseous  $H_2SO_4$ ).

The observations were conducted by P. G. Steffes and J. M. Jenkins of Georgia Tech and by M. J. Klein of JPL. Observations of Venus and several calibration sources in the same approximate regions of the sky (DR21, 3C454.3, P-2134 + 004, Jupiter, and 3C123) were made at 8.42 GHz (3.6 cm), 13.3 GHz (2.25 cm), 18.46 GHz (1.63 cm), and 22.2 GHz (1.35 cm) over a four day period from April 25 through April 28, 1987. Observations at 1.63 cm and 2.25 cm were made using the 140-foot diameter NRAO radio telescope at Greenbank, West Virginia. (The National Radio Astronomy Observatory is operated by Associated Universities, Inc. under contract with the National Science Foundation.) Simultaneous observations at 1.35 cm and 3.6 cm were made with the 64-meter diameter DSN antenna at Goldstone, California. It should also be noted that over the entire month of April 1987, daily observations of the 3.6 cm Venus emission were also made with the 34-meter diameter DSN antenna, also located at Goldstone.

The dates of the observations were selected so that Venus would be close enough to superior conjunction so that it would appear as a very small source (approximately 12 arcseconds), but still far enough from the sun to avoid interference. This was done in order to minimize difficulties with beam size correction and source size correction. The times of observation were selected so as to maximize the elevation angle of the observing antennas, and thus minimize the effects of the earth's atmosphere on the observation. Since the microwave emission spectrum of Venus is a continuum, with little narrow line

structure (due to the high atmospheric pressure), wide receiver bandwidths were used, which also served to increase the accuracy of the radiometric measurement. The bandwidths used were 20 MHz (8.42 GHz), 200 MHz (13.3 GHz), 250 MHz (18.46 GHz), and 20 MHz (22.2 GHz). Receiver system noise temperatures of 81 K or less were achieved at all frequencies further increasing the sensitivity of the measurement.

Initial calibration and study of the observed data is complete and we hope to continue further analysis during the next grant year. Effects such as the variation of receiving antenna aperture efficiency with hour angle, declination, and frequency have presented challenging calibration problems, but we believe our frequent observations of reference sources have provided proper correction for these effects. Calibration of the data has been a very time consuming activity, but has resulted in some surprising findings. The results, shown in Figure 7, show a relatively high Venus brightness of 662 K at 3.6 cm, which almost monotonically decreases with decreasing wavelength down to 523 K at 1.63 cm. This included an emission measurement of 565 K at 2.2 cm. Between 1.63 and 1.35 cm, the emission is relatively constant, with a 1.35 cm value of 519 K. Also shown in Figure 7 are computed emission spectra for Venus (Janssen, personal communication). The upper line represents the disk temperature computed by assuming that the only source of microwave opacity in the atmosphere is CO2, and uses the CO2 opacity results from Ho, Kaufman, and Thaddeus (1966) and the Venus temperature-pressure profile from Seiff et al. (1980). The middle line represents the disk temperature computed as above, but also includes the effects of the opacity of a 10 km deep layer of gaseous  $H_2SO_4$  (altitudes from 42 to 52 km) with a mixing ratio of 1 ppm. (The lower line includes the effects of 20 km deep layer, i.e., altitudes from 32 to 52 km).

The opacity from the gaseous sulfuric acid has been newly computed by adding contributions from over 11,800 resonant lines, and by assuming the Van Vleck-Weisskopf lineshape with a broadening parameter of 3 MHz/Torr for all lines. This new attempt at developing a theoretical formulation for the microwave opacity of gaseous sulfuric acid were stimulated by our previous laboratory results (Steffes, 1986). Typical results for the microwave opacity spectrum of gaseous sulfuric acid using this formulation are shown in Figure 8 (Janssen, personal communication) along with results of laboratory measurements under the same conditions (CO<sub>2</sub> atmosphere, P = 1 Bar, T = 575 K,  $H_2SO_4$ mixing ratio = 0.4%). Comparison of the two results shows a striking correlation in that a peak in opacity occurs near the 2 cm wavelength. Janssen's theoretical formulation differs from the laboratory results in that it indicates an opacity which is greater by a factor of 2 to 3 at the peak, and nearly an order of magnitude greater at longer wavelengths. Some of this apparent variation may be due to mixing ratio uncertainties in the laboratory measurement, since the assumed dissociation factor for  $H_2SO_4$  into  $SO_3$  and  $H_2O_4$ was 0.47, which has never been confirmed by laboratory measurement. This variation may also be partially due to the use of the 3 MHz/Torr broadening parameter for all resonant lines in the theoretical calculation. Another notable difference is the apparent width of the absorptivity peak near 2 cm. Since the laboratory measurements were made only at specific frequencies, the inferred absorption spectrum (solid curve) was developed based on an arbitrary shape. The shape of the curve developed by Janssen, therefore, is still consistent with our results, but it will also affect the range of frequencies or wavelengths over which a depressed disk brightness temperature might be expected.

There are several notable aspects to our results (Figure 7) for the microwave emission from Venus. The first is the relatively high values measured for the brightness temperatures at all wavelengths. Such high values, especially at the shortest wavelengths, suggest a reduced  $SO_2$ abundance in the atmospheric region near the clouds. This may be suggestive of substantial temporal variation in Venus SO2 abundance since the Pioneer-Venus probe entry. At longer wavelengths (3.6 cm), the higher brightness temperatures suggest increased opacity (possibly due to SO2) very low in the atmosphere or some variation in surface emission with planetary phase. The second noticeable feature is the nature of the hoped-for dip in emission around 2 cm. A dip on the order of 50 K below the expected value over a narrow wavelength range was thought possible. Instead, we have observed a much shallower dip over a wider wavelength range. This is consistent with the theoretically-derived absorption spectrum of Janssen. Moreover, the shallowness of the dip suggests an average gaseous  $H_2SO_4$  abundance below the clouds of between 1 and 5 ppm, depending on whether the laboratory-based or the theoretically-based values for  $H_2SO_4$  opacity are employed. Additionally, variations in vertical distribution may allow peak abundances which are even larger (15-20 ppm).

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It should also be noted that our earth-based observations of the Venus microwave emission are inherently weighted toward the equatorial zones. Thus, variations in gaseous sulfuric acid abundance with latitate are not being resolved, nor are contributions from the polar regions. However, our results suggest that the inferred microwave opacity appears to be lower than that inferred from radio occultation measurements in the 1970s and early 1980s (e.g., Cimino, 1982). These may be suggestive of either large scale temporal

variations in the abundance of gaseous  $H_2SO_4$  or large spatial variation between abundances in polar regions (as inferred from radio occultations) and those in equatorial zones (as inferred from our emission measurements). One important tool for determining which effect is occurring would be reduction of the microwave data from the 1986-87 Pioneer-Venus radio occultation measurements. This data was taken over a wide range of latitudes and could be critical for determining whether temporal variations or spatial variations in gaseous  $H_2SO_4$  abundance could be occurring. Thus, we have discussed with Dr. Arvydas J. Kliore (P-V Radioscience Leader) the possibility of our group obtaining the currently unreduced data and (working at JPL) reducing the data to obtain absorptivity profiles for the 1986-87 epoch. A more complete description of our proposed activity is included in the accompanying proposal.

### VI. PUBLICATIONS AND INTERACTION WITH OTHER INVESTIGATORS

At the beginning of the current grant year, a paper was completed and accepted for publication in <u>Icarus</u>, describing results and applications of experiments performed during the previous year of Grant NAGW-533 (Steffes and Jenkins, 1987). This paper is described at length in Section I of this report. More recently, we have submitted summaries of our laboratory measurements for inclusion in the twentieth issue of the <u>Newsletter of Laboratory</u> <u>Spectroscopy for Planetary Science</u>. In November, we presented our most recent results for the laboratory measurements of the millimeter-wave opacity of ammonia under Jovian conditions at the 19th Annual Meeting of the Division of Planetary Sciences of the American Astronomical Society (AAS/DPS Meeting) in a paper by Joiner <u>et al</u>. (1987)\*. In a related meeting held in conjunction with \*Abstract attached--See appendices.

the DPS meeting, entitled, "Laboratory Measurements for Planetary Science," we presented a summary of our millimeter-wave laboratory activities, and their application to the interpretation of radio absorptivity data entitled, "Laboratory Measurements of the Millimeter-Wave Absorption from Gaseous Constituents under Simulated Conditions for the Outer Planets" (Steffes, 1987). Also at the AAS/DPS meeting, we presented results of our microwave measurements of the absorptivity of  $H_2O$  and  $CH_A$  (Jenkins and Steffes, 1987),\* as well as initial results from the 1.3-3.6 cm observation of Venus (Steffes et al., 1987).\* Support for travel by Professor Steffes to Pasadena for both these meetings (and the earlier trip to JPL for analysis of the Venus microwave observations) was provided by Georgia Tech in support of planetary atmospheres research. (It should also be noted that partial support for travel to NRAO for the April 1987 observation was provided by Georgia Tech.) By the end of January (1988), we hope to be able to submit papers on these same subjects to refereed journals (Icarus and Astrophysical Journal) for publication.

In addition to the observations of Venus and analysis work conducted jointly with Dr. Michael J. Klein of JPL, we have also worked closely with Dr. Michael A. Janssen of JPL regarding models for the Venus atmosphere, interpretation of microwave emission measurements, and theoretical models for the absorption spectrum of  $H_2SO_4$ . We have also had productive discussions with Dr. Arvydes J. Kliore regarding our proposed future involvement with the reduction and interpretation of data from recent Pioneer-Venus Radio Occultation Studies. More informal contacts have been maintained with groups at the California Institute of Technology (Dr. Duane O. Muhleman, regarding radio astronomical measurements of Venus opacity), at the Stanford Center for \*Abstracts attached--See Appendices

Radar Astronomy (V. R. Eshleman, G. L. Tyler, and D. P. Hinson, regarding Voyager results for the outer planets, and laboratory measurements), and at JPL (Drs. Robert Poynter and Samuel Gulkis, regarding radio astronomical observations of the outer planets and Venus). We have also been active in the review of proposals submitted to the Planetary Atmospheres Program at NASA and as a reviewer of manuscripts submitted to <u>Icarus</u> and the <u>Journal of Geophysical Research</u>, for which Dr. Steffes is an Associate Editor. Another source of close interaction with other planetary atmospheres principal investigators has been Dr. Steffes' membership in the Planetary Atmospheres Management and Operations Working Group (PAMOWG). Travel support for attendance at PAMOWG meetings has been provided by Georgia Tech.

#### VII. CONCLUSION

Over this past grant year, we have continued to conduct laboratory measurements of the microwave properties of atmospheric gases under simulated conditions for the outer planets. A most significant addition to this effort has been our capability to make such measurements at longer millimeterwavelengths (7-10 mm). In the future, we hope to extend such measurements to even shorter millimeter-wavelengths. We will likewise continue to pursue further analysis and application of our laboratory results to microwave and millimeter-wave absorption data for the outer planets, such as results from Voyager Radio Occultation experiments and earth-based radio astronomical observations. We also intend to pursue the analysis of available multispectral microwave opacity data from Venus, including data from our most recent radio astronomical observations in the 1.3-3.6 cm wavelength range and Pioneer-Venus Radio Occultation measurements at 13 and 3.6 cm, using our

previous laboratory measurements as an interpretive tool. The timely publication of all of these results will be a high priority.

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IX. KEY FIGURES

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Table .
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Date	Temperature K	Mixing Ratio	Pressure (atm.)	a (dB/km) Measured	α (dB/km) Theoretical
<b>9</b> /25/86	206	$3.7 \times 10^{-3}$	6.1	1.273 +1.0	1.35
			4.1	0.0 +1.0	0.66
			2.0	0.0 ±1.0	0.18
9/17/86	193	$3.7 \times 10^{-3}$	6.1	1.83 +1.8	1.58
			4.1	0.92 + 1.3 - 0.9	0.78
			2.0	$0.46 \begin{array}{c} +1.4 \\ -0.5 \end{array}$	0.21

Absorption Summary for 1.6 GHz

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### TABLE II

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### Absorption Summary for 2.2 GHz

Date	Temperature K	Mixing Ratio	Pressure (atm.)	α (dB/km) Measured	α (dB/km) <u>- Theoretical</u>
<b>8/6/8</b> 6	300	$3.7 \times 10^{-3}$	6.1	0.91 ±0.27	0.89
-			4.1	0.36 +0.28 -0.27	- 0.42
			2.0	0.27 +0.28 -0.27	0.11
7/25/86	193	$3.7 \times 10^{-3}$	6.1	1.73 ±0.27	2.58
			4.1	<b>0.91</b> ±0.18	1.23
			2.1	0.50 ±0.18	0.34
8/13/86	193	$3.7 \times 10^{-3}$	6.1	2.10 +0.27 -0.28	2.58
			4.1	0.82 ±0.23	1.23
			2.0	$0.37 \begin{array}{c} +0.18 \\ -0.19 \end{array}$	0.34
9/17/86	193	$3.7 \times 10^{-3}$	6.1	2.19 +0.22 -0.23	2.58
			4.1	1.28 +0.22 -0.23	1.23
			2.0	0.37 +0.22 -0.23	0.34
7/14/86	178	4.3 x 10 <sup>-4</sup>	6.1	0.18 + 0.28 - 0.18	0.36
			3.2	0.18 + 0.22 - 0.13	0.11

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### TABLE III

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### Absorption Summary for Ammonia at 203K

### Mixing Ratio = 0.0184

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Date	Frequency (GHz)	Pressure (atm)	(dB/km) Measured	(dB/km) VVW	(dB/km) BR
10/9/87	32.15	2	327.6+227	462.6	343.5
10/9/87	32.15	1	109.3 <u>+</u> 118	173.9	130.2
10/13/87	32.15	2	382.2+227	462.6	343.5
10/13/87	32.15	1 -	109.1 <u>+</u> 118	173.9	130.2
9/24/87	36.24	2	364.2 <u>+</u> 82	351.4	234.1
9/24/87	36.24	1	91.2 <u>+</u> 73	100.2	73.5
10/9/87	36.24	2	327.8 <u>+</u> 227	351.4	234.1
10/9/87	36.24	1	109.3 <u>+</u> 118	100.2	73.5
10/13/87	36.24	2	273.3 <u>+</u> 91	351.4	234.1
10/13/87	36.24	1	36.5 <u>+</u> 73	100.2	73.5
9/24/87	36.95	2	327.7 <u>+</u> 82	336.8	220.1
9/24/87	36.95	1	63.7 <u>+</u> 63	93.9	67.7
10/9/87	38.99	2	255.0 <u>+</u> 109	300.8	182.1
10/9/87	38.99	1	72.9 <u>+</u> 109	87.0	53.4
10/13/87	38.99	2	164.0 <u>+</u> 82	300.B	182.1
10/13/87	38.99	1	72.9 <u>+</u> 72	87.0	53.4
9/24/87	39.13	2	291.2 <u>+</u> 82	298.8	184.6
9/24/87	39.13	1	109.1 <u>+</u> 64	86.8	45.5
10/9/87	39.66	2	145.6 <u>+</u> 82	289.6	172.5
10/9/87	39.66	1	54.6 <u>+</u> 54	83.1	50.2
10/13/87	39.66	2	164.0 <u>+</u> 82	289.6	172.5
10/13/87	39.66	1	18.3 <u>+</u> 73	83.1	50.2
9/24/87	37.86	2	182.2 <u>+</u> 54	288.0	174.8
9/24/87	39/86	1	72.9 <u>+</u> 54	81.3	50.6



Figure 1: Block diagram of atmospheric simulator, as configured for measurements of the microwave absorption from test gases under simulated conditions for the outer planets.



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Figure 2: Diagram of Fabry-Perot Resonator







### Frequency, GHz

Figure 5: Measured microwave absorption from gaseous ammonia (3.7 ± 0.8 x 10<sup>-3</sup>, by volume) in a 90% hydrogen/10% helium atmosphere as a function of frequency (wavelength), at 193 ± 8 K, at three different pressures. Also shown (solid lines), are theoretically derived absorption profiles.

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Absorption of NH<sub>3</sub> -43in a 90% H<sub>2</sub> 10% He atmosphere (Mixing Ratio: 0.0184 Pressure: 2 atm.)



Figure 6: Ammonia absorption under Jovian conditions (T=203 K).





igure 8: Comparison of theoretically-derived absorption spectrum results (Janssen, personal communication, 1987) and laboratory measurements (Steffes, 1986).

### X. APPENDICES

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LADUTAL	ory mea	surements	s of	the	Opacity	<u>of</u>	Gaseous
Ammonia	$(NH_3)$	in the	7.3-8.	3 mm	(36-41	GHz	) Range
Under S	imulated	<u>Condition</u>	ons for	· Jov	ian Atmo	spher	es

J. Joiner, J. M. Jenkins, P. G. Steffes (Georgia Institute of Technology)

Previous experimental results have verified that the modified Ben-Reuven lineshape correctly describes the opacity of gaseous ammonia (NH3) in an H2/He atmosphere at wavelengths form 1.38 to 18.5 cm (1.62-21.7 GHz). To determine whether the Ben-Reuven lineshape correctly describes the absorption spectrum of NH2 at wavelengths shorward of 1 cm, laboratory measurements of the opacity of NH3 in a 90% H2/10% He atmosphere at pressures up to 2 atm and at wavelengths from 7.3-8.3 mm (36-41 GHz) have been made at room temperature. Further measurements in the same frequency and pressure ranges are presently being made at temperatures as low as 193K. These measurements needed are for inferrina the abundance and distribution of NH3 in the upper atmospheres of the outer planets from radio astronomical observations at these wavelengths.

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Limits of the Microwave Absorption of H<sub>2</sub>O and CH<sub>4</sub> in the Jovian Atmosphere

J. M. Jenkins, P. G. Steffes (Georgia Institute of Technology

The modified Ben-Reuven lineshape used to model the microwave absorption of gaseous ammonia (NH<sub>3</sub>) in a Jovian atmosphere suggests that the absorption due to a solar abundance of NH3(150 ppm) is not large enough to account for the actual absorption from 10 to 20 cm as inferred by the recent spectral emission studies and radio occultation experiments. To determine whether there is an additional microwave absorbing constituent present in Jupiter's atmosphere. we have measured the microwave absorption of methane (CH<sub>4</sub>) and water vapor (H<sub>2</sub>O) in a simulated Jovian atmosphere at 2.25 GHz (13.3 cm), 8.5 GHz (3.5 cm) and 21.7 GHz (1.38 cm). The measurements of the opacity of CH<sub>A</sub> were conducted at 153 K under pressures as high as 6 atmospheres in various proportions of hydrogen (H<sub>2</sub>) and helium (He). The measurements of the opacity of H<sub>2</sub>O were made at 298 K under pressures as high as 6 atmospheres in a 90% H<sub>2</sub>/10% He atmosphere with a mixing ratio of  $3.5 \times 10^{-3}$ . The results of these measurements are consistent with the theoretical expressions for microwave opacity in the Jovian atmosphere at the specified frequencies, and suggest that CHA and H<sub>2</sub>O are not responsible for the additional absorption observed between 10 and 20 cm. Thus ammonia abundances significantly greater than solar abundance are indicated.

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#### REPORT

#### TO THE

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

#### SEMIANNUAL STATUS REPORT #9

for

GRANT NAGW-533

## LABORATORY EVALUATION AND APPLICATION OF MICROWAVE ABSORPTION PROPERTIES UNDER SIMULATED CONDITIONS FOR PLANETARY ATMOSPHERES

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February 1, 1988 through July 31, 1988

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#### I. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often leads to significant misinterpretation of available opacity data. Steffes and Eshleman (1981) showed that under environmental conditions corresponding to the middle atmosphere of Venus, the microwave absorption due to atmospheric SO2 was greater than that calculated Similarly, results obtained for the from Van Vleck-Weisskopf theory. microwave opacity from gaseous H2SO4 under simulated Venus conditions, during the first two years of Grant NAGW-533, showed that not only was the opacity from  $H_2SO_4$  much greater than theoretically predicted, but that its frequency (wavelength) dependence was far different than that theoretically predicted (Steffes, 1985 and Steffes, 1986). Measurements made by Steffes and Jenkins (1987), during the third year of Grant NAGW-533, showed that the microwave opacity of gaseous ammonia (NH<sub>3</sub>) under simulated Jovian conditions did indeed agree with theoretical predictions to within experimental accuracy at wavelengths longward of 1.3 cm. Work performed during the fourth year of Grant NAGW-533 (February 1, 1987 through January 31, 1988) and continuing on into the first half of this current grant year (February 1, 1988 through July 31, 1988) has shown that the millimeter-wave opacity of ammonia between 7.5 mm and

9.3 mm likewise agrees with theoretical predictions. The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by both radio occultation experiments and radio astronomical observations, and over a range of frequencies which correspond to those used in both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has been the goal of this investigation to conduct such measurements and to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profiles of constituents in those planetary atmospheres.

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In some cases, new observations or experiments have been suggested by the results of the laboratory measurements. For example, this facility was initially developed, and then operated, in order to evaluate the microwave absorbing properties of gaseous sulfuric acid  $(H_2SO_4)$  under Venus atmospheric conditions. The results, obtained at 13.4 cm and 3.6 cm wavelengths, were applied to measurements from Mariner 5, Mariner 10, and early Pioneer-Venus Radio Occultation experiments, to determine abundances of gaseous sulfuric acid in the Venus atmosphere, with accuracies exceeding those achieved with <u>in-situ</u> instruments (Steffes, 1985). Later efforts concentrated on making laboratory measurements of the microwave absorption from gaseous  $H_2SO_4$  at wavelengths from 1.3 to 22 cm under simulated Venus conditions. We applied these results to radio astronomical observations of Venus which have been made in the same wavelength range, in order to better model the structure of  $H_2SO_4$ and  $SO_2$  abundance in the Venus atmosphere, and to resolve temporal variations of their abundances on a planet-wide basis.

Our laboratory measurements also suggested that a substantial variation in the Venus microwave emission, related to the abundance of gaseous sulfuric acid, might exist near the 2 cm wavelength. Since no observations of the Venus emission at this wavelength have ever been published, we conducted observations of Venus using the 140-foot NRAO telescope and the 64-meter DSN/Goldstone antenna in April 1987 to not only search for the presence of the predicted feature, but to use such a feature to determine a planet-wide average for sulfuric acid vapor abundance below the main cloud layer. The results of this observation are substantial in that they not only place limits on the abundance and spatial distribution of gaseous  $H_2SO_4$  and  $SO_2$ , but they also suggest some limits to long term temporal variations for the abundance of these two gases. During the first half of this current grant year, we have completed calibration and interpretive studies on the data from these observations and are submitting a paper entitled, "Observations of the Microwave Emission of Venus from 1.3 to 3.6 cm," by P. G. Steffes, M. J. Klein, and J. M. Jenkins, to the journal Icarus. (Preprints of this paper will accompany this report under separate cover.)

Another important tool for evaluating potential spatial and temporal variations in abundance and distribution of gaseous  $H_2SO_4$  is the reduction and analysis of recently obtained Pioneer-Venus radio occultation measurements. The 13 cm microwave absorptivity profiles, which can be obtained from the radio occultation data, are closely related to the abundance profiles for gaseous  $H_2SO_4$ . Within the last month, we have begun the reduction of the 1986-87 Pioneer-Venus radio occultation measurements (working at JPL with support from the Pioneer-Venus Guest Investigator Program) in order to obtain the needed 13 cm microwave absorptivity profiles. This reduction effort, and its potential results, are discussed in Section V of this report.

An equally important activity for the first half of this grant year has continued to be laboratory measurements of the microwave and millimeter-wave properties of the simulated atmospheres of the outer planets and their satellites. As described in the previous Annual Status Report for Grant NAGW-533 (February 1, 1987 through January 31, 1988), initial laboratory measurements of the millimeter-wave opacity of gaseous ammonia (NH<sub>3</sub>) in a hydrogen/helium (H<sub>2</sub>/He) atmosphere, under simulated conditions for the outer planets were begun in 1987. These measurements were conducted at frequencies from 32 to 40 GHz (wavelengths from 7.5 to 9.3 mm). It has been found by some (e.g., de Pater and Massie, 1985) that the observed millimeter-wave emission from Jupiter is inconsistent with the millimeter-wave absorption spectrum predicted using the modified Ben-Reuven line shape for ammonia. In order to investigate this, we developed a Fabry-Perot spectrometer system capable of operation from 30 to 41 GHz (wavelengths from 7.3 to 10 mm). This system has been used at pressures up to 2 Bars and temperatures as low as 150 K, which corresponds closely to the conditions at altitudes in the Jovian atmosphere most responsible for the observed millimeter-wave absorption. A complete description of the millimeter-wave spectrometer is given in Section II.

Initially, we used this spectrometer to complete laboratory measurements of the 7.5 to 9.3 mm absorption spectrum of ammonia. The results of these measurements were substantive in that they suggested the possibility that neither the modified Ben-Reuven lineshape nor the Van Vleck-Weisskopf lineshape best describe the 7.5 to 9.3 mm (32 to 40 GHz) absorption from gaseous  $NH_3$  under simulated Jovian conditions. However, because of the large error bars for these initial measurements, it was not possible to determine the specific absorption spectrum. In order to resolve this uncertainty, we

have found that it is desirable to characterize the opacity of ammonia to an accuracy of  $\pm 20\%$ . This would also help to determine upper level ammonia abundance and distribution in the atmospheres of the outer planets (from radio emission measurements), and to properly account for contributions to opacity from other absorbing constituents. For our initial measurements, accuracies of no better than  $\pm 60\%$  were achieved (see Joiner <u>et al.</u>, 1987). Thus, we have devoted a great deal of effort during the first half of this current grant year to improve the sensitivity of our 7.5 to 10 mm spectrometer system, as described in Section II. The effect of this improvement can be seen in the laboratory results described in Section IV.

Since larger variations from theoretically-derived opacity values are expected at shorter millimeter-wavelengths (see de Pater and Massie, 1985), we hope to pursue (in the second half of this grant year) further laboratory measurements at shorter millimeter-wavelengths, especially near 3.2 mm(94 GHz), where a large number of observations of the emission from the outer planets have been made. A better knowledge of the millimeter-wave absorption properties of NH<sub>3</sub> is essential, not only to help better characterize the distribution and abundance of ammonia at high levels in Jovian atmospheres, but to make it possible to resolve the contributions from other absorbing constituents such as  $H_2S$  (see Bezard et al., 1983).

Of equal importance, however, will be the further application of our laboratory results for the microwave and millimeter-wave absorption from various gases under simulated planetary conditions to available absorptivity data. These data sets include microwave and millimeter-wave emission measurements from Venus and the outer planets, from which opacity can be inferred. Other sources of absorptivity data include the Pioneer-Venus radio

occultation experiments, discussed in Section V, and Voyager radio occultation experiments at the outer planets. Results of our laboratory measurements have also been used in the study and planning of future missions (see Section VI).

#### II. THE GEORGIA TECH RADIO ASTRONOMY AND PROPAGATION (R.A.P.) FACILITY

The basic configuration of the planetary atmospheres simulator developed at Georgia Tech for use in measurement of the microwave absorptivity of gases under simulated conditions for planetary atmospheres is described at length in the previous Annual Status Report(s) for Grant NAGW-533. It is also discussed at length in Steffes (1986) and Steffes and Jenkins (1987). The most recent addition to the Georgia Tech Radio Astronomy and Propagation Facility has been a Fabry-Perot type resonator capable of operation between 30 and 41 GHz. As shown in Figure 1, the resonator consists of two gold plated mirrors (one with a flat surface, and one with a parabolic surface) separated by a distance of about 20 cm. The mirrors are contained in a T-shaped glass pipe which serves as a pressure vessel capable of withstanding over 2 atm of pressure. Each of the three open ends of the pipe is sealed with an O-ring sandwiched between the lip of the glass and a flat brass plate which is bolted to an inner flange. Electromagnetic energy is coupled both to and from the resonator by twin irises located on the surface of the flat mirror. Two sections of WR-28 waveguide which are attached to the irises pass through the brass plate to the exterior of the pressure vessel. The end of each waveguide section is pressure-sealed by a rectangular piece of mica which is held in place by a mixture of rosin and beeswax. As shown in Figure 2, one of these ends is connected to the sweep oscillator through a waveguide section. A Ka-band (26-40 GHz) mixer is attached to the other section of waveguide and is coupled to

the high resolution spectrum analyzer through a calibrated section of coaxial cable. The entire resonator, including its glass pressure envelope, is placed in the temperature chamber, which is a low-temperature freezer capable of operation down to 150 K. A network of stainless steel tubing and valves connects other components such as gas storage tanks, vacuum gauges, the pressure gauge, and the vacuum pump to the resonator assembly, so that each component may be isolated from the system as necessary. When properly secured, the system is capable of containing up to two atmospheres of pressure without detectable leakage. The sensitivities (minimum detectable opacities) previously achievable with this system are shown in Figure 3 (solid line). However, as previously discussed, these minimum sensitivities were too large to allow an unambiguous determination of the absorption spectrum of ammonia under simulated Jovian conditions.

In order to achieve a better system sensitivity, which corresponds to a higher "Q" or quality factor for the Fabry-Perot resonator (see Section III), all losses in the resonator must be minimized, since the quality factor is defined as  $2\pi$  times the ratio of the average energy stored in the resonator to the energy lost (per cycle) in the resonator. There are three sources of loss which typically affect a Fabry-Perot resonator (Collin, 1966):

- (1) Resistive losses on the surfaces of the mirrors.
- (2) Coupling losses due to the energy coupling out of the resonator through the irises on the flat-surfaced mirror.
- (3) Diffraction losses around the sides of the mirrors.

For previous measurements made at frequencies below 22 GHz (wavelengths longer than 1.35 cm), it was found that the resistive losses were predominant. This was because the measurements were conducted using cylindrical resonators,
for which no diffraction losses existed. (See Steffes and Jenkins, 1987.) Also, coupling losses were held to a minimum by using very small coupling loops and irises. The predominance of the resistive losses in the cylindrical resonators was demonstrated when such resonators were cooled to 193 K in the atmospheric simulator. Significant improvement in the quality factor of the resonators were observed when compared with their room temperature values. This was consistent with the expected reduction in the resistive losses at lower temperatures.

When the newer, higher frequency (30 to 41 GHz) Fabry-Perot resonator (Figure 1) was first cooled from room temperature down to 203 K for tests under simulated Jovian conditions, its quality factor appeared to worsen rather than improve. Initially, it was thought that this might have been caused by separation of the gold plating on the mirror surfaces from the backstructure (which had been machined from aluminum) due to differential thermal contraction. As a result, new mirrors were machined (to high tolerance) from brass, and then were plated with titanium and then gold, to assure no separation would occur. The performance of the new mirrors was only marginally better when installed in the resonator. Computation of the resistive losses from the mirrors showed that, in the absence of all other losses, the Q of our Fabry-Perot resonator should be on the order of 250,000; whereas its actual Q was on the order of 10,000. Therefore, it became clear that either coupling losses or diffraction losses were the limiting factor in its performance, and that even the introduction of high-temperature superconducting material would not significantly improve the sensitivity of the system. (Note, however, that we are still studying the possibility of using high temperature superconductors in our lower frequency, cylindrical resonators in order to obtain increased sensitivity at frequencies below 22 GHz.)

In order to further improve the guality factor of the 30 to 41 GHz system, some additional improvements were made. First, adjustable irises were developed so that the smallest possible coupling losses would occur, while still allowing sufficient signal coupling in and out of the resonator so as to make accurate absorptivity measurements. Since the irises are actually circular holes which are placed near the center of the flat-surfaced mirror, adjustment of their sizes is difficult. However, two small metal sheets with V-shaped cuts were placed immediately behind each iris in an area where the mirror surface thickness is very small. The two sheets could be moved together or apart so as to adjust the effective size, and therefore the coupling, of each iris. Even when adjusted for minimal coupling, however, the resonator Q was only slightly improved, suggesting that diffraction losses were the major limiting factor to system sensitivity.

Diffraction losses occur due to energy being lost around the edges of the mirrors. These losses can be minimized by assuring that both mirrors are oriented directly toward each other (i.e., the centerlines for each mirror, which are orthogonal to the planes of each mirror at their centerpoints, must be colinear). Since the positioning of the mirrors can vary with temperature, due to thermal contraction or expansion of metallic mounting structures, the temperature dependence of the quality factor which has been observed is consistent with a system which is limited by diffraction losses.

Two approaches can be used to minimize diffraction losses. The first involves precise pointing of the mirrors. This was accomplished by directing the beam of a helium-neon laser through the input waveguide and iris and into the resonator. Mirror positioner screws could then be adjusted so that the reflected beam focused precisely on the output iris. Since the parabolic

mirror has a precisely defined focus, adjustment of its exact position is far more critical than that of the flat mirror. The second technique for reducing diffraction loss involves the use of larger mirrors in the resonator. However, because of size limitations set by our pressure vessel, we are unable to significantly increase the size of the mirrors in our system.

Overall, our efforts during the first half of this grant year at improving the quality factor of our 30 to 41 GHz Fabry-Perot resonator have been successful, but in themselves have not been enough to provide the needed increase in system sensitivity. However, since absorptivity is measured by monitoring the <u>change</u> in the quality factor of the resonator which is caused by the absorbing gas mixture, improvements in our measurement technique, described in Section III, have allowed us to achieve the required system sensitivity.

#### **III. EXPERIMENTAL APPROACH**

The approach used to measure the microwave absorptivity of test gases in an  $H_2/He$  atmosphere is similar to that used previously by Steffes and Jenkins (1987) for simulated Jovian atmospheres. At frequencies between 30 and 41 GHz, the changes in the Q of the numerous resonances of the Fabry-Perot resonator (see Figure 2) are related to the absorptivity of the test gas mixture at these frequencies. The changes in the Q of the resonances which are induced by the introduction of an absorbing gas mixture can be monitored by the high resolution microwave spectrum analyzer, since Q is simply the ratio of the cavity resonant frequency to its half-power bandwidth. For relatively low-loss gas mixtures, the relation between the absorptivity of the gas mixture and its effect on the Q of a resonance is straightforward:

$$\alpha \simeq (Q_{\rm L}^{-1} - Q_{\rm C}^{-1}) \pi / \lambda \tag{1}$$

where  $\alpha$  is absorptivity of the gas mixture in Nepers km<sup>-1</sup>. (Note, for example, that an attenuation constant or absorption coefficient or absorptivity of 1 Neper km<sup>-1</sup> = 2 optical depths per km (or km<sup>-1</sup>) = 8.686 dB km<sup>-1</sup>, where the first notation is the natural form used in electrical engineering, the second is the usual form in physics and astronomy, and the third is the common (logarithmic) form. The third form is often used in order to avoid a possible factor-of-two ambiguity in meaning.) Q<sub>L</sub> is the quality factor of the cavity resonator when the gas mixture is present, Q<sub>C</sub> is the quality factor of the resonance in a vacuum, and  $\lambda$  is the wavelength (in km) of the test signal in the gas mixture.

In the first half of this grant year, we have attempted to make high accuracy measurements of the millimeter-wave absorption from gaseous ammonia  $(NH_3)$  in a 90%  $H_2/10\%$  He atmosphere at a temperature of 203 K. While even lower temperatures could be achieved, the need to avoid the risk of ammonia condensation kept our operating temperatures relatively high. As in the previous experiments, the bandwidth and center frequencies of each of several resonances between 32 and 41 GHz were measured in a vacuum. Next, 28 torr of gaseous ammonia is added to the system. The pressure of the ammonia gas is measured with the high-accuracy thermocouple vacuum gauge, as shown in Figure 2.

In addition, the ammonia abundance can be monitored by measuring refractivity of the introduced gas. Since the index of refraction (relative to unity) is proportional to the ammonia gas abundance, the ability of the system

to accurately measure refractivity (through measurement of the frequency shift of resonances) can be used to infer the relative vapor abundance or pressure. Note that it is not yet possible to use this approach for the accurate determination of absolute  $NH_3$  pressure since accurate refractivity data for the 7.3 to 10 mm wavelength range is not available. (In fact, by using our thermocouple vacuum gauge, we have made measurements of the density-normalized refractivity of gaseous ammonia at 39 GHz, and found it to be  $8.8 \times 10^{-17}$ N-units/molecule/cm, which is nearly 8 times the value at optical wavelengths.)

Next, 1.8 atm of hydrogen  $(H_2)$  and 0.2 atm of helium (He) are added to the chamber, bringing the total pressure to 2 atm. The bandwidth of each resonance is then measured and compared with its value when the chamber was evacuated in order to determine the absorptivity of the gas mixture at 2 atm total pressure. The total pressure is then reduced, by venting, to 1 atm, and the bandwidths are again measured. Finally, the pressure vessel is again evacuated and the bandwidths again measured so as to assure no variation of the Q's of the evacuated resonator has occurred. As with the previous measurements, the measured changes of bandwidths (Q's) can then be used to compute the absorptivity of the gas mixtures at each of the resonant frequencies.

This approach has the advantage that the same gas mixture is used for the absorptivity measurements at the various pressures. Thus, even though some small uncertainty may exist as to the mixing ratio of the initial mixture, the mixing ratios at all pressures are the same, and thus the uncertainty for any derived pressure <u>dependence</u> is due only to the accuracy limits of the absorptivity measurements, and not to uncertainty in the mixing ratio. (This

assumes that the mixing ratio is small, so that foreign-gas broadening predominates, as is the case for our measurements.) Similarly, measurements of the frequency <u>dependence</u> of the absorptivity from the mixture would likewise be immune to any mixing ratio uncertainty, since foreign-gas broadening predominates.

For the measurements described, the amount of absorption being measured is extremely small. Thus, any errors in measurements of (or other changes in) the apparent bandwidth of the resonances, not caused by the absorbing gases, could lead to significant errors in the absorption measurement. The contribution of instrumental errors and noise-induced errors on such absorptivity measurements have been discussed at length in Steffes and Jenkins (1987). However, because our latest measurements represent such small percentage changes in bandwidth, another instrumental source of error which we refer to as dielectric loading becomes a concern.

As can be seen in Figure 2, the resonator, which operates as a bandpass filter, is connected to a signal source (the millimeter-wave sweep oscillator) and to a signal receiver (the high-resolution spectrum analyzer). The "Q" of the resonator, which is defined as the ratio of energy stored in the resonator to the energy lost per cycle, equals the ratio of resonant center frequency to resonance half-power bandwidth. It is not surprising, therefore, that the stronger the coupling between the resonator and the spectrum analyzer or sweep oscillator, the lower will be the Q of the resonance, since more energy will be lost per cycle through the waveguides connecting the resonator to the spectrum analyzer and sweep oscillator. For this reason, we have always designed our resonators (both the coaxially-coupled cylindrical cavity resonators used below 25 GHz and the waveguide-coupled Fabry-Perot resonator

used above 30 GHz) with minimal coupling, so as to maximize Q and to minimize the changes in Q that might result from changes in coupling that occur when gases are introduced into the resonators. It should be noted that these changes in coupling, which are due to the presence of the test gas mixtures, are not related to the absorptivity of the gases, but rather to the dielectric constant or permittivity of the test gas mixtures. (Hence, the term "dielectric loading.")

We have always strived to design the coupling elements of the resonators so that the changes in lossless test gas abundances (and resulting changes in dielectric constant) had little or no effect on the Q of the resonator as measured in the system. This has been no small feat in that slight imperfections in resonators, cables, coupling loops, or waveguides can make the apparent Q of the resonator appear to vary with the abundance of such lossless gases. It has now become a standard part of our experimental procedure to repeat absorption measurements for gas mixtures in which the absorbing gas is a minor constituent, without the absorbing gas present. For example, after measurements were made of the microwave and millimeter-wave absorption from ammonia as a minor constituent in an  $H_2/He$  atmosphere, measurements of the apparent absorption of the H2/He atmosphere without the ammonia gas were made. Since, for the pressures and wavelengths involved, the  $H_2/He$  atmosphere is essentially transparent, no absorption was expected. If any apparent absorption was detected, "dielectric loading," or a change in coupling due to the dielectric properties of the gases, was indicated.

Initially, if any evidence of dielectric loading existed, the experiments were terminated and the apparatus disassembled, including pressure seals. The cables and coupling loops were then readjusted, and the system reassembled and

tested again. The entire procedure was repeated until the dielectric loading effect was eliminated or minimized. If some small variation in the resonant Q or bandwidth due to the presence of the non-absorbing gases still remained, it was added to the uncertainty or error bars for each experiment. More recently however, we have found that the effects of dielectric loading are additive, in that they add to the apparent changes of resonator bandwidth caused by the absorbing gases. Thus, as long as the effects of dielectric loading are not time variable, they can be removed by using the measured value of the Q of a resonance with the non-absorbing gases present (instead of the Q of the resonance in a vacuum) for the quantity Q<sub>c</sub> in equation (1).

Another potential source of instrumental error which we have recently detected has to do with nonlinearities in the spectrum analyzer display. We have found that depending on the vertical position of the bandpass spectrum on the spectrum analyzer CRT display, the peak signal level (and, therefore, the apparent half-power bandwidth and resulting quality factor) of the resonator seems to vary slightly. This is due to nonlinearities in the CRT vertical deflection amplifier. We have minimized this potential error by always resetting the vertical position of the displayed spectrum to the same portion of the CRT screen.

As previously discussed, once a test gas mixture is formed in the pressure vessel, the same mixture is used for measurement of absorption at several frequencies and pressures. Thus, even though some uncertainty in absolute mixing ratio exists, the pressure and frequency <u>dependences</u> of the millimeter-wave absorption can be measured to high accuracy. However, in order to properly characterize the magnitude of the absorption, the ammonia mixing ratio must be known precisely. Using the high accuracy thermocouple

vacuum gauge shown in Figure 2, the actual  $NH_3$  mixing ratio can only be determined to an accuracy of ±20% of its value. (Note: This corresponds to  $(1.85 \pm 0.37)$ %  $NH_3$  volume mixing ratio.) However, since our required overall accuracy for the  $NH_3$  absorptivity measurement is ±20%, this mixing ratio uncertainty is excessive. In order to reduce this uncertainty, we arranged for a local gas products supplier (Matheson Gas Products) to provide us with a pre-mixed hydrogen/helium/ammonia atmosphere which was analyzed with a mass spectrometer so that mixing ratio accuracies of better than 2% (i.e., (1.85 ±.04)%) were obtained. We have used this mixture (1.85%  $NH_3$ , 9.81% He, and 88.34%  $H_2$ ) for the high accuracy absorptivity measurements which are required to accurately infer ammonia abundance from millimeter-wave opacity data for the Jovian planets.

### IV. RESULTS OF LABORATORY MEASUREMENTS AND THEIR APPLICATION

Initial measurements of the 7.5 to 9.2 mm absorptivity from  $NH_3$  in a hydrogen/helium atmosphere were conducted at 203 K as described in Section III, with an ammonia mixing ratio of 0.0186, at pressures of 1 and 2 Bars. An examination of these early experimental results revealed that because of large error bars, we could not determine whether the modified Ben-Reuven lineshape best described the absorption profile of gaseous ammonia shortward of 1 cm, as discussed in Sections I and II. As a result, we have undertaken higher accuracy measurements in the first half of this current grant year.

Results of measurements of the 32 to 40 GHz (7.5 to 9.2 mm) absorptivity of gaseous ammonia under simulated Jovian conditions (203 K) are shown in Figure 4. These measurements were made using a 88.34% hydrogen/9.81% helium

atmosphere with a total pressure of 2 Bars. The ammonia mixing ratio was 0.0185. Triangular points represent measurements of gas mixtures formed using the thermocouple vacuum gauge (NH<sub>3</sub> mixing ratio accuracy =  $\pm 20\%$  of its value) and the circular points represent measurements of the pre-mixed, analyzed gas mixture described in Section III. With this mixing ratio, temperatures as low as 190 K could have been used before saturation would have become a problem, but 203 K was used so as to be consistent with earlier measurements.

Also, shown in Figure 4 are solid lines which represent the theoretically-computed opacity using the Van Vleck-Weisskopf lineshape (upper line), the modified Ben-Reuven lineshape (middle line), and the Zhevakin-Naumov lineshape (lower line). The Van Vleck-Weisskopf calculation was performed using linewidths and line intensities as per Wrixon et al. (1971). The Ben-Reuven calculation was made as per Berge and Gulkis (1976), by employing a Ben-Reuven lineshape which has been modified so as to be consistent with the laboratory results of Morris and Parsons (1970), in which the 9.58 GHz absorption from  $NH_3$  (in a high pressure  $H_2$ /He atmosphere) at room temperature was measured. The Zhevakin-Naumov calculation used the lineshape of Zhevakin and Naumov (1967) and linewidths and line intensities from Wrixon et al. (1971). These theoretical spectra were computed using generalized computer programs for which the partial pressures from H<sub>2</sub>, He, and NH<sub>3</sub>, as well as frequency and temperature, were adjustable variables. The values picked for these variables matched our experimental conditions.

Inspection of the results in Figure 4 shows that most of the measured data points lie nearest to the theoretically-derived absorptivity expression based on the Zhevakin-Naumov lineshape. This differs from our preliminary measurements which suggested that the ammonia opacity might actually exceed

that indicated by the modified Ben-Reuven lineshape. Such results are not surprising, however, in that the accuracy of the new measurements is far greater than that from previous measurements. It is also noteworthy that other researchers, such as de Pater and Massie (1985), have found that in order to best explain the 1-10 mm Jupiter emission spectrum, a different sort of lineshape was needed to characterize the ammonia opacity. Likewise, previous laboratory measurements of NH<sub>3</sub> opacity at frequencies below 22 GHz showed opacities less than those indicated by the modified Ben-Reuven lineshape under the same conditions of temperature and pressure (Steffes and Jenkins, 1987). Since even larger variations from either the modified Ben Reuven formulation or the Van Vleck-Weisskopf formulation for ammonia opacity are expected at shorter millimeter-wavelengths, we hope to pursue further laboratory measurements, especially near 3.2 mm (94 GHz), where a larger number of observations of the emission from the outer planets have been made.

A better knowledge of the millimeter-wave absorption properties of  $NH_3$  is essential, not only to help better characterize the distribution and abundance of ammonia at high levels in Jovian atmospheres, but to make it possible to resolve the contributions from other absorbing constituents such as  $H_2S$  (see Bezard <u>et al.</u>, 1983). Our goal to better characterize the millimeter-wave absorption spectrum of ammonia will not only involve increasing the range of frequencies over which measurements are made, but to increase the sensitivity of the measuring systems.

### V. OBSERVATIONAL AND INTERPRETIVE STUDIES

As described in the previous <u>Annual Status Report</u> for Grant NAGW-533 (February 1, 1987 through January 31, 1988), studies of our recent measure-

ments of the 1.35 to 3.6 cm emission from Venus have suggested that long term temporal and/or significant spatial variations in the abundances of  $SO_2$  and gaseous  $H_2SO_4$  may occur immediately below the main cloud layer (48 km and below). Our observation, which was predominantly of equatorial and midlatitude regions of Venus, indicated a significantly lower  $SO_2$  abundance than was measured in 1978 by the Pioneer-Venus Sounder Probe, and a lower average abundance of gaseous  $H_2SO_4$  than would have been inferred from earlier Pioneer-Venus radio occultation studies of subcloud opacity at 13 cm. Some or all of this difference may be due to spatial variations in the subcloud  $H_2SO_4$ abundance since most of the early Pioneer-Venus results were for polar latitudes (Cimino, 1982). Similarly, our results may be consistent with the earlier equatorial 13 cm radio occultation opacity measurements made with the Mariner 10 spacecraft (Lipa and Tyler, 1979), where the peak opacity would correspond to a very large abundance of gaseous  $H_2SO_4$ , but the average subcloud opacity was significantly lower.

One important tool for evaluating these effects is the reduction of the microwave data from the 1986-87 Pioneer-Venus radio occultation measurements. This data was taken over a wide range of latitudes and could be critical for determining whether temporal variations or spatial variations in gaseous  $H_2SO_4$  abundance could be occurring. Working with Dr. Arvydas J. Kliore (P-V Radioscience Leader), our group has obtained the currently unreduced data and (working at JPL) has begun reducing the data to obtain absorptivity profiles for the 1986-87 epoch.

Over the next year, we will reduce data obtained from the Fall 1986/ Winter 1987 Pioneer-Venus Radio Occultation Observations. Since the initial conversion from amplitude and doppler (frequency) data to refractivity and

absorptivity profiles can be most efficiently completed at JPL, we have made arrangements to send graduate students to JPL for this activity. Support for travel and student salaries for the reduction effort at JPL has been obtained from the Pioneer-Venus Guest Investigator Program.

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Since the overall effort is of limited scope, we have selected a small number of radio occultations which are spread over a range of latitudes from polar to equatorial. We will correct the "raw" radio occultation data (amplitude and doppler) for spacecraft motions, frequency drift, and for antenna pointing inaccuracies, in order to then invert the data to obtain reliable refractivity and absorptivity profiles. Since the Pioneer-Venus Orbiter can no longer be positioned to track the planetary limb during occultations and because of spacecraft "wobble," use of the 3.6 cm wavelength has been extremely limited. This is not a severe constraint, however, since under optimal conditions the 3.6 cm wavelength can probe no deeper than 50 km altitude (Cimino, 1982), and we are most interested in altitudes below 48 km.

After the initial reduction, we hope to have dependable 13 cm wavelength refraction and absorption profiles for a range of altitudes in the Venus atmosphere reaching down to 38 km and for latitudes ranging from equatorial to polar. Figure 5 shows a preliminary 13 cm absorptivity profile derived from radio data obtained during the entry occultation of Orbit 2801 on August 6, 1986. (Note: This is the first such absorptivity profile which has been derived since Orbit 358 - November 28, 1979.) This occultation probed the Venus atmosphere at 52°N latitude and the ray path traversed mainly the night side of the planet. For this orbit, the spacecraft signal was only receivable at the Goldstone DSS-14 receiver down to a periapsis altitude of 43.7 km before receiver lock was lost. (This assumes a planetary radius of 6052 km.)

It is hoped that other orbits may be found which probe deeper into the atmosphere before loss of signal, but because the Pioneer-Venus steerable antenna no longer tracks the limb of the planet (the direction the radio ray travels back to earth) during the occultation, the resulting lower signal level may prevent probing deeper than the 40 km altitude.

To show the usefulness of the 13 cm opacity data for inferring the nature of the gaseous  $H_2SO_4$  abundance, we compare in Figure 6 the measured absorptivity profile from orbit 2801N with that absorptivity which would result from a saturation abundance of gaseous  $H_2SO_4$  (from Steffes, 1985) in the 43 to 55 km altitude range. It can be seen that for altitudes in the 49 to 51 km altitude range (the nominal altitude range of the Venus lower cloud - see Ragent and Blamont, 1980), absorptivity values close to those caused by a saturation abundance of gaseous  $H_2SO_4$  are seen. At lower altitudes, most values for absorptivity are below those caused by a saturation abundance. It should be noted that error bars for this preliminary absorptivity data have not yet been computed, but are expected to be on the order of  $\pm 0.001$  dB/km  $(\pm 0.00023 \text{ km}^{-1})$ .

Figure 7 shows the full extent to which application of our laboratory results can be carried. The abundance of gaseous  $H_2SO_4$  (derived from the absorptivity profile in Figure 5 by using laboratory results from Steffes, 1985) is plotted as a function of altitude, along with a plot of the saturation abundance of gaseous  $H_2SO_4$ , for comparison. As our work in the Pioneer-Venus Guest Investigator Program yields more absorptivity profiles for a wide range of locations in the Venus atmosphere, we hope to be able to well characterize the abundance, structure, and spatial variations of gaseous  $H_2SO_4$  in the Venus atmosphere. We also hope to make comparative studies with earlier

radio occultation measurements in order to detect possible temporal variations in  $H_2SO_4$  abundance and structure.

# VI. PUBLICATIONS AND INTERACTION WITH OTHER INVESTIGATORS

In the first half of the current grant year, a paper was completed and accepted for publication in <u>Icarus</u>, describing results and applications of some experiments performed during the previous year of Grant NAGW-533 (Jenkins and Steffes, 1988). This paper describes laboratory measurements of the microwave absorption of methane  $(CH_4)$  and water vapor  $(H_2O)$  under simulated Jovian conditions. The paper also concludes, that based on these laboratory results, neither methane nor water vapor can be responsible for the excess microwave opacity detected at wavelengths between 10 and 20 cm in the atmosphere of Jupiter. This supports the presence of an ammonia abundance which exceeds solar abundance by a factor of 1.5 in the 2 to 6 Bar levels in Jupiter's atmosphere.

In addition, as discussed in Section I, we have just completed a paper describing observations and interpretive studies of the 1.3 to 3.6 cm Venus emission spectrum (Steffes <u>et al.</u>, 1988). Likewise, a paper describing the results and applications of the laboratory measurements of the millimeter-wave opacity of ammonia described in Section IV is currently in preparation. We also submitted updated summaries of our most recent laboratory measurements for inclusion in the twenty-first issue of the <u>Newsletter of Laboratory</u> Spectroscopy for Planetary Science.

In addition to the observations of Venus and analysis work conducted jointly with Dr. Michael J. Klein of JPL, we have also worked with Dr. Michael

A. Janssen of JPL regarding models for the Venus atmosphere, interpretation of microwave emission measurements, and theoretical models for the absorption spectrum of H<sub>2</sub>SO<sub>4</sub>. We have also worked with Dr. Arvydas J. Kliore of JPL on the reduction and interpretation of data from recent Pioneer-Venus Radio Occultation Studies as part of our involvement in the Pioneer-Venus Guest Investigator Program. More informal contacts have been maintained with groups at the California Institute of Technology (Dr. Duane O. Muhleman and his students, regarding interpretation of radio astronomical measurements of Venus opacity), at the Stanford Center for Radar Astronomy (V. R. Eshleman, G. L. Tyler, and T. Spilker, regarding Voyager results for the outer planets, and laboratory measurements), and at JPL (Drs. Robert Poynter and Samuel Gulkis, regarding radio astronomical observations of the outer planets and Venus). We have also worked with Dr. Imke de Pater (University of California-Berkeley) by using our laboratory measurements of atmospheric gases in the interpretation of radio astronomical observations of Venus and the outer planets. We have also studied possible effects of the microwave opacity of cloud layers in the outer planets' atmospheres. In this area, we have worked both with Dr. de Pater and with Dr. Paul Romani (NRC Associate, Goddard SFC). Dr. Steffes has also been active in the review of proposals submitted to the Planetary Atmospheres Program at NASA (both as a "by-mail" reviewer and as a panel member) and as a reviewer of manuscripts submitted to Icarus and the Journal of Geophysical Research, for which Dr. Steffes is an Associate Editor. We have also continued to serve the planetary community through the distribution of reprints of our articles describing our laboratory measurements and their application to microwave and millimeter-wave data from planetary atmospheres. The results of these measurements have been used in the

mission planning for radio and radar systems aboard the Galileo and Magellan missions, and more recently, for proposed experiments for the Cassini mission. Dr. Steffes also participated as a member of the International Jupiter Watch (IJW) Laboratory/ Theory Discipline Team. Another source of close interaction with other planetary atmospheres principal investigators has been Dr. Steffes' membership in the Planetary Atmospheres Management and Operations Working Group (PAMOWG). Travel support for attendance at PAMOWG meetings has been provided by Georgia Tech in support of Planetary Atmospheres Research. Also in support of Planetary Atmospheres Research, Georgia Tech provided \$2,000 for required repairs and maintenance to the ultra-cold freezer system used in the outer planets atmospheric simulator.

### VII. CONCLUSION

In the first half of this grant year, we have continued to conduct laboratory measurements of the millimeter-wave properties of atmospheric gases under simulated conditions for the outer planets. Significant improvements in our current system have made it possible to accurately characterize the opacity from gaseous NH<sub>3</sub> at longer millimeter-wavelengths (7 to 10 mm) under simulated Jovian conditions. In the second half of this grant year, we hope to extend such measurements to even shorter millimeter-wavelengths. We will likewise continue to pursue further analysis and application of our laboratory results to microwave and millimeter-wave absorption data for the outer planets, such as results from Voyager Radio Occultation experiments and earthbased radio astronomical observations. We also intend to pursue the analysis of available multi-spectral microwave opacity data from Venus, including data from our most recent radio astronomical observations in the 1.3 to 3.6 cm

wavelength range and newly obtained Pioneer-Venus Radio Occultation measurements at 13 cm, using our laboratory measurements as an interpretive tool.

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Figure 1: Diagram of Fabry-Perot Resonator



Figure 2: Block Diagram of Atmospheric Simulator (30-40GHz)

rigure 3: Performance (minimum detectable absorptivities) for the millimeter-wave planetary atmospheres simulator (operating at 200 K) at the beginning -31of the current grant year.



ABSORPTION of NH<sub>3</sub> in a -32-88.34% H<sub>2</sub>,9.81% He, 1.85% NH<sub>3</sub> ixture (Mixing ratio: 0.0185±0.0005) Pressure: 2 atm. Temp: 203K





-2 -4 -4

Figure 6: Comparison of absorptivities measured with radio occultation technique (circular points -- from Pioneer-Venus Orbit 2801entry occultation) with absorptivity which would result from saturation abundance of H<sub>2</sub>SO<sub>4</sub> (from Steffes, 1985). The absorption coefficient scale is logarithmic (exponents of 10). All measurements were made at the 13-cm wavelength (2.293 GHz).



Figure 7: Abundances of gaseous H<sub>2</sub>SO<sub>4</sub> inferred from Pioneer-Venus 13-cm absorptivity profiles (circular points) compared with the saturation abundance profile of gaseous H<sub>2</sub>SO<sub>4</sub> (from Steffes, 1985). The mixing ratio scale is logarithmic (exponents of 10).



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#### REPORT

#### TO THE

### NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

### ANNUAL STATUS REPORT (Includes Semiannual Status Report #10)

for

GRANT NAGW-533

#### LABORATORY EVALUATION AND APPLICATION OF

### MICROWAVE ABSORPTION PROPERTIES UNDER SIMULATED

#### CONDITIONS FOR PLANETARY ATMOSPHERES

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February 1, 1988 through January 31, 1989

Submitted by

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#### I. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often leads to significant misinterpretation of available opacity data. Results obtained for the microwave opacity from gaseous H<sub>2</sub>SO<sub>4</sub> under simulated Venus conditions, during the first two years of Grant NAGW-533, showed that not only was the opacity from  $H_2SO_4$  much greater than theoretically predicted, but that its frequency (wavelength) dependence was far different than that theoretically predicted (Steffes, 1985 and Steffes, 1986). Measurements made by Steffes and Jenkins (1987), during the third year of Grant NAGW-533, have shown that the microwave opacity of gaseous ammonia (NH<sub>3</sub>) under simulated Jovian conditions did indeed agree with theoretical predictions to within experimental accuracy at wavelengths longward of 1.3 cm. Work performed during the fourth year of Grant NAGW-533 (February 1, 1987 through January 31, 1988) and continuing on into this current grant year (February 1, 1988 through January 31, 1989) has shown that laboratory measurements of the millimeter-wave opacity of ammonia between 7.5 mm and 9.3 mm requires a different lineshape to be used in the theoretical prediction for millimeter-wave ammonia opacity than had been previously used (see Joiner et al., 1988). The recognition of the need to make such

laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by both radio occultation experiments and radio astronomical observations, and over a range of frequencies which correspond to those used in both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has been the goal of this investigation to conduct such measurements and to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profiles of constituents in those planetary atmospheres.

The key activity for this grant year has continued to be laboratory measurements of the microwave and millimeter-wave properties of the simulated atmospheres of the outer planets and their satellites. As described in the previous Annual Status Report for Grant NAGW-533 (February 1, 1987 through January 31, 1988), initial laboratory measurements of the millimeter-wave opacity of gaseous ammonia  $(NH_3)$  in a hydrogen/helium  $(H_2/He)$  atmosphere, under simulated conditions for the outer planets were begun in 1987. These measurements were conducted at frequencies from 32 to 40 GHz (wavelengths from 7.5 to 9.3 mm). It has been found by some (e.g., de Pater and Massie, 1985) that the observed millimeter-wave emission from Jupiter is inconsistent with the millimeter-wave absorption spectrum predicted using the modified Ben-Reuven lineshape for ammonia. In order to investigate this, we developed a Fabry-Perot spectrometer system capable of operation from 32 to 41 GHz. This system has been used at pressures up to 2 Bars and temperatures as low as 150 K, which corresponds closely to the conditions at altitudes in the Jovian atmosphere most responsible for the observed millimeter-wave absorption.

A complete description of the millimeter-wave spectrometer is given in Section II.

Initially, we used this spectrometer to complete laboratory measurements of the 7.5 to 9.3 mm absorption spectrum of ammonia. The results of these measurements were substantive in that they suggested the possibility that neither the modified Ben-Reuven lineshape nor the Van Vleck-Weisskopf lineshape best described the 7.5 to 9.3 mm (32 to 40 GHz) absorption from gaseous NH3 under simulated Jovian conditions. However, because of the large error bars for these initial measurements, it was not possible to determine the specific absorption spectrum. In order to resolve this uncertainty, we have found that it is desirable to characterize the opacity of ammonia to an accuracy of ±20%. For our initial measurements, accuracies of no better than ±60% were achieved (see Joiner et al., 1987). Thus, we devoted a great deal of effort during the first half of this current grant year to improve the sensitivity of our 7.5 to 9.3 mm spectrometer system, as described in The effect of this improvement can be seen in the laboratory Section II. results described in Section IV.

Since larger variations from theoretically-derived opacity values were expected at shorter millimeter-wavelengths (see de Pater and Massie, 1985), we began (in the second half of this grant year) laboratory measurements at wavelengths near 3.2 mm (94 GHz), where a large number of observations of the emission from the outer planets have been made. A description of this new system is presented in Section II. A better knowledge of the millimeter-wave absorption properties of  $NH_3$  is essential, not only to help better characterize the distribution and abundance of ammonia at high levels in Jovian atmospheres, but to make it possible to resolve the contributions from other absorbing constituents such as  $H_2S$  (see Bezard <u>et al.</u>, 1983).

In some cases, new observations or experiments have been suggested by the results of our laboratory measurements. For example, this facility was initially developed, and then operated, in order to evaluate the microwave absorbing properties of gaseous sulfuric acid  $(H_2SO_4)$  under Venus atmospheric conditions. The results, obtained at 13.4 cm and 3.6 cm wavelengths, were applied to measurements from Mariner 5, Mariner 10, and early Pioneer-Venus Radio Occultation experiments, to determine abundances of gaseous sulfuric acid in the Venus atmosphere, with accuracies exceeding those achieved with in-situ instruments (Steffes, 1985). Further laboratory measurements also suggested that a substantial variation in the Venus microwave emission, related to the abundance of gaseous sulfuric acid, might exist near the 2.2 cm Since no observations of the Venus emission at this wavelength wavelength. had ever been published, we conducted observations of Venus using the 140-foot NRAO telescope and the 64-meter DSN/Goldstone antenna in April 1987 to not only search for the presence of the predicted feature, but to use such a feature to determine a planet-wide average for sulfuric acid vapor abundance below the main cloud layer. The results of this observation were substantial in that they not only placed limits on the abundance and spatial distribution of gaseous  $H_2SO_L$  and  $SO_2$ , but they also suggested some limits to long term temporal variations for the abundance of these two gases. During the first half of this current grant year, we have completed calibration and interpretive studies on the data from these observations and have submitted a paper entitled, "Observations of the Microwave Emission of Venus from 1.3 to 3.6 cm," by P. G. Steffes, M. J. Klein, and J. M. Jenkins, to the journal Icarus.

Another important tool for evaluating potential spatial and temporal variations in abundance and distribution of gaseous  $H_2SO_4$  is the reduction and analysis of recently obtained Pioneer-Venus radio occultation measurements. The 13 cm microwave absorptivity profiles, which can be obtained from the radio occultation data, are closely related to the abundance profiles for gaseous  $H_2SO_4$ . Starting in June, we began the reduction of the 1986-87 Pioneer-Venus radio occultation measurements (working at JPL with support from the Pioneer-Venus Guest Investigator Program) in order to obtain the needed 13 cm microwave absorptivity profiles. This reduction effort, and its potential results, are discussed in Section V of this report.

Over the next nine months of Grant NAGW-533, we intend to complete our laboratory analysis of the millimeter-wave absorption from gaseous NH3 under simulated Jovian conditions, by completing our laboratory studies at 3.2 mm (94 GHz). We will then develop a formulation which accurately predicts the opacity from gaseous ammonia in a Jovian-type atmosphere over the entire 3 mm to 20 cm wavelength range (frequencies from 1.5 to 100 GHz). With such a formulation at our disposal, we will then develop models for microwave and millimeter-wave emission from the Jovian planets and adjust aomononia abundance profiles so as the match the emission spectrum observed from earth-based radio telescopes. We may even be able to take advantage of the availability of several millimeter-wave radio telescope arrays in order to make observations from which we could develop localized ammonia abundance profiles over the entire disk of one or more Jovian planets. Likewise, we will continue to take advantage of the availability of profiles of the 13 cm absorptivity in the Venus atmosphere, which we are developing as part of the Pioneer-Venus Guest Investigator Program. These profiles, which are related to the distribution

of gaseous H<sub>2</sub>SO<sub>4</sub> will be invaluable for characterizing the spatial and temporal variabilities of H<sub>2</sub>SO<sub>4</sub> in the Venus atmosphere. Finally, we will complete designs for laboratory instrumentation which will allow us to measure the microwave and millimeter-wave properties of liquids and solids under simulated planetary conditions. A more complete discussion of this proposed future activity is included in the accompanying proposal to NASA entitled, "Laboratory Evaluation and Application of Microwave Absorption Properties under Simulated Conditions for Planetary Atmospheres."

### II. THE GEORGIA TECH RADIO ASTRONOMY AND PROPAGATION (R.A.P.) FACILITY

The basic configuration of the planetary atmospheres simulator developed at Georgia Tech for use in measurement of the microwave absorptivity of gases under simulated conditions for planetary atmospheres is described at length in the previous Annual Status Report(s) for Grant NAGW-533. It is also discussed at length in Steffes (1986) and Steffes and Jenkins (1987). The most recent addition to the Georgia Tech Radio Astronomy and Propagation Facility have been Fabry-Perot type resonators capable of operation between 30 and 41 GHz and between 93 and 95 GHz. As shown in Figure 1, the Ka-band resonator (32-40 GHz) consists of two gold plated mirrors (one with a flat surface, and one with a parabolic surface) separated by a distance of about 20 cm. The mirrors are contained in a T-shaped glass pipe which serves as a pressure vessel capable of withstanding over 2 atm of pressure. Each of the three open ends of the pipe is sealed with an O-ring sandwiched between the lip of the glass and a flat brass plate which is bolted to an inner flange. Electromagnetic energy is coupled both to and from the resonator (which operates as a bandpass filter) by twin irises located on the surface of the
flat mirror. Two sections of WR-28 waveguide which are attached to the irises pass through the brass plate to the exterior of the pressure vessel. The end of each waveguide section is pressure-sealed by a rectangular piece of mica which is held in place by a mixture of rosin and beeswax. As shown in Figure 2, one of these ends is connected to the sweep oscillator through a A Ka-band (26-40 GHz) mixer is attached to the other waveguide section. section of waveguide and is coupled to the high resolution spectrum analyzer through a calibrated section of coaxial cable. The entire resonator, including its glass pressure envelope, is placed in the temperature chamber, which is a low-temperature freezer capable of operation down to 150 K. network of stainless steel tubing and valves connects other components such as gas storage tanks, vacuum gauges, the pressure gauge, and the vacuum pump to the resonator assembly, so that each component may be isolated from the system as necessary. When properly secured, the system is capable of containing up to two atmospheres of pressure without detectable leakage.

In order to achieve a better system sensitivity, which corresponds to a higher "Q" or quality factor for the Fabry-Perot resonator (see Section III), all losses in the resonator must be minimized, since the quality factor is defined as  $2\pi$  times the ratio of the average energy stored in the resonator to the energy lost (per cycle) in the resonator. There are three sources of loss which typically affect a Fabry-Perot resonator (Collin, 1966):

- (1) Resistive losses on the surfaces of the mirrors.
- (2) Coupling losses due to the energy coupling out of the resonator through the irises on the flat-surfaced mirror.
- (3) Diffraction losses around the sides of the mirror.

For previous measurements made at frequencies below 22 GHz (wavelengths longer than 1.35 cm), it was found that the resistive losses were predominant. This was because the measurements were conducted using <u>cylindrical</u> resonators, for which no diffraction losses existed. (See Steffes and Jenkins, 1987). Also, coupling losses were held to a minimum by using very small coupling loops and irises. The predominance of the resistive losses in the cylindrical resonators was demonstrated when such resonators were cooled to 193 K in the atmospheric simulator. Significant improvement in the quality factor of the resonators were observed when compared with their room temperature values. This was consistent with the expected reduction in the resistive losses at lower temperatures.

When the newer, higher frequency (32 to 40 GHz) Fabry-Perot resonator (Figure 1) was first cooled from room temperature down to 203 K for tests under simulated Jovian conditions, its quality factor appeared to worsen rather than improve. Initially, it was thought that this might have been caused by separation of the gold plating on the mirror surfaces from the backstructure (which had been machined from aluminum) due to differential thermal contraction. As a result, new mirrors were machined (to high tolerance) from brass, and then were plated with titanium and then gold, to assure no separation would occur. The performance of the new mirrors was only marginally better when installed in the resonator. Computation of the resistive losses from the mirrors showed that, in the absence of all other losses, the Q of our Fabry-Perot resonator should be on the order of 250,000; whereas its actual Q was on the order of 10,000. Therefore, it became clear that either coupling losses or diffraction losses were the limiting factor in its performance, and that even the introduction of high-temperature superconducting material would

not significantly improve the sensitivity of the system. (Note, however, that we are still studying the possibility of using high temperature superconductors in our lower frequency, <u>cylindrical</u> resonators in order to obtain increased sensitivity at frequencies below 22 GHz.)

In order to further improve the quality factor of the 32 to 40 GHz system, some additional improvements were made. First, adjustable irises were developed so that the smallest possible coupling losses would occur, while still allowing sufficient signal coupling in and out of the resonator so as to make accurate absorptivity measurements. Since the irises are actually circular holes which are placed near the center of the flat-surfaced mirror, adjustment of their sizes is difficult. However, two small metal sheets with V-shaped cuts were placed immediately behind each iris in an area where the mirror surface thickness is very small. The two sheets could be moved together or apart so as to adjust the effective size, and therefore the coupling, of each iris. Even when adjusted for minimal coupling, however, the resonator Q was only slightly improved, suggesting that diffraction losses were the major limiting factor to system sensitivity.

Diffraction losses occur due to energy being lost around the edges of the mirrors. These losses can be minimized by assuring that both mirrors are oriented directly toward each other (i.e., the centerlines for each mirror, which are orthogonal to the planes of each mirror at their centerpoints, must be colinear). Since the positioning of the mirrors can vary with temperature, due to thermal contraction or expansion of metallic mounting structures, the temperature dependence of the quality factor which has been observed is consistent with a system which is limited by diffraction losses.

Two approaches can be used to minimize diffraction losses. The first involves precise pointing of the mirrors. This was accomplished by directing the beam of a helium-neon laser through the input waveguide and iris and into the resonator. Mirror positioner screws could then be adjusted so that the reflected beam focused precisely on the output iris. Since the parabolic mirror has a precisely defined focus, adjustment of its exact position is far more critical than that of the flat mirror. The second technique for reducing diffraction loss involves the use of larger mirrors in the resonator. However, because of size limitations set by our pressure vessel, we are unable to significantly increase the size of the mirrors in our system.

Overall, our efforts during the first half of this grant year at improving the quality factor of our 32 to 40 Ghz Fabry-Perot resonator were successful, but in themselves were not enough to provide the needed increase in system sensitivity. However, since absorptivity is measured by monitoring the <u>change</u> in the quality factor of the resonator which is caused by the absorbing gas mixture, improvements in our measurement technique, described in Section III, have allowed us to achieve the required system sensitivity.

In the second half of this grant year, we implemented a similar system for operation around 94 GHz (3.2 mm). As shown in Figure 3, the 94 GHz resonator resembles the Ka-band Fabry-Perot resonator in that it consists of two gold plate mirrors, one with a flat surface and one with a parabolic surface. However, the 94 GHz resonator is significantly different in that the flat mirror has a much smaller radius than the parabolic mirror, and the focal length of the parabolic mirror is much shorter, resulting in "tighter" focusing. Mechanically, the 94 GHz resonator is superior in that the parabolic mirror rests on two support arms and can be adjusted in distance from

the flat mirror without disturbing the other angular adjustments. The result has been a resonator with a quality factor (Q) of about 30,000.

The 94 GHz system (Figure 4) functions in the same fashion as does the Ka-band system in that changes in the Q of the Fabry-Perot resonator are monitored as lossy gas mixtures are introduced into the pressure vessel. However, because of the extremely high frequency of operation, costly millimeter-wave components (e.g., signal sources, waveguides, and mixers) are required. Fortunately, such a resource of millimeter-wave equipment exists at Georgia Tech within the Georgia Tech Research Institute (GTRI) and has been made available to us for these experiments. Figure 4, shows a W-band (75-110 GHz) sweep oscillator whose signal is fed into the Fabry-Perot resonator via an isolator, so as to provide a constant impedance both for the backward wave oscillator (BWO) tube in the sweeper and for the resonator. The resonator output at 94 GHz is then down-converted to around 1.55 GHz using a harmonic mixer which employs the tenth harmonic of a stabilized 9.24 GHz oscillator. The 1.5 GHz signal is then fed to the high-resolution spectrum analyzer by which the bandwidth (and, therefore, the Q) of the resonance can be measured. The pressurization systems for both systems are essentially identical.

#### III. EXPERIMENTAL APPROACH

The approach used to measure the microwave absorptivity of test gases in an  $H_2/He$  atmosphere is similar to that used previously by Steffes and Jenkins (1987) for simulated Jovian atmospheres. At frequencies between 32 and 40 GHz, the changes in the Q of the numerous resonances of the Fabry-Perot resonator (see Figure 2) are related to the absorptivity of the test gas mixture at these frequencies. Similarly, the changes in the Q of the single

94 GHz resonance are related to the absorptivity at that frequency. The changes in the Q of the resonances which are induced by the introduction of an absorbing gas mixture can be monitored by the high resolution microwave spectrum analyzer, since Q is simply the ratio of the cavity resonant frequency to its half-power bandwidth. For relatively low-loss gas mixtures, the relation between the absorptivity of the gas mixture and its effect on the Q of a resonance is straightforward:

$$\alpha \simeq \left(Q_{\rm L}^{-1} - Q_{\rm C}^{-1}\right) \pi / \lambda \tag{1}$$

where a is absorptivity of the gas mixture in Nepers km<sup>-1</sup>. (Note, for example, that an attenuation constant or absorption coefficient or absorptivity of 1 Neper km<sup>-1</sup> = 2 optical depths per km (or km<sup>-1</sup>) = 8.686 dB km<sup>-1</sup>, where the first notation is the natural form used in electrical engineering, the second is the usual form in physics and astronomy, and the third is the common (logarithmic) form. The third form is often used in order to avoid a possible factor-of-two ambiguity in meaning.)  $Q_L$  is the quality factor of the cavity resonator when the gas mixture is present,  $Q_C$  is the quality factor of the resonance in a vacuum, and  $\lambda$  is the wavelength (in km) of the test signal in the gas mixture.

In the first half of this grant year, we have made high accuracy measurements of the 7.5 to 9.3 mm absorption from gaseous ammonia  $(NH_3)$  in a 90%  $H_2/10\%$  He atmosphere. at a temperature of 203 K. While even lower temperatures could be achieved, the need to avoid the risk of ammonia condensation kept our operating temperatures relatively high. As in the previous experiments, the bandwidth and center frequencies of each of several

resonances between 32 and 40 GHz were measured in a vacuum. Next, 28 torr of gaseous ammonia is added to the system. The pressure of the ammonia gas is measured with the high-accuracy thermocouple vacuum gauge, as shown in Figure 2.

In addition, the ammonia abundance can be monitored by measuring refractivity of the introduced gas. Since the index of refraction (relative to unity) at low pressure is proportional to the ammonia gas abundance, the ability of the system to accurately measure refractivity (through measurement of the frequency shift of resonances) can be used to infer the relative vapor abundance or pressure. Note that it is not yet possible to use this approach for the accurate determination of absolute NH<sub>3</sub> pressure since accurate refractivity data for the 7.3 to 10 mm wavelength range is not available. (In fact, by using our thermocouple vacuum gauge, we have made measurements of the density-normalized refractivity of gaseous ammonia at 39 GHz, and found it to be 6.1 x  $10^{-17}$  N-units/molecule/cm<sup>3</sup>, which is nearly 6 times the value at optical wavelengths.)

Next, 1.8 atm of hydrogen  $(H_2)$  and 0.2 atm of helium (He) are added to the chamber, bringing the total pressure to 2 atm. The bandwidth of each resonance is then measured and compared with its value when the chamber was evacuated in order to determine the absorptivity of the gas mixture at 2 atm total pressure. The total pressure is then reduced, by venting, to 1 atm, and the bandwidths are again measured. Finally, the pressure vessel is again evacuated and the bandwidths again measured so as to assure no variation of the Q's of the evacuated resonator has occurred. As with the previous measurements, the measured changes of bandwidths (Q's) can then be used to compute the absorptivity of the gas mixtures at each of the resonant frequencies.

This approach has the advantage that the same gas mixture is used for the absorptivity measurements at the various pressures. Thus, even though some small uncertainty may exist as to the mixing ratio of the initial mixture, the mixing ratios at all pressures are the same, and thus the uncertainty for any derived pressure <u>dependence</u> is due only to the accuracy limits of the absorptivity measurements, and not to uncertainty in the mixing ratio. (This assumes that the mixing ratio is small, so that foreign-gas broadening predominates, as is the case for our measurements.) Similarly, measurements of the frequency <u>dependence</u> of the absorptivity from the mixture would likewise be immune to any mixing ratio uncertainty, since foreign-gas broadening predominates.

For the measurements described, the amount of absorption being measured is extremely small. Thus, any errors in measurements of (or other changes in) the apparent bandwidth of the resonances, not caused by the absorbing gases, could lead to significant errors in the absorption measurement. The contribution of instrumental errors and noise-induced errors on such absorptivity measurements have been discussed at length in Steffes and Jenkins (1987). However, because our latest measurements represent such small percentage changes in bandwidth, another instrumental source of error which we refer to as dielectric loading becomes a concern.

As can be seen in Figure 2, the resonator, which operates as a bandpass filter, is connected to a signal source (the millimeter-wave sweep oscillator) and to a signal receiver (the high-resolution spectrum analyzer). The "Q" of the resonator, which is defined as the ratio of energy stored in the resonator to the energy lost per cycle, equals the ratio of resonant center frequency to resonance half-power bandwidth. It is not surprising, therefore, that the

stronger the coupling between the resonator and the spectrum analyzer or sweep oscillator, the lower will be the Q of the resonance, since more energy will be lost per cycle through the waveguides connecting the resonator to the spectrum analyzer and sweep oscillator. For this reason, we have always designed our resonators (both the coaxially-coupled cylindrical cavity resonators used below 25 GHz and the waveguide-coupled Fabry-Perot resonators used above 30 GHz) with minimal coupling, so as to maximize Q and to minimize the changes in Q that might result from changes in coupling that occur when gases are introduced into the resonators. It should be noted that these changes in coupling, which are due to the presence of the test gas mixtures, are not related to the absorptivity of the gases, but rather to the dielectric constant or permittivity of the test gas mixtures. (Hence, the term "dielectric loading.")

We have always strived to design the coupling elements of the resonators so that the changes in lossless test gas abundances (and resulting changes in dielectric constant) had little or no effect on the Q of the resonator as measured in the system. This has been no small feat in that slight imperfections in resonators, cables, coupling loops, or waveguides can make the apparent Q of the resonator appear to vary with the abundance of such lossless gases. It has now become a standard part of our experimental procedure to repeat absorption measurements for gas mixtures in which the absorbing gas is a minor constituent, without the absorbing gas present. For example, after measurements were made of the microwave and millimeter-wave absorption from ammonia as a minor constituent in an  $H_2/He$  atmosphere, measurements of the apparent absorption of the  $H_2/He$  atmosphere without the ammonia gas were made. Since, for the pressures and wavelengths involved, the  $H_2/He$  atmosphere is

essentially transparent, no absorption was expected. If any apparent absorption was detected, "dielectric loading," or a change in coupling due to the dielectric properties of the gases, was indicated.

Initially, if any evidence of dielectric loading existed, the experiments were terminated and the apparatus disassembled, including pressure seals. The cables and coupling loops were then readjusted, and the system reassembled and tested again. The entire procedure was repeated until the dielectric loading effect was eliminated or minimized. If some small variation in the resonant Q or bandwidth due to the presence of the non-absorbing gases still remained, it was added to the uncertainty or error bars for each experiment. More recently however, we have found that the effects of dielectric loading are additive, in that they add to the apparent changes of resonator bandwidth caused by the absorbing gases. Thus, as long as the effects of dielectric loading are not time variable, they can be removed by using the measured value of the Q of a resonance with the non-absorbing gases present (instead of the Q of the resonance in a vacuum) for the quantity  $Q_c$  in equation (3).

Another potential source of instrumental error which we have recently detected has to do with nonlinearities in the spectrum analyzer display. We have found that depending on the vertical position of the bandpass spectrum on the spectrum analyzer CRT display, the peak signal level (and, therefore, the apparent half-power bandwidth and resulting quality factor) of the resonator seems to vary slightly. This is due to nonlinearities in the CRT vertical deflection amplifier. We have minimized this potential error by always resetting the vertical position of the displayed spectrum to the same portion of the CRT screen.

As previously discussed, once a test gas mixture is formed in the pressure vessel, the same mixture is used for measurement of absorption at several frequencies and pressures. Thus, even though some uncertainty in absolute mixing ratio exists, the pressure and frequency dependences of the millimeter-wave absorption can be measured to high accuracy. However, in order to properly characterize the magnitude of the absorption, the ammonia mixing ratio must be known precisely. Using the high accuracy thermocouple vacuum gauge shown in Figure 2, the actual NH, mixing ratio can only be determined to an accuracy of  $\pm 20\%$  of its value. (Note: This corresponds to  $(1.85 \pm 0.37)$ % NH<sub>3</sub> volume mixing ratio.) However, since our required overall accuracy for the NH, absorptivity measurement is ±20%, this mixing ratio uncertainty is excessive. In order to reduce this uncertainty, we arranged for a local gas products supplier (Matheson Gas Products) to provide us with a pre-mixed hydrogen/helium/ammonia atmosphere which was analyzed with a mass spectrometer so that mixing ratio accuracies of better than 2% (i.e., (1.85 ± .04)%) were obtained. We have used this mixture (1.85% NH<sub>3</sub>, 9.81% He, and 88.34% H<sub>2</sub>) for the high accuracy absorptivity measurements which are required to accurately infer ammonia abundance from millimeter-wave opacity data for the Jovian planets.

This same "custom" gas mixture has been measured under the same conditions (total pressures of 1 and 2 Bars and temperature 203 K), using the same techniques, at a single resonance near 94 GHz (3.2 mm). However, because the absorptivity of ammonia is less at 3.2 mm than in the 7.5-9.3 mm wavelength range, the percentage accuracy of the measurements have been worse, even though the quality factor of the 3.2 mm resonator is higher. As a result, a second "custom" gas mixture has been obtained with an even higher

ammonia mixing ratio  $(5.07\% \text{ NH}_3$ . 85.56% H<sub>2</sub>, 9.37% He). Because of the higher ammonia mixing ratio (and the need to avoid condensation), measurements of the 94 GHz absorptivity of this new mixture have been conducted at a slightly higher temperature (210 K) for total pressures of 1, 1.3, and 2 Bars.

### IV. RESULTS OF LABORATORY MEASUREMENTS AND THEIR APPLICATION

Initial measurements of the 7.5 to 9.3 mm absorptivity from NH<sub>3</sub> in a hydrogen/helium atmosphere were conducted at 203 K as described in Section III, with an ammonia mixing ratio of 0.0186, at pressures of 1 and 2 Bars. An examination of these early experimental results revealed that because of large error bars, we could not determine whether the modified Ben-Reuven lineshape best described the absorption profile of gaseous ammonia shortward of 1 cm, as discussed in Sections I and II. As a result, we undertook higher accuracy measurements in the first half of this current grant year.

Results of measurements of the 32 to 40 GHz (7.5 to 9.3 mm) absorptivity of gaseous ammonia under simulated Jovian conditions (203 K) are shown in Figure 5. These measurements were made using a 88.3% hydrogen/9.81% helium atmosphere with a total pressure of 2 Bars. The ammonia mixing ratio was 0.0185. Triangular points represent measurements of gas mixtures formed using the thermocouple vacuum gauge (NH<sub>3</sub> mixing ratio accuracy =  $\pm 20\%$  of its value) and the circular points represent measurements of the pre-mixed, analyzed gas mixture described in Section III. With this mixing ratio, temperatures as low as 190 K could have been used before saturation would have become a problem, but 203 K was used so as to be consistent with earlier measurements.

are solid lines which represent Also. shown in Figure 5 the theoretically-computed opacity using the Van Vleck-Weisskopf lineshape (upper line), the modified Ben-Reuven lineshape (middle line), and the Zhevakin-Naumov lineshape (lower line). The Van Vleck-Weisskopf calculation was performed using linewidths and line intensities as per Wrixon et al. (1971). The Ben-Reuven calculation was made as per Berge and Gulkis (1976), by employing a Ben-Reuven lineshape which has been modified so as to be consistent with the laboratory results of Morris and Parsons (1970), in which the 9.58 GHz absorption from NH<sub>3</sub> (in a high pressure  $H_2/He$  atmosphere) at room temperature was measured. The Zhevakin-Naumov calculation used the lineshape of Zhevakin and Naumov (1967) and linewidths and line intensities from Wrixon These theoretical spectra were computed using generalized et al. (1971). computer programs for which the partial pressures from H2, He, and NH3, as well as frequency and temperature, were adjustable variables. The values picked for these variables matched our experimental conditions.

Inspection of the results in Figure 5 shows that most of the measured data points lie nearest to the theoretically-derived absorptivity expression based on the Zhevakin-Naumov lineshape. This differs from our preliminary measurements which suggested that the ammonia opacity might actually exceed that indicated by the modified Ben-Reuven lineshape. Such results are not surprising, however, in that the accuracy of the new measurements is far greater than that from previous measurements. It is also noteworthy that other researchers, such as de Pater and Massie (1985), have found that in order to best explain the 1-10 mm Jupiter emission spectrum, a different sort of lineshape was needed to characterize the ammonia capacity. Likewise, previous laboratory measurements of NH<sub>3</sub> opacity at frequencies below 22 GHz

showed opacities slightly less than those indicated by the modified Ben-Reuven lineshape under the same conditions of temperature and pressure (Steffes and Jenkins, 1987). However, it has been noted by Spilker (private communication) that slight changes to the modified Ben-Reuven formulation can be made which make it more consistent with longer wavelength laboratory results, and make it essentially identical with the Zhevakin-Naumov formulation at frequencies between 32 and 40 GHz (see discussion below).

Our preliminary results at 94 GHz, shown in Figure 6, definitely favor the modified Ben-Reuven formalism. When the data points for opacity are compared with the theoretically-computed opacity using the Van Vleck-Weisskopf lineshape (upper line), the modified Ben-Reuven lineshape (middle line), and the Zhevakin-Naumov lineshape (lower line), it becomes clear that the modified Ben-Reuven lineshape best characterizes the opacity at 3.2 mm under the conditions of this measurement (Total Pressure: 2 Bars; Temperature: 213 K; 5.07% NH, 85.56% H, 9.37% He). Mixing Ratios: Note that these three theoretical computations were made using the same techniques used for the 7.5-9.3 mm calculation. This result is especially significant in that it contradicts the suggestion by de Pater and Massie (1985) that ammonia opacity at wavelengths shortward of 7 mm must be substantially greater than indicated by the modified Ben-Reuven lineshape. Instead it suggests an ammonia abundance distribution different than that proposed, or the presence of other millimeter-wave absorbing constituents, such as was suggested by Bézard (1983).

Our preliminary results at 94 GHz are also significant in that they are not consistent with the Zhevakin-Naumov lineshape, and therefore, confirm the suggestion by Spilker and Eshleman (1988) that a further modification to Ben-

Reuven lineshape best describes the microwave absorption spectrum of  $NH_3$  under Jovian conditions. However, it should be noted that this only applies at pressures of 2 Bars or greater. At pressures well below 2 Bars, it is expected that the absorption of  $NH_3$  will revert to that computed using the Van Vleck-Weisskopf formulation, which will be especially noticeable at frequencies far from 1-2 cm inversion resonances. Therefore, we hope to be able to characterize this transition from the modified Ben-Reuven lineshape to the Van Vleck-Weisskopf lineshape by further study of the 94 GHz opacity of  $NH_3$  (in an  $H_2/He$  atmosphere) in the 1 to 2 Bar pressure range.

## V. OBSERVATIONAL AND INTERPRETIVE STUDIES

As described in the previous Annual Status Report for Grant NAGW-533 (February 1, 1987 through January 31, 1988), studies of our recent measurements of the 1.35 to 3.6 cm emission from Venus have suggested that long term temporal and/or significant spatial variations in the abundance of SO<sub>2</sub> and gaseous H<sub>2</sub>SO<sub>4</sub> may occur immediately below the main cloud layer (48 km and Our observation, which was predominantly of equatorial and midbelow). latitude regions of Venus, indicated a significantly lower SO<sub>2</sub> abundance than was measured in 1978 by the Pioneer-Venus Sounder Probe, and a lower average abundance of gaseous H<sub>2</sub>SO<sub>4</sub> than would have been inferred from earlier Pioneer-Venus radio occultation studies of subcloud opacity at 13 cm. Some or all of this difference may be due to spatial variations in the subcloud H<sub>2</sub>SO<sub>4</sub> abundance since most of the early Pioneer-Venus results were for polar latitudes (Cimino, 1982). Similarly, our results may be consistent with the earlier equatorial 13 cm radio occultation opacity measurements made with the Mariner 10 spacecraft (Lipa and Taylor, 1979), where the peak opacity would

correspond to a very large abundance of gaseous  $H_2SO_4$ , but the average subcloud opacity was significantly lower.

One important tool for evaluating these effects is the reduction of the microwave data from the 1986-87 Pioneer-Venus radio occultation measurements. This data was taken over a wide range of latitudes and could be critical for determining whether temporal variations or spatial variations in gaseous  $H_2SO_4$  abundance could be occurring. Working with Dr. Arvydas J. Kliore (P-V Radioscience Leader), our group has obtained the currently unreduced data and (working at JPL) begun reducing the data to obtain absorptivity profiles for the 1986-87 epoch.

Over the next year, we will reduce data obtained from the Fall 1986/ Winter 1987 Pioneer-Venus Radio Occultation Observations. Since the initial conversion from amplitude and doppler (frequency) data to refractivity and absorptivity profiles can be most efficiently completed at JPL, we have made arrangements to send graduate students to JPL for this activity. Support for travel and student salaries for the reduction effort at JPL has been obtained from the Pioneer-Venus Guest Investigator Program.

After the initial reduction, we hope to have dependable 13 cm wavelength refraction and absorption profiles for a range of altitudes in the Venus atmosphere reaching down to 38 km and for latitudes ranging form equatorial to polar. Figure 7 shows a preliminary 13 cm absorptivity profile derived from radio data obtained during the entry occultation of Orbit 2801 on August 6, 1986. (Note: This is the first such absorptivity profile which has been derived since Orbit 358 - November 28, 1979). This occultation probed the Venus atmosphere at 52°N latitude and the ray path traversed mainly the night side of the planet. For this orbit, the spacecraft signal was only receivable

at the Goldstone DSS-14 receiver down to a periapsis altitude of 43.7 km before receiver lock was lost. (This assumes a planetary radius of 6052 km.) It is hoped that other orbits may be found which probe deeper into the atmosphere before loss of signal, but because the Pioneer-Venus steerable antenna no longer tracks the limb of the planet (the direction the radio ray travels back to earth) during the occultation, the resulting lower signal level may prevent probing deeper than the 40 km altitude.

To show the usefulness of the 13 cm opacity data for inferring the nature of the gaseous  $H_2SO_4$  abundance, we compare in Figure 8 the measured absorptivity profile from orbit 2801N with that absorptivity which would result from a saturation abundance of gaseous  $H_2SO_4$  (from Steffes, 1985) in the 43 to 55 km altitude range. It can be seen that for altitudes in the 49 to 51 km altitude range (the nominal altitude range of the Venus lower cloud - see Ragent and Blamont, 1980), absorptivity values close to those caused by a saturation abundance of gaseous  $H_2SO_4$  are seen. At lower altitudes, most values for absorptivity are below those caused by a saturation abundance. It should be noted that error bars for this preliminary absorptivity data have not yet been computed, but are expected to be on the order of  $\pm 0.001$  dB/km  $(\pm 0.00023 \text{ km}^{-1})$ .

Figure 9 shows the full extent to which application of our laboratory results can be carried. The abundance of gaseous  $H_2SO_4$  (derived from the absorptivity profile in Figure 7 by using laboratory results from Steffes, 1985) is plotted as a function of altitude, along with a plot of the saturation abundance of gaseous  $H_2SO_4$ , for comparison. As our work in the Pioneer-Venus Guest Investigator Program yields more absorptivity profiles for a wide range of locations in the Venus atmosphere, we hope to be able to well

characterize the abundance, structure, and spatial variations of gaseous  $H_2SO_4$ in the Venus atmosphere. We also hope to make comparative studies with earlier radio astronomical and radio occultation measurements in order to detect possible temporal variations in  $H_2SO_4$  abundance and structure.

Also, our new results for the 3.2 mm opacity of ammonia under Jovian conditions holds promise for new interpretive studies and possibly some new observational studies of the millimeter-wave emission spectra from the outer planets. We include further discussion of this in the attached proposal.

# VI. PUBLICATIONS AND INTERACTION WITH OTHER INVESTIGATORS

In the first half of the current grant year, a paper was completed and accepted for publication in <u>Icarus</u>, describing results and applications of some experiments performed during the previous year of Grant NAGW-533 (Jenkins and Steffes, 1988a). This paper describes laboratory measurements of the microwave absorption of methane  $(CH_4)$  and water vapor  $(H_2O)$  under simulated Jovian conditions. The paper also concludes, that based on these laboratory results, neither methane nor water vapor can be responsible for the excess microwave opacity detected at wavelengths between 10 and 20 cm in the atmosphere of Jupiter. This supports the presence of an ammonia abundance which exceeds solar abundance by a factor of 1.5 in the 2 to 6 Bar levels in Jupiter's atmosphere.

In addition, as discussed in Section I, we completed a paper describing observations and interpretive studies of the 1.3 to 3.6 cm Venus emission spectrum (Steffes <u>et al.</u>, 1988). Likewise, a paper describing the results and applications of the laboratory measurements of the millimeter-wave opacity of ammonia described in Section IV has been submitted for publication in Icarus

(Joiner <u>et al.</u>, 1988a). We also submitted updated summaries of our most recent laboratory measurements for inclusion in the twenty-second issue of the Newsletter of Laboratory Spectroscopy for Planetary Science.

We attended the 20th Annual Meeting of the Division for Planetary Sciences of the American Astronomical Society the week of October 30 through November 4. In addition to presenting our work involving the Venus studies (Jenkins and Steffes, 1988b), our latest laboratory results for the millimeter-wave opacity from ammonia were presented by GRA Joanna Joiner (Joiner et al., 1988b -- abstract attached).

In addition to the observations of Venus and analysis work conducted jointly with Dr. Michael J. Klein of JPL, we have also worked with Dr. Michael A. Janssen of JPL regarding models for the Venus atmosphere, interpretation of microwave emission measurements, and theoretical models for the absorption spectrum of  $H_2SO_4$ . A presentation of our laboratory results for simulated Venus and Jovian atmospheres was given at JPL on September 9, 1988. We have also worked with Dr. Arvydas J. Kliore of JPL on the reduction and interpretation of data from recent Pioneer-Venus Radio Occultation Studies as part of our involvement in the Pioneer-Venus Guest Investigator Program. More informal contacts have been maintained with groups at the California Institute of Technology (Dr. Duane O. Muhleman and his students, regarding interpretation of radio astronomical measurements of Venus and the outer planets), at the Stanford Center for Radar Astronomy (V. R. Eshleman, G. L. Tyler, and T. Spilker, regarding Voyager results for the outer planets, and laboratory measurements), and at JPL (Drs. Robert Poynter and Samuel Gulkis, regarding radio astronomical observations of the outer planets and Venus). We have also worked with Dr. Imke de Pater (University of California-Berkeley) by using our

laboratory measurements of atmospheric gases in the interpretation of radio astronomical observations of Venus and the outer planets. We have also studied possible effects of the microwave opacity of cloud layers in the outer planets atmospheres. In this area, we have worked both with Dr. de Pater and with Dr. Paul Romani (NRC Associate, Goddard SFC).

Dr. Steffes has also been active in the review of proposals submitted to the Planetary Atmospheres Program at NASA (both as a "by-mail" reviewer and as a member of both the March 1988 and September 1988 review panels) and as a reviewer of manuscripts submitted to Icarus and the Journal of Geophysical Research, for which Dr. Steffes is an Associate Editor. We have also continued to serve the planetary community through the distribution of reprints of our articles describing our laboratory measurements and their application to microwave and millimeter-wave data from planetary atmospheres. The results of these measurements have been used in the mission planing for radio and radar systems aboard the Galileo and Magellan missions, and more recently, for proposed experiments for the Cassini mission. Dr. Steffes also participated as a member of the International Jupiter Watch (IJW) Laboratory/ Theory Discipline Team. Another source of close interaction with other planetary atmospheres principal investigators has been Dr. Steffes' membership in the Planetary Atmospheres Management and Operations Working Group (PAMOWG). Travel support for Dr. Steffes' attendance at PAMOWG meetings, as well as the November AAS/DPS meeting, has been provided by Georgia Tech in support of Planetary Atmospheres Research. Also in support of Planetary Atmospheres Research, Georgia Tech provided \$2,000 for required repairs and maintenance to the ultra-cold freezer system used in the outer planets atmospheric simulator.

As in the past, we have maintained contact with members of the Georgia congressional delegation, keeping them aware of our work and aware of our continued support for the solar system exploration program. We were especially pleased with the support received from Senator Wyche Fowler for the CRAF/Cassini "new start," after briefing his staff on this issue.

## VII. CONCLUSION

In the first half of this grant year, we continued to conduct laboratory measurements of the millimeter-wave properties of atmospheric gases under simulated conditions for the outer planets. Significant improvements in our system made it possible to accurately characterize the opacity from gaseous NH<sub>3</sub> at longer millimeter-wavelengths (7.5 to 9.3 mm) under simulated Jovian conditions. In the second half of this grant year, we extended such measurements to even shorter millimeter-wavelengths (3.2 mm). The preliminary results of these measurements have been significant in that they seem to indicate that the large opacities predicted by a number of workers at these wavelengths are indeed incorrect and that a form of the modified Ben-Reuven formalism for computing the millimeter-wave opacity from ammonia is correct. In the next nine months of the grant, we will complete this study. We will also pursue further analysis and application of our laboratory results to microwave and millimeter-wave absorption data for the outer planets, such as results from Voyager Radio Occultation experiments and earth-based radio astronomical observations. We also intend to pursue the anlaysis of available multi-spectral microwave opacity data from Venus, including data from our most recent radio astronomical observations in the 1.3 to 3.6 cm wavelength range and newly obtained Pioneer-Venus Radio Occultation measurements at 13 cm, using our laboratory measurements as an interpretive tool.

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Figure 1: Diagram of Fabry-Perot Resonator



FIGURE 3: DIAGRAM OF FABRY-PEROT RESONATOR (94GHZ)





™ 5 Absorption of NH3 in a -35-88.34% H2,9.81% He, 1.85% NH3 hixture (Mixing ratio:0.0185±0.0005) Pressure:2 atm. Temp:203K





Figure 6: Absorptivity of gaseous mixture (5.07% NH<sub>3</sub>, 9.37% He, and 85.56% H<sub>2</sub>) under simulated Jovian conditions (Total pressure: 2 Bars, Temperature: 210 K). Shown for comparison (horizontal lines), are the theoretically computed values from the Van Vleck-Weisskopf formalism (top line), the modified Ben Reuven formalism (middle line), and the Zhevakin-Naumov formalism (lower line).



Figure 8: Comparison of absorptivities measured with radio occultation technique (circular points -- from Pioneer-Venus Orbit 2801entry occultation) with absorptivity which would result from saturation abundance of H<sub>2</sub>SO<sub>4</sub> (from Steffes, 1985). The absorption coefficient scale is logarithmic (exponents of 10). All measurements were made at the 13-cm wavelength (2.293 GHz).



Figure 9: Abundances of gaseous  $H_2SO_4$  inferred from Pioneer-Venus 13-cm absorptivity profiles (circular points) compared with the saturation abundance profile of gaseous  $H_2SO_4$  (from Steffes, 1985). The mixing ratio scale is logarithmic (exponents of 10).



X. APPENDIX

## <u>Millimeter-Wave Measurements of the Opacity of</u> <u>Gaseous Ammonia (NH<sub>3</sub>) Under Simulated Conditions</u> for the Jovian Atmosphere

J. Joiner, J.M. Jenkins, P.G. Steffes (Georgia Institute of Technology)

Last year, preliminary results of laboratory measurements of the opacity of gaseous ammonia  $(NH_3)$  from 32-40 GHz (7.5 - 9.3 mm) in an H<sub>2</sub>/He atmosphere were presented. These measurements were conducted at a pressure of two atmospheres, a temperature of 203K, and a mixing ratio of However, due to large uncertainties, 0.0185. the results were inconclusive as to which theoretical lineshape best described the observed behavior under these conditions. Over the past year, we have been able to significantly reduce the uncertainties in these measurements. Our final results show that the Zhevakin-Naumov lineshape best describes the absorption of ammonia under these conditions between 32 and 40 GHz. Thus, both the Ben-Reuven lineshape and the VanVleck-Weisskopf overstate the absorption due to ammonia in this region. We also plan to make similar measurements at 94 GHz (3.2mm), where several radio emission studies have been made and even larger variations from theoretically derived lineshapes are expected.

This work was supported by the Planetary Atmospheres Program of the National Aeronautics and Space Administration under Grant NAGW-533.

Division for Planetary Sciences - Austin meeting erican Astronomical Society le Submitted



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Renewal Proposal

and Semiannual Status Report #11

to the

THE NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

for

Grant NAGW-533

#### LABORATORY EVALUATION AND APPLICATION OF

## MICROWAVE ABSORPTION PROPERTIES UNDER SIMULATED

## CONDITIONS FOR PLANETARY ATMOSPHERES

in Response to

NRA 89-OSSA-5

Report Period: February 1, 1989 through April 30, 1989 Proposed Renewal Period: November 1, 1989 through October 31, 1990 Requested Funding Level: \$70,887 (FY90)

Submitted by

Professor Paul G. Steffes School of Electrical Engineering Georgia Institute of Technology Atlanta, Georgia 30332-0250 (404) 894-3128

March 1989
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COVER SHEET

RTOP No Grant/Contract No Date Received				
• NATIONAL AERONAUTICS AND SPACE ADMINISTRATION SOLAR SYSTEM EXPLORATION DIVISION PLANETARY ATMOSPHERES PROGRAM				
X Full Proposal Progress Report				
TITLE: Laboratory Evaluation and Application of Microwave Absorption Properties under Simulated Conditions for Planetary Atmospheres TYPE OF ORGANIZATION*): University				
RESEARCH AREA**): Microwave/Millimeter-wave Laboratory Measurements				
DATE SUBMITTED: March 27, 1989				
DESIRED STARTING AND ENDING DATES: 11/1/89 10/31/92				
PRINCIPAL INVESTIGATOR:				
Name: Paul G. Steffes				
Title: Associate Professor				
Address: School of Electrical Engineering Georgia Institute of Technology Telephone: Atlanta, GA 30332-0250 (404) 894-3128				
AUTHORIZING INSTITUTIONAL OFFICIAL:				
Name: R. Dennis Farmer				
Title: Contracting Officer				
Address: Georgia Tech Research Corporation Centennial Research Building				
Georgia Institute of Technology Telephone: Atlanta, GA 30332-0420				
(404) 894-4817 *)e.g., profit, nonprofit, university, other educational institutions, etc.				
<pre>**)Please indicate main research area, such as structure, dynamics, composition, aeronomy, laboratory measurements, other.</pre>				

1

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### B. COGNIZANT PERSONNEL

For scientific or technical matters relating to the contract (Principal Investigator):

Paul G. Steffes School of Electrical Engineering Georgia Institute of Technology Atlanta, Georgia 30332-0250 Telephone: (404) 894-3128

For contractual and business matters:

٠

Georgia Tech Research Corporation Attn: R. Dennis Farmer Centennial Research Building Georgia Institute of Technology Atlanta, Georgia 30332-0420 Telephone: (404) 894-4817

### C. PROPOSAL SUMMARY

TITLE: Laboratory Evaluation and Application of Microwave Absorption Properties Under Simulated Conditions for Planetary Atmospheres PRINCIPAL INVESTIGATOR: Paul G. Steffes (Georgia Institute of Technology)

### ABSTRACT

. Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave and millimeter-wave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often leads to significant misinterpretation of available opacity data. The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by both radio occultation experiments and radio astronomical observations, and over a range of frequencies which correspond to those used in both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has been the goal of this investigation to conduct such measurements and to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profile of constituents in those planetary atmospheres.

This document contains an Introduction and Summary (Section II) and a Proposal (Section III), which together form a "stand alone" Renewal Proposal. Section IV contains Semiannual Status Report #11 for this grant (NAGW-533), which provides other useful information about this research program.

The key activities proposed for the next grant year include:

- Complete development of generalized formulation for the microwave and millimeter-wave opacity of gaseous NH<sub>3</sub> under Jovian conditions, based on laboratory results.
- 2. Develop radiative transfer models for microwave and millimeter-wave emission from Jovian planets. Decide whether absorption from gases other than NH<sub>3</sub> (and clouds) is significant enough to require additional laboratory measurements.
- 3. Develop and perform laboratory measurement of the "dissociation factor" for gaseous  $H_2SO_4$ . Use results to correct existing expressions for the microwave opacity from gaseous  $H_2SO_4$ .
- 4. Develop and begin laboratory measurements of the millimeter-wave opacity of gaseous H<sub>2</sub>SO<sub>4</sub>.
- 5. Perform comparative studies of microwave and millimeter-wave opacity of Venus atmosphere as inferred from radio emission and radio occultation studies so as to characterize possible spatial or temporal variabilities in the abundance of microwave absorbing constituents.

Several longer term projects, such as laboratory measurements of the microwave properties of Jovian cloud materials, are also discussed.

PRINC	IPAL INVESTIGATOR:	Paul G. Steffes		
TITLE	:	Laboratory Evaluation and Appli Absorption Properties Under Sim Planetary Atmospheres	cation of Micr ulated Conditi	owave ons for
GRANT	NUMBER:	NAGW-533 For the period of November 1, 1 October 31, 1990	.989 through	
		Estimated Cost Breakdown		
1.	DIRECT SALARIES AN	D WAGES*:		\$37,220
	A. Principal Inv P.G. Steffes	estigator		
	25% time, cal B. 1 Graduate St	endar year (.25 person-years) udent	\$15 <b>,6</b> 20	
	50% time, cal	endar year (.5 person-years) udant	\$12,960	
	A. K. Fahd 33% time, cal	endar year (.33 person-years)	\$ 8,640	
11.	FRINGE BENEFITS**: \$3, 25.5% of Direct Salaries & Wages (less students)		\$ 3,984	
111.	MATERIALS, SUPPLIE	S, AND SERVICES:		\$ 1,300
	A. Gases and Liquid B. Miscellaneous P	ids for Experiments Project Supplies	<b>\$ 8</b> 00	
	and Component	8	<u>\$ 500</u>	
IV.	TRAVEL:			\$ 1,800
	A. Travel for St (Providence,	udents (2) to AAS/DPS Meeting RI, 4 days duration)	\$ 1,800	
		SUBTOTAL - ESTIMATE OF DIRE	CT COSTS:	\$44,304
v.	OVERHEAD (Indirect 60% of Modified To	Expense)**: tal Direct Cost Base		\$26,583

### TOTAL ESTIMATED COST: \$70,887

\*The salary and wage rates are based on current FY89 salaries for the Georgia Institute of Technology. An increase of 10% is estimated for the performance in Georgia Tech FY90. The Georgia Tech Fiscal Year is July 1 through June 30.

#\*Rates are effective for the period July 1, 1988 through June 30, 1989 and are subject to adjustment thereafter upon DCAA audit and ONR negotiations.

# E. LIST OF CURRENT AND PENDING RESEARCH SUPPORT FOR PRINCIPAL INVESTIGATOR (Prof. Paul G. Steffes)

# A. Current Support

1. National Aeronautics and Space Administration - Grant NAGW-533, "Laboratory Evaluation and Application of Microwave Absorption Properties Under Simulated Conditions for Planetary Atmospheres," \$47,616 for 9-month period (2/1/89-10/31/89). P.I. time commitment: 27% (2.4 person-months). .

L

- National Aeronautics and Space Administration, Ames Research Center - Grant NAG 2-515, "Pioneer-Venus Radio Occultation (ORO) Data Reduction: Profiles of 13 cm Absorptivity," \$34,828 for 12-month period (7/1/88-6/30/88). P.I. time commitment: 5% (0.6 personmonths).
- GTE Spacenet Corporation Contract C-10070, "Research in Development of Satellite Interference Location System (SILS) at Georgia Tech," \$32,640 for 11-month period (1/20/89-12/31/89). P.I. time commitment: 10% (1.1 person-months).
- 4. Emory-Georgia Tech Biomedical Technology Research Center, "Research in Development of a Non-Invasive Blood Glucose Monitoring Technique," \$29,000 for 12-month period (7/1/88-6/30-89). P.I. time commitment: 10% (1.2 person-months).
- B. Pending Support (other than this proposal)
  - RENEWAL: National Aeronautics and Space Administration, Ames Research Center - Grant NAG 2-515, "Pioneer-Venus Radio Occultation (ORO) Data Reduction: Profiles of 13 cm Absorptivity," \$34,828 for 12-month period (7/1/89-6/30/90). P.I. time commitment: 12% (1.5 person-months).
  - <u>RENEWAL</u>: Emory-Georgia Tech Biomedical Technology Research Center, "Research in Development of a Non-Invasive Blood Glucose Monitoring Technique," \$29,000 for 12-month period (7/1/89-6/30/90). P.I. time commitment: 10% (1.2 person-months).

absorptivity data for planetary atmospheres obtained from Radio spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often leads to significant misinterpretation of available opacity data. For example, results obtained for the microwave opacity from gaseous H2SO4 under simulated Venus conditions, during the first two years of Grant NAGW-533, showed that not only was the opacity from  $H_2SO_4$  much greater than theoretically predicted, but that its frequency (wavelength) dependence was far different than that theoretically predicted (Steffes, 1985 and Steffes, 1986). Measurements made by Steffes and Jenkins (1987), during the third year of Grant NAGW-533, have shown that the microwave opacity of gaseous ammonia (NH3) under simulated Jovian conditions did indeed agree with theoretical predictions to within experimental accuracy at wavelengths longward of 1.3 cm. Work performed during the fourth and fifth years of Grant NAGW-533 (February 1, 1987 through January 31, 1989) and continuing into this current grant year (February 1, 1989 through October 31, 1989) has shown that laboratory measurements of the millimeter-wave opacity of ammonia between 7.5 mm and 9.3 mm and also at the 3.2 mm wavelength require a different lineshape to be used in the theoretical prediction for millimeter-wave ammonia opacity than had been previously used

(see Joiner <u>et al.</u>, 1989). The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by both radio occultation experiments and radio astronomical observations, and over a range of frequencies which correspond to those used in both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has been the goal of this investigation to conduct such measurements <u>and</u> to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profiles of constituents in those planetary atmospheres.

1

A key activity for the current grant year has continued to be laboratory measurements of the microwave and millimeter-wave properties of the simulated atmospheres of the outer planets and their satellites. As described in the appended <u>Semiannual Status Report</u> for Grant NAGW-533 (February 1, 1989 through April 30, 1989), initial laboratory measurements of the millimeter-wave opacity of gaseous ammonia (NH<sub>3</sub>) in a hydrogen/helium (H<sub>2</sub>/He) atmosphere, under simulated conditions for the outer planets were completed in 1988. These measurements were conducted at frequencies from 32 to 40 GHz (wavelengths from 7.5 to 9.3 mm). It had been found by some (e.g., de Pater and Massie, 1985) that the observed millimeter-wave emission from Jupiter was inconsistent with the millimeter-wave absorption spectrum predicted using the modified Ben-Reuven lineshape for ammonia. In order to investigate this, we developed a Fabry-Perot spectrometer system capable of operation from 32 to 41 GHz. This system can be used at pressures up to 2 Bars and temperatures as low as 150 K, which corresponds closely to the conditions at altitudes in the

Jovian atmosphere most responsible for the observed millimeter-wave absorption. A complete description of this millimeter-wave spectrometer is given in previous <u>Annual Status Reports</u> for Grant NAGW-533, and in Joiner <u>et al.</u> (1989).

Since larger variations from theoretically-derived opacity values were expected at shorter millimeter-wavelengths (see de Pater and Massie, 1985), we have conducted laboratory measurements at wavelengths near 3.2 mm (94 GHz), where a large number of observations of the emission from the outer planets have been made. A description of this new system is presented in the appended <u>Semiannual Status Report</u>. A better knowledge of the millimeter-wave absorption properties of NH<sub>3</sub> is essential, not only to help better characterize the distribution and abundance of ammonia at high levels in Jovian atmospheres, but to make it possible to resolve the contributions from other absorbing constituents such as  $H_2S$  (see Bezard <u>et al.</u>, 1983). This knowledge will be of considerable importance for millimeter-wave instruments proposed for future missions, such as the MSAR (microwave and spectrometer and radiometer) proposed for the Cassini mission.

In some cases, new observations or experiments have been suggested by the results of our laboratory measurements. For example, this facility was initially developed, and then operated, in order to evaluate the microwave absorbing properties of gaseous sulfuric acid  $(H_2SO_4)$  under Venus atmospheric conditions. The results, obtained at 13.4 cm and 3.6 cm wavelengths, were applied to measurements from Mariner 5, Mariner 10, and early Pioneer-Venus Radio Occultation experiments, to determine abundances of gaseous sulfuric acid in the Venus atmosphere (Steffes, 1985). Further laboratory measurements also suggested that a substantial variation in the Venus microwave emission,

related to the abundance of gaseous sulfuric acid, might exist near the 2.2 cm wavelength. Since no observations of the Venus emission at this wavelength had ever been published, we conducted observations of Venus using the 140-foot NRAO telescope and the 64-meter DSN/Goldstone antenna in April 1987 to not only search for the presence of the predicted feature, but to use such a feature to determine a planet-wide average for sulfuric acid vapor abundance below the main cloud layer. The results of this observation were substantial in that they not only placed limits on the abundance and spatial distribution of gaseous  $H_2SO_4$  and  $SO_2$ , but they also suggested some limits to long term temporal variations for the abundance of these two gases.

Recently, we completed calibration and interpretive studies on the data from these observations and submitted a paper entitled, "Observations of the Microwave Emission of Venus from 1.3 to 3.6 cm," by P. G. Steffes, M. J. Klein, and J. M. Jenkins, to the journal Icarus. One important issue which was discussed in this paper is the discovery that the microwave absorptivity for gaseous  $H_2SO_4$  which was measured by Steffes (1985 and 1986) appears to differ from a theoretical spectrum newly computed from over 11,000 lines by Janssen (personal communication) by a scale factor. That scale factor suggests that the theoretically-derived "dissociation factor" for gaseous  $H_2SO_4$  (i.e. the percentage of  $H_2SO_4$  which breaks down to form  $SO_3$  and  $H_2O$ ) may have been underestimated. This results in an underestimation of the absorption from gaseous  $H_2SO_4$ . Therefore, an experiment is being planned which will correctly evaluate the "dissociation factor" and thus allow unambiguous calibration of laboratory data for  $H_2SO_4$  opacity (see Section III). This will be critical for the proper interpretation of a wide range of opacity data.

Another important tool for evaluating potential spatial and temporal variations in abundance and distribution of gaseous  $H_2SO_4$  is the reduction and malysis of recently obtained Pioneer-Venus radio occultation measurements. The 13 cm microwave absorptivity profiles, which can be obtained from the adio occultation data, are closely related to the abundance profiles for aseous H<sub>2</sub>SO<sub>4</sub>. Starting in June 1988, we began the reduction of the 1986-87 ioneer-Venus radio occultation measurements (working at JPL with support from he Pioneer-Venus Guest Investigator Program) in order to obtain the needed 3 cm microwave absorptivity profiles. Yet another important source of information is the increasing number of high-resolution millimeter-wavelength Jenus emission measurements which have been recently conducted. Correlative studies of these measurements with radio occultation measurements and our onger wavelength emission measurements (Steffes et al., 1989) should provide. the necessary data for characterizing temporal and spatial variations in the abundance of gaseous  $H_2SO_4$  and for modeling its role in the subcloud itmosphere. However, unambiguous results will require that we have dependable mowledge of the equilibrium between gaseous  $H_2SO_4$ ,  $SO_3$ , and  $H_2O$ , both so as o properly interpret laboratory measurements of the microwave and millimeterave opacity of the gases which elute from liquid sulfuric acid, as well as to odel their relation within the Venus atmosphere.

In the next year of Grant NAGW-533, we propose to use the results of our aboratory analysis of the millimeter-wave absorption from gaseous NH<sub>3</sub> under mulated Jovian conditions to complete a formulation which accurately edicts the opacity from gaseous ammonia in a Jovian-type atmosphere over the tire 3 mm to 20 cm wavelength range (frequencies from 1.5 to 100 GHz). With ch a formulation at our disposal, we will then develop models for microwave

and millimeter-wave emission from the Jovian planets and adjust ammonia abundance profiles so as to match the emission spectrum observed from earth-based radio telescopes. (We should be able to take advantage of the availability of data from several millimeter-wave radio telescope arrays in order to develop localized ammonia abundance profiles over the entire disk of one or more Jovian planets.) Further discussion of this and other planned work related to the outer planets is presented in Section III (Proposal). Likewise, we will continue to make laboratory measurements which will support our interpretive work of the 13 cm absorptivity profiles in the Venus atmosphere, which we are developing as part of the Pioneer-Venus Guest Investigator Program, as well as interpreting microwave and millimeter-wave emission measurements of Venus. These data sets will be invaluable for characterizing the spatial and temporal variabilities of H<sub>2</sub>SO<sub>4</sub> in the Venus atmosphere (see Section III). Finally, we will complete designs for laboratory instrumentation which will allow us to measure the microwave and millimeter-wave properties of liquids and solids under simulated planetary conditions.

#### III. PROPOSAL

### A. Outer Planets Studies

As described in the appended <u>Semiannual Status Report</u> for Grant NAGW-533, we are currently analyzing our measurements of the 94 GHz opacity of gaseous ammonia (NH<sub>3</sub>) under simulated Jovian conditions. Laboratory measurement and analysis of the opacity of two different gas mixtures (1.85% NH<sub>3</sub>, 9.81% He, and 88.34% H<sub>2</sub>) and (5.07% NH<sub>3</sub>, 9.37% He, and 85.56% H<sub>2</sub>) have been conducted in the 203 K - 213 K temperature range and in the 1 to 2 Bar pressure range. These measurements are of importance for a number of reasons:

- They are being conducted at a wavelength (3.2 mm) near which a large number of observations of the emissions from the outer planets have been made. Thus, they are critical for proper interpretation of the atmospheric opacity inferred from such observations.
- 2. They are being conducted over a range of pressures which actually exist at the altitudes of the peak "emission weighting functions" for this wavelength range.
- 3. The measurements are being conducted in a pressure range over which the theoretical formalism for computing the opacity shifts from the Van Vleck-Weisskopf formalism (pressures less than 1 Bar) to the modified Ben Reuven formalism (pressures greater than or equal to 2 Bars). They are also being conducted at a wavelength (3.2 mm) where this shift is most pronounced. (For further discussion, consult the accompanying Semiannual Status Report.)

Therefore, we are deriving from these measurements and our previous measurements of NH<sub>3</sub> opacity a generalized formulation for computing the microwave and millimeter-wave opacity from ammonia under the conditions existing at the altitudes of Jovian atmospheres which are probed at these wavelengths.

Upon completion of the development of the generalized formulation for  $NH_3$  opacity under Jovian conditions, we will then develop a radiative transfer model for microwave and millimeter-wavelengths. Ammonia abundance profiles which result in emission spectra which are consistent with the large number of microwave and millimeter-wave observations will then be derived. (We are currently compiling a data base of such observations.) Should the resulting ammonia abundance profiles appear excessive relative to the chemically

acceptable limits for the Jovian atmosphere, we will pursue further review of the observed emission spectra, and possibly make additional observations, if necessary. However, should we conclude that the observed emission spectra are correct, but inconsistent with a chemically tenable ammonia distribution profile, we will investigate other potential sources of microwave and millimeter-wave opacity. Since gaseous  $H_2S$  and the aqueous  $NH_3$  cloud have been identified as possible sources of additional opacity (especially at millimeter-wavelengths), we will study their absorption properties and evaluate their relative effect on the emission spectra of the outer planets. If it is necessary, we will make additional laboratory measurements of the millimeter-wave opacity from gaseous  $H_2S$ . It will be our long term goal (FY91) to measure the microwave and millimeter-wave opacity from potential cloud condensates (such as aqueous  $NH_3$  solutions) under simulated Jovian conditions.

These laboratory measurements of the microwave and millimeter-wave properties of gaseous constituents will provide a means for accurately interpreting the microwave and millimeter-wave emission spectra of Jovian planets. With the increasing availability of high angular resolution emission measurements of Jovian planets, this will provide an opportunity not only to infer average ammonia abundance profiles for an entire planet, but to infer localized ammonia abundance profiles, as well as localized profiles for other constituents. Thus, it will be possible to better characterize both spatial and temporal variations of the abundance profile for the Jovian planets.

### B. Venus Studies

The same techniques for developing constituent abundance profiles are also applicable for the atmosphere of Venus. However, for Venus we are fortunate to have the additional resource of localized 13 cm absorptivity profiles provided by the Pioneer-Venus Orbiter Radio Occultation (ORO) experiment. As described in the accompanying <u>Semiannual Status Report</u>, we are working under the Pioneer-Venus Guest Investigator Program to reduce radio occultation data obtained over a wide range of locations in the Venus atmosphere so as to obtain 13 cm absorptivity profiles. Since these profiles have been shown to be directly related to the abundance profiles of gaseous  $H_2SO_4$  (Steffes, 1985), it will be possible to study these profiles for spatial variation in the abundance of gaseous  $H_2SO_4$ , and to compare them with earlier 13 cm absorptivity profiles (e.g. Cimino, 1982) in order to infer long-term temporal variations in  $H_2SO_4$  abundance.

Proper interpretation of Venus microwave opacity data inferred from radio occultation and radio emission measurements requires accurate data on the opacity of the microwave-absorbing constituents. Such measurements were conducted (Steffes, 1985 and 1986) at wavelengths from 1.3 to 22 cm, and showed that gaseous  $H_2SO_4$  would be the predominant microwave absorber at altitudes above 30 km in the Venus atmosphere. The unique frequency dependence of the microwave absorption spectrum of gaseous  $H_2SO_4$ , which was measured in these laboratory experiments, prompted a re-evaluation of the theoretical formulation for explaining such opacity. M. A. Janssen (JPL, personal communication) set about such a computation by adding contributions from 11,892 resonant lines, and by assuming the Van-Vleck lineshape with a broadening parameter of 3 MHz for all  $H_2SO_4$  lines in a  $CO_2$  atmosphere.

Typical results for the microwave opacity spectrum of gaseous sulfuric acid using this formulation are shown in Figure 1 (from Steffes <u>et al.</u>, 1989) along with results of laboratory measurements under the same conditions (CO<sub>2</sub> atmosphere, P = 1 Bar, T = 575 K,  $H_2SO_4$  mixing ratio = 0.4%). Comparison of the two results shows a striking correlation in that a peak in opacity occurs near the 2 cm wavelength. Janssen's theoretical formulation differs from the laboratory results in that it indicates an opacity which is greater by a factor of 2 to 8 at the peak, and nearly an order of magnitude greater at longer wavelengths. Some of this difference may be due to the use of the 3 MHz/Torr broadening parameter for all resonant lines in the theoretical calculation. However, since the laboratory data points (including error bars) can all be made consistent with the theoretical spectrum if increased by a scale factor of approximately 8, a scale factor error in either the laboratory result or the theoretical computation is suggested.

It should be remembered that the laboratory measurements of Steffes (1985 and 1986) were conducted by allowing liquid  $H_2SO_4$  to reach vapor pressure equilibrium with an evacuated chamber of known volume. After the experiment was completed, the amount of liquid  $H_2SO_4$  which evaporated to form the vapor was precisely measured, and the  $H_2SO_4$  vapor pressure was inferred.  $CO_2$  was then added and the microwave absorptivity was then measured. Thus, the microwave absorption (shown in Figure 1) was known quite precisely, as was the number of  $H_2SO_4$  molecules which evaporated to form the gas mixture. However, since it was expected that some of the  $H_2SO_4$  molecules in gas phase would dissociate to form gaseous  $SO_3$  and  $H_2O$  (whose microwave absorption is small compared to  $H_2SO_4$ ), the actual  $H_2SO_4$  mixing ratio was uncertain. Steffes (1985 and 1986) assumed that 47% of the vaporized  $H_2SO_4$  dissociated to form



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Figure 1: Comparison of laboratory measurements of the absorptivity spectrum for gaseous  $H_2SO_4$  (solid line, from Steffes, 1986) with that computed theoretically by Janssen and Gulkis (personal communication).

gaseous  $SO_3$  and  $H_2O$ , as computed by Gmitro and Vermeulen (1964) for the concentration used in the experiment. Since the resulting  $H_2SO_4$  vapor pressure expression was consistent with another laboratory measurement which was conducted using a totally different technique (Ayers <u>et al.</u>, 1980), our confidence in the result was increased. However, it should be noted that Ayers <u>et al.</u> were unable to discern  $H_2SO_4$  vapor pressure from  $SO_3$  vapor pressure and were likewise required to use the dissociation factor of Gmitro and Vermeulen (1964).

From this discussion, it becomes clear that a laboratory measurement of the dissociation factor is desperately needed in order to allow for proper interpretation of both future and existing laboratory measurements of opacity and to properly model the gaseous  $H_2SO_4$  saturation abundance in the Venus atmosphere. We propose to conduct such a measurement, using the apparatus shown in Figure 2. In this experiment, a large vacuum chamber and a flask containing a carefully measured quantity of liquid  $H_2SO_4$  are heated to the measurement temperature (approximately 550 K) and then a valve is opened allowing the liquid  $H_2SO_4$  to reach vapor pressure equilibrium with the evacuated chamber. The presence of the vapor ( $H_2SO_4$ ,  $SO_3$ , and  $H_2O$ ) within the chamber is then measured directly using the high accuracy vacuum gauge. Next, after the system has been allowed to cool, the volume of the remaining acid is then measured. Given the density of the liquid sulfuric acid,  $\rho(gm/md)$ , it is possible to compute the number of  $H_2SO_4$  molecules which were converted to vapor phase by the relation

$$n_{vapor} = V_{1iquid} \rho/98$$
 (1)



Figure 2: Laboratory apparatus used to measure the dissociation factor, D, for gaseous  $\rm H_2SO_4$ 

where  $n_{vapor}$  is the number of vaporized molecules (in moles) and  $V_{1iquid}$  is the volume of the liquid (in ml) which vaporized during the experiment. The total number of molecules in the chamber which resulted from these vaporized molecules can be determined assuming a dissociation factor D:

$$n_{total} = n_{vapor} (1 + D)$$
(2)

where D is the portion of  $H_2SO_4$  molecules which dissociate to form  $SO_3$  and  $H_2O$ . (The range of D is between 0 and 1.) The pressure in the chamber, which can be directly measured, is related to  $n_{total}$  by the ideal gas equation:

$$PV_{chamber} = n_{total} RT$$
(3)

where P is the measured pressure in the chamber,  $V_{chamber}$  is the volume of the chamber, R is the ideal gas constant, and T is the temperature of the chamber, which is carefully monitored using a thermocouple. It is, therefore, a straightforward process to solve for the dissociation factor:

$$D = (98 PV_{chamber} / \rho RTV_{liquid}) - 1 \qquad (4)$$

Since the dissociation constant may vary with temperature, we will attempt to make this measurement at as many temperatures as possible. Once the values for the dissociation constant are known, we will correct, as necessary, expressions for microwave absorption and  $H_2SO_4$  vapor pressure, which have been derived from our previous laboratory measurements. These results will likewise be critical for the proper interpretation of planned measurements of the millimeter-wave absorptivity from gaseous  $H_2SO_4$ .

At the November 1988, AAS/DPS (Division for Planetary Sciences of the American Astronomical Society) Meeting, two independent groups brought to our attention observations made of diurnal variations in the Venus millimeterwave emission spectrum. Results reported by both the Cal.-Tech. group (Pierce et al., 1988) and by the U.C.-Berkeley group (de Pater, personal communication) showed a higher brightness temperature on the night side of the planet at wavelengths near 2.6 mm. While some atmospheric opacity near 2.6 mm is due to high altitude carbon monoxide (CO), both groups reported the variation at wavelengths well separated from the J = 0 + 1 CO transition. Since the highest level of significant millimeter-wave opacity at these wavelengths appears to occur in the 45-55 km altitude range, it is possible that either diurnal variations in the main cloud bulk density or diurnal variations in the abundance of gaseous  $H_2SO_4$  are responsible for the observed emission variations. (Note that no measurements, either remote or in-situ, have suggested diurnal variations in atmospheric or surface temperatures.) Since the 13 cm opacity is mainly due to the abundance of gaseous  $H_2SO_4$  with little relation to cloud bulk density (see Steffes, 1985), we hope to be able to search for diurnal variations in the 13 cm opacity measured from Pioneer-Venus radio occultation studies. If no variations are detected, then the 2.6 mm variations will be attributable to the clouds. Since the millimeterwave opacity from such clouds is not well understood, laboratory measurements of the millimeter-wave properties of aqueous  $H_2SO_4$  will be necessary. If such 13 cm variations are detected, we will attempt to correlate them with the variations in 2.6 mm emission. Further study and laboratory measurement of the 2.6 mm absorption from gaseous  $H_2SO_4$  will then be necessary.

Measurement of the millimeter-wave absorption from gaseous  $H_2SO_4$  will be conducted using the same general system used for measurement of the 3.2 mm absorption from NH<sub>3</sub> (Section IV, Figure 4), except that the Fabry-Perot resonator would be tuned to 110 GHz (2.7 mm wavelength) and it would be placed in a high-temperature chamber (up to 600 K). The source of the gaseous  $H_2SO_4$ would be a flask of liquid sulfuric acid. Gaseous  $CO_2$  would then be added to simulate Venus conditions. Measurements of the millimeter-wave opacity from liquid  $H_2SO_4$  would be conducted by placing a sealed sample of liquid  $H_2SO_4$ within the Fabry-Perot resonator and measuring its effects on the resonator response.

# C. Facilities

The School of Electrical Engineering of the Georgia Institute of Technology has extensive physical facilities devoted to a wide variety of research and development problems. The large faculty and staff conduct teaching, research, and applied research in almost every area of electrical engineering including microwave and millimeter-wave propagation. In addition to the school itself, the facilities of the Georgia Tech Research Institute (GTRI), a world-renown organization in the area of microwave/millimeter-wave propagation and systems, are also available. Finally, the facilities of the Atmospheric Sciences Program of the School of Geophysical Sciences at Georgia Tech are also available in this area. Overall, the ability to perform the proposed research at the Georgia Institute of Technology is excellent.

The specific measurements described in this proposal will be conducted at the Radio Astronomy and Propagation (R.A.P.) Laboratory which is located within the School of Electrical Engineering. A complete description of the

equipment being used for these measurements is included in the accompanying Semiannual Status Report for Grant NAGW-533.

For support of any required data analysis and computing activities, a wide range of computing services for education, research, and administration is provided by the Georgia Tech Office of Computing Services. Since 1955 this centralized service facility has operated a variety of computer systems. In 1983, a Control Data Corporation CYBER 180/855 was installed; in 1984, a second of these systems began operation. These two systems share disk storage and use the NOS operating system. Early in 1985, an IBM 4381 system was installed running MVS and VM. The hardware configuration, a broad variety of programming languages, applications programs, and library subroutines, all combine to provide an impressive amount of computer power to both timesharing, remote batch, and on-site batch users. Time-sharing terminals, CALCOMP and Versatec plotters, and an optical scanner offer additional Other computers available within the School of Electrical versatility. Engineering include a Data General Eclipse/S-250 and five Digital Equipment Corporation VAX systems, in addition to numerous personal computers.

# D. Proposed Procedure and Level of Effort

The proposed program will continue an ambitious effort to resolve many of the uncertainties involved in the interpretation of microwave and millimeterwave absorptivity data from Venus, Jupiter, Saturn, Uranus, and Neptune. Completion of all of the activities described (both laboratory measurements and all interpretive studies) will require approximately 36 months (November 1, 1989 through October 31, 1992). Several of these activities will be conducted simultaneously over the next 12 month period (Movember 1, 1989

through October 31, 1990). Early in the 12 month period, we expect to complete the generalized formulation for the microwave and millimeter-wave opacity of gaseous  $NH_3$  under Jovian conditions. Subsequently, we will complete development of the microwave and millimeter-wave radiative transfer models for the Jovian planets. Simultaneously, we will perform laboratory measurements of the "dissociation factor" for gaseous  $H_2SO_4$ , and use the results to correct existing expressions for the microwave opacity of the Venus atmosphere as measured at different times and different locations in the Venus atmosphere. Finally, we hope to have assembled the equipment so as to begin measurements of the <u>millimeter-wave</u> opacity from gaseous and liquid  $H_2SO_4$ .

The proposed level of effort in the 12 month period proposed for FY90 (November 1, 1989 through October 31, 1990) involves one professor (P. G. Steffes, Associate Professor of Electrical Engineering) at 25% time, one graduate student (Graduate Research Assistant, Joanna Joiner) at 50% time, and a second graduate student assistant at 33% time, with supplies and other support as indicated in the attached cost breakdown (see Section F). In addition to the participation in the program by Professor Steffes and the paid graduate research assistants, contributions to the program from both graduate and undergraduate students working on special projects for academic credit have been substantial. Topics of these projects have included both laboratory measurements and the application and analysis of microwave refractivity and absorptivity data from simulated planetary atmospheres, in addition to repair of laboratory microwave equipment supporting the project. We expect such contributions to continue in the future. Likewise, in the spirit of the NASA Graduate Student Researchers Program (Underrepresented Minority Focus), we continue to seek out talented underrepresented minority students and involve them in our program.

### E. References for Proposal

Ayers, R. P., R. W. Gillet, and J. L. Gras (1980). On the vapor pressure of sulfuric acid. Geophysical Research Letters 7, 433-436.

Bézard, B., A. Marten, J. P. Baluteau, D. Gautier, J. M. Flaud, and C. Camy-Peyret (1983). On the detectability of H<sub>2</sub>S in Jupiter. <u>Icarus 55</u>, 259-271.

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Gmitro, J. I. and T. Vermeulen (1964). Vapor-liquid equilibria for aqueous sulfuric acid. Amer. Inst. Chem. Eng. J. 10, 740-746.

Joiner, J., P. G. Steffes, and J. M. Jenkins (1989). Laboratory measurements of the 7.5-9.38 mm absorption of gaseous ammonia  $(NH_3)$  under simulated Jovian conditions. Icarus, in press.

Pierce, K., D. O. Muhleman, and G. Berge (1988). Spatial resolution of the CO J = 0 + 1 line on Venus. <u>Bull. Amer. Astron. Soc.</u> 20, 832.

Steffes, P. G. (1985). Laboratory measurements of the microwave opacity and vapor pressure of sulfuric acid under simulated conditions for the middle atmosphere of Venus. <u>Icarus 64</u>, 576-585.

Steffes, P. G. (1986). Evaluation of the microwave spectrum of Venus in the 1.2 to 22 centimeter wavelength range based on laboratory measurements of constituent gas opacities. <u>Astrophysical Journal</u> 310, 482-489, November 1, 1986.

Steffes, P. G. and J. M. Jenkins (1987). Laboratory measurements of the microwave opacity of gaseous ammonia  $(NH_3)$  under simulated conditions for the Jovian atmosphere. Icarus 72, 35-47.

Steffes, P. G., M. J. Klein, and J. M. Jenkins (1989). Observation of the microwave emission of Venus from 1.3 to 3.6 cm. Submitted to Icarus.

### F. Projected Budget

through June 30.

For the period of November 1, 1989 through October 31, 1990

# Estimated Cost Breakdown

1.	DIRECT SALARIES AND WAGES*:	\$37,220
	A. Principal Investigator P.G. Steffes	
	25% time, calendar year \$15,620	
	B. 1 Graduate Student	
	50% time, calendar year \$12,960	
	C. 1 Graduate Student	
	$337$ time, calendar year $\frac{58,640}{58,640}$	
II.	FRINGE BENEFITS**:	\$ 3,984
	(less students)	
III.	MATERIALS, SUPPLIES, AND SERVICES:	<b>\$ 1,3</b> 00
	A. Gases and Liquids for Experiments - \$ 800	
	B. Miscellaneous Project Supplies and Components \$ 500	
IV.	TRAVEL:	\$ 1,800
	A. Travel for Students (2) to AAS/DPS Meeting (Providence, RI, 4 days duration) <u>\$1,800</u>	
	SUBTOTAL - ESTIMATE OF DIRECT COSTS:	\$44,304
v.	OVERHEAD (Indirect Expense)**: 60% of Modified Total Direct Cost Base	\$26,583
	TOTAL ESTIMATED COST:	\$70,887

\*The salary and wage rates are based on current FY89 salaries for the Georgia Institute of Technology. An increase of 10% is estimated for the performance in Georgia Tech FY90. The Georgia Tech Fiscal Year is July I

\*\*Rates are effective for the period July 1, 1988 through June 30, 1989 and are subject to adjustment thereafter upon DCAA audit and ONR negotiations.

# G. Cognizant Personnel

For scientific or technical matters relating to the contract:

Paul G. Steffes School of Electrical Engineering Georgia Institute of Technology Atlanta, Georgia 30332-0250 Telephone: (404) 894-3128

For contractual and business matters:

Georgia Tech Research Corporation Centennial Research Building Georgia Institute of Technology Atlanta, Georgia 30332-0420 Telephone: (404) 894-4817

### H. Biographical Sketch

STEFFES, PAUL G. - Associate Professor of Electrical Engineering Georgia Institute of Technology

### EDUCATION

S.B., S.M., Massachusetts Institute of Te	echnology 1977
Electrical Engineering	
Ph.D., Stanford University, Electrical En	ngineering 1982

### EMPLOYMENT HISTORY

Georgia Institute of Technology, School of Electrical 1982-Present Engineering, Assistant Professor

Duties include both research and teaching.

Research Activities: Principal Investigator--NASA Planetary Atmospheres Program, "Laboratory Evaluation and Application of Microwave Absorption Properties under Simulated Conditions for Planetary Atmospheres." This research includes study of the interaction between a number of atmospheric constituents and electromagnetic waves, along with applications of these studies to spacecraft measurements of the microwave absorption in atmospheres of Venus and the outer planets (1984-1989). Principal Investigator--NASA Pioneer Venus Guest Investigator Program, "Pioneer Venus Radio Occultation (ORO) Data Reduction: Profiles of 13 cm Absorptivity." This research infers 13 cm wavelength absorptivity profiles using the Pioneer Venus Orbiter, and then uses such profiles to characterize abundance profiles for gaseous  $H_2SO_4$  in the Venus atmosphere. Principal Investigator--National Science Foundation Grant, "Remote Sensing of Clouds Bearing Acid Rain." This research studied and designed a microwave/millimeter-wave system for remotely sensing the pH of acidic clouds (1982-1983). Principal Investigator--GTE Spacenet Program: "Satellite Interference Locating System (SILS)." The program involves location of interfering signals on the surface of the earth (geodesy) without disrupting regular satellite operations (1986-1989). Principal Investigator--Emory University-Georgia Tech Biomedical Technology Research Center, "Research in Development of a Non-Invasive Blood Glucose Monitoring Technique." This research involves the use of active infrared systems to determine glucose levels in the human eye and bloodstream. Co-director--Ku Band Satellite Earth Station System. "Responsible for development of a Ku-band uplink/ downlink system for use in inter-university networks. Co-investigator--"Radar Warning Receiver Evaluations" with Georgia Tech Research Institute (GTRI).

Teaching Effort: Resource Professor for Satellite Communications Systems (graduate course) and <u>Electromagnetics III</u> (undergraduate required course covering waves, waveguides, and antennas). Have also taught Electromagnetics II (electrodynamics), Signals and Systems, and Survey of Remote Sensing.

#### PAST ACADEMIC RESEARCH

Stanford University Electronics Lab

1979-1982

1977-1982

- Graduate Research Assistant, Center for Radar Astronomy Supervisor: Prof. Von R. Eshleman

Research was concentrated in the area of microwave radio occultation experiments from Voyager and Mariner spacecraft, with specific interest in microwave absorption in planetary atmospheres. Work included computer-based theoretical development of microwave absorption coefficients for planetary atmospheres, to facilitate the use of radio occultation-derived microwave absorption profiles in determining constituent densities. Additional work included the development of a fully instrumented experimental facility for use in measuring the microwave properties of planetary atmospheres under simulated planetary The research resulted in a Ph.D. dissertation entitled conditions. "Abundances of Cloud-Related Gases in the Venus Atmosphere as Inferred from Observed Radio Opacity."

Massachusetts Institute of Technology1976-1977Graduate Research Assistant, Research Laboratory of<br/>Electronics (Radio Astronomy and Remote Sensing Group)976-1977Supervisor:Prof. David H. Staelin-

Responsible for development, operation, and data analysis for an 8-channel, 118 GHz radiometer system flown aboard the NASA Flying Laboratory (CV-990) as an engineering model for a meteorological sensing satellite. Duties included hardware development of millimeter-wave, microwave, analog, and A to D segments of the system, in addition to airborne operation and reduction of data. The research resulted in a Master's thesis entitled "Atmospheric Absorption at 118 GHz" detailing the first airborne measurement of high altitude atmospheric absorption in the 2.5 millimeter wavelength range, due to atmospheric oxygen.

#### INDUSTRIAL EXPERIENCE

Watkins-Johnson Company, San Jose, California Member of the Technical Staff of the Sensor Development Section of the Recon Division

"Responsibilities included customer proposals and system design and development, particularly in the area of millimeter-wave systems. Responsibility for millimeter-wave systems development included government sponsored study and development of ELINT (Electronic Intelligence) and radar warning receiving systems to frequencies as high as 110 GHz, as well as internal company-sponsored development projects including a 60 GHz communications system, and millimeter-wave downconverters.

### AWARDS AND PROFESSIONAL AFFILIATIONS

Member of the American Association for the Advancement of Science, American Astronomical Society (Division for Planetary Sciences), the American Geophysical Union, American Institute of Physics, American Society for Engineering Education, Association of Old Crows, Astronomical Society of • the Pacific, Eta Kappa Nu, IEEE, Sigma Xi, and U.R.S.I. Received the Stewart Award (M.I.T.), for exceptional contribution to student extracurricular life, in 1977. Associate editor, Journal of Geophysical Research (JGR-Atmospheres), 1984-89. Metro Atlanta "Young Engineer of the Year," presented by Society of Professional Engineers, 1985. Sigma Xi Young Faculty Research Award, 1988. Reviewer/referee for Icarus (International Journal of Solar System Studies), Journal of Geophysical Research, Radioscience, and for several textbooks in the area of electromagnetics. Proposal reviewer for the NASA Planetary Astronomy Program and the NASA Planetary Atmospheres Program. Appointed member of the NASA Management and Operations Working Group for the Planetary Atmospheres Program. Officer of the Atlanta Chapter, IEEE Antennas and Propagation Society and Microwave Theory and Techniques Society (Chairman, 1986-88). Director of Atlanta Section, IEEE (1988-1989).

#### PUBLICATIONS

- 1. Steffes, P. G., "A Microwave (UHF) Television Repeater System," B.S. Thesis, Massachusetts Institute of Technology, 1976.
- 2. Steffes, P. G., "Atmospheric Absorption at 118 GHz," M.S. Thesis, Massachusetts Institute of Technology, 1977.
- 3. Steffes, P. G., "Sulfur Dioxide and Other Cloud-Related Gases as Microwave Absorbers in the Middle Atmosphere of Venus," <u>Bull. Amer. Astron.</u> Soc., 12, 719, 1980.
- 4. Steffes, P. G. and Meck, R. A., "Prototype Tests Secure Millimeter Communications," <u>Microwave Systems News</u>, 10, 59-68, October 1980.
- 5. Eshleman, V. R., Muhleman, D. O., Nicholson, P. D., and Steffes, P. G., "Comment on Absorbing Regions in the Atmosphere of Venus as Measured by Radio Occultation," <u>Icarus</u>, <u>44</u>, 793-803, December 1980.
- 6. Steffes, P. G. and Eshleman, V. R., "Sulfur Dioxide and Other Cloud-Related Gases as the Source of the Microwave Opacity of the Middle Atmosphere of Venus," Icarus, 46, 127-131, April 1981.
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- Steffes, P. G. and Eshleman, V. R., "Laboratory Measurements of the Microwave Opacity of Cloud Related Gases under Simulated Conditions for the Venus Atmosphere," Bull. Amer. Astron. Soc., 13, 716, 1981.

- 9. Steffes, P. G. and Eshleman, V. R., "Abundances of Cloud-Related Gases in the Venus Atmosphere: Implications from Observed Radio Opacity," <u>Proceedings of the International Conference on the Venus Environment</u>, vol. 1, p. 20, Palo Alto, CA, November 1981.
- Steffes, P. G. and Eshleman, V. R., "Sulfuric Acid Vapor and Other Cloud- Related Gases in the Venus Atmosphere: Abundances Inferred from Observed Radio Opacity," <u>Abstracts of the Fourth Annual Meeting of Planetary</u> <u>Atmospheres Principal Investigators</u>, vol. 1, pp. 20-21, University of Michigan, April 21-23, 1982.
- Steffes, P. G. and Eshleman, V. R., "Sulfuric Acid Vapor and Other Cloud-Related Gases in the Venus Atmosphere: Abundances Inferred from Observed Radio Opacity," <u>Icarus</u>, 51, 322-333, August 1982.
- Steffes, P. G., "Abundances of Cloud-Related Gases in the Venus Atmosphere as Inferred from Observed Radio Opacity," Ph.D. Dissertation, Stanford University, 1982.
- 13. Steffes, P. G., "Millimeter-Wavelength Remote Sensing of Stratospheric Sulfur Dioxide," EOS, 64, 198-199, May 1983.
- 14. Steffes, P. G., "Microwave Remote Sensing of Gases and Clouds Involved in the Formation of Acid Precipitation," <u>Digest of 1983 International</u> <u>Geoscience and Remote Sensing Symposium</u> (IGARSS '83), v. 2, FA-4, pp. 3.1-3.4, San Francisco, CA, 1983.
- 15. Steffes, P. G., "A Millimeter-Wave System for the Remote Sensing of Acidic Clouds and Precursor Gases in the Troposphere," <u>Digest of the</u> <u>Eighth International Conference on Infrared and Millimeter Waves</u>, vol. 1, pp. 264-265, Miami Beach, FL, December 16, 1983.
- 16. Steffes, P. G., Stellitano, P. S., and Lott, R. C., "Measurements of the Microwave Opacity and Vapor Pressure of Gaseous Sulfuric Acid under Simulated Venus Conditions," <u>Bull. Amer. Astron. Soc.</u>, <u>16</u>, 694, 1984. Presented at the 16th Annual Meeting of the American Astronomical Society, Kona, HA, October 1984.
- 17. Steffes, P. G., "Laboratory Measurements of Microwave Absorption from Gaseous Constituents under Conditions for the Outer Planets," <u>The Jovian</u> <u>Atmospheres</u> (NASA Conference Publication 2441), 111-116, given at the Conference on the Jovian Atmospheres, New York, May 1985.
- 18. Steffes, P. G., "Microwave Absorption from Cloud-Related Gases in Planetary Atmospheres," <u>Proceedings of the 1985 Joint Assembly of the Inter-</u> national Association of Meteorology and Atmospheric Physics (IAMAP) and the International Association of Physical Sciences of the Ocean (IAPSO), M10-13, p. 96, August 1985.

- 19. Steffes, P. G. and Watson, D. H., "Constraints on Constituent Abundances in the Venus Atmosphere from the Microwave Emission Spectrum in the 1 to 20 cm Wavelength Range," <u>Bull. Amer. Astron. Soc.</u>, <u>17</u>, p. 720. Presented at the 17th Annual Meeting of the Division of Planetary Sciences of the American Astronomical Society, Baltimore, MD, October 1985.
- 20. Steffes, P. G., "Laboratory Measurements of the Microwave Opacity and
  Vapor Pressure of Sulfuric Acid under Simulated Conditions for the Middle Atmosphere of Venus," Icarus, 64, 576-585, December 1985.
- 21. Steffes, P. G., "Evaluation of the Microwave Spectrum of Venus in the 1.2 to 22 cm Wavelength Range Based on Laboratory Measurements of Constituent Gas Opacities," <u>Astrophysical Journal, 310</u>, 482-489, November 1, 1986.
- 22. Steffes, P. G., "Microwave Properties of the Atmospheres of the Outer Planets: Laboratory Measurements with the Georgia Tech Planetary Atmospheres Simulator," Proceedings of the Workshop Laboratory Measurements for Planetary Science, p. L-6. Presented in Meudon, France, November 3, 1986.
- 23. Steffes, P. G., Jenkins, J. M., Selman, M. F., and Gregory, W. W., "Laboratory Measurements of the Microwave Opacity of Gaseous Ammonia (NH<sub>3</sub>) under Simulated Conditions for Jovian Atmospheres," <u>Bull. Amer.</u> <u>Astron. Soc. 18</u>, 787. Presented at the 18th Annual Meeting of the Division of Planetary Sciences, AAS, Paris, France, November 5, 1986.
- 24. Steffes, P. G., and Jenkins, J. M., "Laboratory Measurements of the Microwave Opacity of Gaseous Ammonia (NH<sub>3</sub>) under Simulated Conditions for the Jovian Atmosphere," Icarus 52, 35-47, October 1987.
- 25. Smith, W. W. and Steffes, P. G., "Time Delay Techniques for a Satellite Interference Location System," <u>IEEE Transactions on Aerospace and</u> <u>Electronic Systems 25</u>, 401-408, July 1989.
- 26. Jenkins, J. M., and Steffes, P. G., "Limits of the Microwave Absorption of H<sub>2</sub>O and CH<sub>4</sub> in the Jovian Atmosphere," <u>Bull. Amer. Astron. Soc. 19</u>, p. 695. Presented at the 19th Annual Meeting of the Division of Planetary Sciences of the American Astronomical Society, Pasadena, California, November 11, 1987.
- 27. Joiner, J., Jenkins, J. M., and Steffes, P. G., "Laboratory Measurements of the Opacity of Gaseous Ammonia (NH<sub>3</sub>) in the 7.3-8.3 mm (36-41 GHz) Range under Simulated Conditions for Jovian Atmospheres," <u>Bull. Amer. Astron. Soc. 19</u>, p. 694. Presented at the 19th Annual Meeting of the Division of Planetary Sciences of the American Astronomical Society, Pasadena, California, November 11, 1987.
- 28. Steffes, P. G., Jenkins, J. M., and Klein, M. J., "Observation of the Microwave Emission Spectrum of Venus from 1.3 to 3.6 cm," <u>Bull. Amer.</u> <u>Astron. Soc. 19</u>, p. 780. Presented at the 19th Annual Meeting of the Division of Planetary Sciences of the American Astronomical Society, Pasadena, California, November 12, 1987.

- 29. Steffes, P. G., "Laboratory Measurements of the Millimeter-Wave Absorption from Gaseous Constituents under Simulated Conditions for the Outer Planets," Proceedings of the Workshop, Laboratory Measurements for Planetary Science II, pp. 6-7 through 6-8. Presented in Pasadena, California, November 9, 1987.
- 30.- Jenkins, J. M. and Steffes, P. G., "Constraints on the Microwave Opacity of Gaseous Methane and Water Vapor in the Jovian Atmosphere," <u>Icarus 76</u>, 378-382, November 1988.
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MARCH 1989

### A. The Georgia Tech Radio Astronomy and Propagation (R.A.P.) Facility

The basic configuration of the planetary atmospheres simulator developed at Georgia Tech for use in measurement of the microwave absorptivity of gases under simulated conditions for planetary atmospheres is described at length in the previous Annual Status Report(s) for Grant NAGW-533. It is also discussed at length in Steffes (1986) and Steffes and Jenkins (1987). The most recent additions to the Georgia Tech Radio Astronomy and Propagation Facility have been Fabry-Perot type resonators capable of operation between 30 and 41 GHz and between 93 and 95 GHz. As shown in Figure 1, the Ka-band resonator (32-40 GHz) consists of two gold plated mirrors (one with a flat surface, and one with a parabolic surface) separated by a distance of about 20 cm. The mirrors are contained in a T-shaped glass pipe which serves as a pressure vessel capable of withstanding over 2 atm of pressure. Each of the three open ends of the pipe is sealed with an O-ring sandwiched between the lip of the glass and a flat brass plate which is bolted to an inner flange. Electromagnetic energy is coupled both to and from the resonator (which operates as a bandpass filter) by twin irises located on the surface of the flat mirror. Two sections of WR-28 waveguide which are attached to the irises pass through the brass plate to the exterior of the pressure vessel. The end of each waveguide section is pressure-sealed by a rectangular piece of mica which is held in place by a mixture of rosin and beeswax. As shown in Figure 2, one of these ends is connected to the sweep oscillator through a waveguide section. A Ka-band (26-40 GHz) mixer is attached to the other section of waveguide and is coupled to the high resolution spectrum analyzer through a calibrated section of coaxial cable. The entire resonator,

including its glass pressure envelope, is placed in the temperature chamber, which is a low-temperature freezer capable of operation down to 150 K. A network of stainless steel tubing and valves connects other components such as gas storage tanks, vacuum gauges, the pressure gauge, and the vacuum pump to the resonator assembly, so that each component may be isolated from the system as necessary. When properly secured, the system is capable of containing up to two atmospheres of pressure without detectable leakage.

In order to achieve a better system sensitivity, which corresponds to a higher "Q" or quality factor for the Fabry-Perot resonator, all losses in the resonator must be minimized, since the quality factor is defined as  $2\pi$  times the ratio of the average energy stored in the resonator to the energy lost (per cycle) in the resonator. There are three sources of loss which typically affect a Fabry-Perot resonator (Collin, 1966):

- (1) Resistive losses on the surfaces of the mirrors.
- (2) Coupling losses due to the energy coupling out of the resonator through the irises on the flat-surfaced mirror.
- (3) Diffraction losses around the sides of the mirror.

For previous measurements made at frequencies below 22 GHz (wavelengths longer than 1.35 cm), it was found that the resistive losses were predominant. This was because the measurements were conducted using <u>cylindrical</u> resonators, for which no diffraction losses existed. (See Steffes and Jenkins, 1987). Also, coupling losses were held to a minimum by using very small coupling loops and irises. The predominance of the resistive losses in the cylindrical resonators was demonstrated when such resonators were cooled to 193 K in the atmospheric simulator. Significant improvement in the quality factor of the resonators was observed when compared with their room temperature values.
This was consistent with the expected reduction in the resistive losses at lower temperatures.

When the newer, higher frequency (32 to 40 GHz) Fabry-Perot resonator (Figure 1) was first cooled from room temperature down to 203 K for tests under simulated Jovian conditions, its quality factor appeared to worsen Initially, it was thought that this might have been rather than improve. caused by separation of the gold plating on the mirror surfaces from the backstructure (which had been machined from aluminum) due to differential thermal contraction. As a result, new mirrors were machined (to high tolerance) from brass, and then were plated with titanium and then gold, to assure no separation would occur. The performance of the new mirrors was only marginally better when installed in the resonator. Computation of the resistive losses from the mirrors showed that, in the absence of all other losses, the Q of our Fabry-Perot resonator should be on the order of 250,000; whereas its actual Q was on the order of 10,000. Therefore, it became clear that either coupling losses or diffraction losses were the limiting factor in its performance, and that even the introduction of high-temperature superconducting material would not significantly improve the sensitivity of the system. (Note, however, that we are still studying the possibility of using high temperature superconductors in our lower frequency, cylindrical resonators in order to obtain increased sensitivity at frequencies below 22 GHz.)

In order to further improve the quality factor of the 32 to 40 GHz system, some additional improvements were made. First, adjustable irises were developed so that the smallest possible coupling losses would occur, while still allowing sufficient signal coupling in and out of the resonator so as to make accurate absorptivity measurements. Since the irises are actually

circular holes which are placed near the center of the flat-surfaced mirror, adjustment of their sizes is difficult. However, two small metal sheets with V-shaped cuts were placed immediately behind each iris in an area where the mirror surface thickness is very small. The two sheets could be moved together or apart so as to adjust the effective size, and therefore the coupling, of each iris. Even when adjusted for minimal coupling, however, the resonator Q was only slightly improved, suggesting that diffraction losses were the major limiting factor to system sensitivity.

Diffraction losses occur due to energy being lost around the edges of the mirrors. These losses can be minimized by assuring that both mirrors are oriented directly toward each other (i.e., the centerlines for each mirror, which are orthogonal to the planes of each mirror at their centerpoints, must be colinear). Since the positioning of the mirrors can vary with temperature, due to thermal contraction or expansion of metallic mounting structures, the temperature dependence of the quality factor which has been observed is consistent with a system which is limited by diffraction losses.

Two approaches can be used to minimize diffraction losses. The first involves precise pointing of the mirrors. This was accomplished by directing the beam of a helium-neon laser through the input waveguide and iris and into the resonator. Mirror positioner screws could then be adjusted so that the reflected beam focused precisely on the output iris. Since the parabolic mirror has a precisely defined focus, adjustment of its exact position is far more critical than that of the flat mirror. The second technique for reducing diffraction loss involves the use of larger mirrors in the resonator. However, because of size limitations set by our pressure vessel, we are unable to significantly increase the size of the mirrors in our system.

Overall, our efforts at improving the quality factor of our 32 to 40 Ghz Fabry-Perot resonator were successful. Since absorptivity is measured by monitoring the <u>change</u> in the quality factor of the resonator which is caused by the absorbing gas mixture, improvements in our measurement technique have further improved the system sensitivity.

At the end of the previous grant year (February 1, 1988 through January 31, 1989), we implemented a similar system for operation around 94 GHz (3.2 mm). As shown in Figure 3, the 94 GHz resonator resembles the Ka-band Fabry-Perot resonator in that it consists of two gold plated mirrors, one with a flat surface and one with a parabolic surface. However, the 94 GHz resonator is significantly different in that the flat mirror has a much smaller radius than the parabolic mirror, and the focal length of the parabolic mirror is much shorter, resulting in "tighter" focusing. Mechanically, the 94 GHz resonator is superior in that the parabolic mirror rests on two support arms and can be adjusted in distance from the flat mirror without disturbing the other angular adjustments. The result has been a resonator with a guality factor (Q) of about 30,000.

The 94 GHz system (Figure 4) functions in the same fashion as does the Ka-band system in that changes in the Q of the Fabry-Perot resonator are monitored as lossy gas mixtures are introduced into the pressure vessel. However, because of the extremely high frequency of operation, costly millimeter-wave components (e.g., signal sources, waveguides, and mixers) are required. Fortunately, such a resource of millimeter-wave equipment exists at Georgia Tech within the Georgia Tech Research Institute (GTRI) and has been made available to us for these experiments. Figure 4, shows a W-band (75-110 GHz) sweep oscillator whose signal is fed into the Fabry-Perot resonator via

an isolator, so as to provide a constant impedance both for the backward wave oscillator (BWO) tube in the sweeper and for the resonator. The resonator output at 94 GHz is then down-converted to around 1.55 GHz using a harmonic mixer which employs the tenth harmonic of a stabilized 9.24 GHz oscillator. The 1.5 GHz signal is then fed to the high-resolution spectrum analyzer by which the bandwidth (and, therefore, the Q) of the resonance can be measured. The pressurization systems for both systems are essentially identical.

### B. Experimental Approach

The approach used to measure the microwave absorptivity of test gases in an  $H_2/He$  atmosphere is similar to that used previously by Steffes and Jenkins (1987) for simulated Jovian atmospheres. At frequencies between 32 and 40 GHz, the changes in the Q of the numerous resonances of the Fabry-Perot resonator (see Figure 2) are related to the absorptivity of the test gas mixture at these frequencies. Similarly, the changes in the Q of the single 94 GHz resonance are related to the absorptivity at that frequency. The changes in the Q of the resonances which are induced by the introduction of an absorbing gas mixture can be monitored by the high resolution microwave spectrum analyzer, since Q is simply the ratio of the cavity resonant frequency to its half-power bandwidth. For relatively low-loss gas mixtures, the relation between the absorptivity of the gas mixture and its effect on the Q of a resonance is straightforward:

$$\alpha \simeq (Q_{\rm L}^{-1} - Q_{\rm C}^{-1}) \pi / \lambda \tag{1}$$

where  $\alpha$  is absorptivity of the gas mixture in Nepers km<sup>-1</sup>. (Note, for example, that an attenuation constant or absorption coefficient or absorptivity of 1 Neper km<sup>-1</sup> = 2 optical depths per km (or km<sup>-1</sup>) = 8.686 dB km<sup>-1</sup>, where the first notation is the natural form used in electrical engineering, the second is the usual form in physics and astronomy, and the third is the common (logarithmic) form. The third form is often used in order to avoid a possible factor-of-two ambiguity in meaning.) Q<sub>L</sub> is the quality factor of the cavity resonator when the gas mixture is present, Q<sub>C</sub> is the quality factor of the resonance in a vacuum, and  $\lambda$  is the wavelength (in km) of the test signal in the gas mixture.

In the previous grant year, we made high accuracy measurements of the 7.5 to 9.3 mm absorption from gaseous ammonia  $(NH_3)$  in a 90%  $H_2/10$ % He atmosphere. at a temperature of 203 K. While even lower temperatures could be achieved, the need to avoid the risk of ammonia condensation kept our operating temperatures relatively high. A complete description of these measurements is provided in Joiner <u>et al.</u> (1989) and in the <u>Annual Status Report</u> for the previous grant year.

It is noteworthy that with our technique, once a test gas mixture is formed in the pressure vessel, the same mixture is used for measurement of absorption at several frequencies and pressures. Thus, even though some uncertainty in absolute mixing ratio exists, the pressure and frequency <u>dependences</u> of the millimeter-wave absorption can be measured to high accuracy. However, in order to properly characterize the magnitude of the absorption, the ammonia mixing ratio must be known precisely. Using the high accuracy thermocouple vacuum gauge shown in Figure 2, the actual NH<sub>3</sub> mixing ratio can only be determined to an accuracy of  $\pm 20\%$  of its value. (Note:

This corresponds to  $(1.85 \pm 0.37)$ % NH<sub>3</sub> volume mixing ratio.) However, since our required overall accuracy for the NH<sub>3</sub> absorptivity measurement is  $\pm 20$ %, this mixing ratio uncertainty is excessive. In order to reduce this uncertainty, we arranged for a local gas products supplier (Matheson Gas Products) to provide us with a pre-mixed hydrogen/helium/ammonia atmosphere which was analyzed with a mass spectrometer so that mixing ratio accuracies of better than 2% (i.e., (1.85  $\pm$  .04)%) were obtained. We have used this mixture (1.85% NH<sub>3</sub>, 9.81% He, and 88.34% H<sub>2</sub>) for the high accuracy absorptivity measurements which are required to accurately infer ammonia abundance from millimeter-wave opacity data for the Jovian planets.

This same "custom" gas mixture has been measured under the same conditions (total pressures of 1 and 2 Bars and temperature 203 K), using the same techniques, at a single resonance near 94 GHz (3.2 mm). However, because the absorptivity of ammonia is less at 3.2 mm than in the 7.5-9.3 mm wavelength range, the percentage accuracy of the measurements have been worse, even though the quality factor of the 3.2 mm resonator is higher. As a result, a second "custom" gas mixture has been obtained with an even higher ammonia mixing ratio (5.07% NH<sub>3</sub>. 85.56% H<sub>2</sub>, 9.37% He). Because of the higher ammonia mixing ratio (and the need to avoid condensation), measurements of the 94 GHz absorptivity of this new mixture have been conducted at a slightly higher temperature (210 K) for total pressures of 1, 1.3, 1.7, and 2 Bars.

# C. Results of Laboratory Measurements and Their Application

Results of measurements of the 32 to 40 GHz (7.5 to 9.3 mm) absorptivity of gaseous ammonia under simulated Jovian conditions (203 K) are shown in Figure 5. These measurements were made using a 88.3% hydrogen/9.81% helium

atmosphere with a total pressure of 2 Bars. The ammonia mixing ratio was 0.0185. Triangular points represent measurements of gas mixtures formed using the thermocouple vacuum gauge (NH<sub>3</sub> mixing ratio accuracy =  $\pm 20\%$  of its value) and the circular points represent measurements of the pre-mixed, analyzed gas mixture described in Section B. With this mixing ratio, temperatures as low as 190 K could have been used before saturation would have become a problem, but 203 K was used so as to be consistent with earlier measurements.

Also, shown in Figure 5 are solid lines which represent the theoretically-computed opacity using the Van Vleck-Weisskopf lineshape (upper line), the modified Ben-Reuven lineshape (middle line), and the Gross lineshape (lower line). The Van Vleck-Weisskopf calculation was performed using linewidths and line intensities as per Wrixon et al. (1971). The Ben-Reuven calculation was made as per Berge and Gulkis (1976), by employing a Ben-Reuven lineshape which has been modified so as to be consistent with the laboratory results of Morris and Parsons (1970), in which the 9.58 GHz absorption from  $NH_3$  (in a high pressure  $H_2/He$  atmosphere) at room temperature was measured. The Gross lineshape formulation used the lineshape of Gross (1955) and linewidths and line intensities from Wrixon et al. (1971). These theoretical spectra were computed using generalized computer programs for which the partial pressures from  $H_2$ , He, and  $NH_3$ , as well as frequency and temperature, were adjustable variables. The values picked for these variables matched our experimental conditions.

Inspection of the results in Figure 5 shows that most of the measured data points lie nearest to the theoretically-derived absorptivity expression based on the Gross lineshape. The Gross lineshape has also been used to describe the absorption due to atmospheric water vapor (Zhevakin and Naumov,

1963) and has been found to give better results than the Van Vleck theory. The Van Vleck-Weisskopf lineshape overstates the opacity of NH3 by nearly a factor of 2, while the modified Ben-Reuven lineshape overstates the opacity of NH3 by an average of over 40%. Such results are not surprising, however, in that the accuracy of the new measurements is far greater than that from previous measurements. It is also noteworthy that other researchers, such as de Pater and Massie (1985), have found that in order to best explain the 1-10 mm Jupiter emission spectrum, a different sort of lineshape was needed to characterize the ammonia opacity. Likewise, previous laboratory measurements of NH<sub>3</sub> opacity at frequencies below 22 GHz showed opacities slightly less than those indicated by the modified Ben-Reuven lineshape under the same conditions of temperature and pressure (Steffes and Jenkins, 1987). However, it has been noted by Spilker (private communication) that slight changes to the modified Ben-Reuven formulation can be made which make it more consistent with longer wavelength laboratory results, and make it essentially identical with the Gross formulation at frequencies between 32 and 40 GHz (see discussion below).

Our preliminary results at 94 GHz, shown in Figure 6, definitely favor the modified Ben-Reuven formalism. When the data points for opacity are compared with the theoretically-computed opacity using the Van Vleck-Weisskopf lineshape (upper line), the modified Ben-Reuven lineshape (middle line), and the Gross lineshape (lower line), it becomes clear that the modified Ben-Reuven lineshape best characterizes the opacity at 3.2 mm under the conditions of this measurement (Total Pressure: 2 Bars; Temperature: 213 K; Mixing Ratios: 5.07% NH, 85.56% H, 9.37% He). Note that these three theoretical computations were made using the same techniques used for the 7.5-9.3 mm calculation. This result is especially significant in that it contradicts the

suggestion by de Pater and Massie (1985) that ammonia opacity at wavelengths shortward of 7 mm must be substantially greater than indicated by the modified Ben-Reuven lineshape. Instead it suggests an ammonia abundance distribution different than that proposed, or the presence of other millimeter-wave absorbing constituents, such as was suggested by Bézard (1983).

Our results at 94 GHz are also significant in that they are not consistent with the Gross lineshape, and therefore, confirm the suggestion by Spilker and Eshleman (1988) that a further modification to Ben-Reuven lineshape best describes the microwave absorption spectrum of  $NH_3$  under Jovian conditions. However, it should be noted that this only applies at pressures of 2 Bars or greater. At pressures well below 2 Bars, it is expected that the absorption of  $NH_3$  will revert to that computed using the Van Vleck-Weisskopf formulation, which will be especially noticeable at frequencies far from 1-2 cm inversion resonances. Therefore, we hope to be able to characterize this transition from the modified Ben-Reuven lineshape to the Van Vleck-Weisskopf lineshape by further study of the 94 GHz opacity of  $NH_3$  (in an  $H_2/He$ atmosphere) in the 1 to 2 Bar pressure range, in the second half of the current grant year.

## D. Observational and Interpretive Studies

As described in the previous <u>Annual Status Report(s)</u> for Grant NAGW-533, and in Steffes <u>et al.</u> (1989), studies of our recent measurements of the 1.35 to 3.6 cm emission from Venus have suggested that long term temporal and/or significant spatial variations in the abundance of  $SO_2$  and gaseous  $H_2SO_4$  may occur immediately below the main cloud layer (48 km and below). Our observation, which was predominantly of equatorial and mid-latitude regions of

Venus, indicated a significantly lower  $SO_2$  abundance than was measured in 1978 by the Pioneer-Venus Sounder Probe, and a lower average abundance of gaseous  $H_2SO_4$  than would have been inferred from earlier Pioneer-Venus radio occultation studies of subcloud opacity at 13 cm. Some or all of this difference may be due to spatial variations in the subcloud  $H_2SO_4$  abundance since most of the early Pioneer-Venus results were for polar latitudes (Cimino, 1982). Similarly, our results may be consistent with the earlier equatorial 13 cm radio occultation opacity measurements made with the Mariner 10 spacecraft (Lipa and Taylor, 1979), where the peak opacity would correspond to a very large abundance of gaseous  $H_2SO_4$ , but the average subcloud opacity was significantly lower.

One important tool for evaluating these effects is the reduction of the microwave data from the 1986-87 Pioneer-Venus radio occultation measurements. This data was taken over a wide range of latitudes and could be critical for determining whether temporal variations or spatial variations in gaseous  $H_2SO_4$  abundance could be occurring. Working with Dr. Arvydas J. Kliore (P-V Radioscience Leader), our group has obtained the currently unreduced data and (working at JPL) begun reducing the data to obtain absorptivity profiles for the 1986-87 epoch, as part of the Pioneer-Venus Guest Investigator Program.

By then end of 1989, we will have reduced data obtained from the Fall 1986/ Winter 1987 Pioneer-Venus Radio Occultation Observations. After the initial reduction, we hope to have dependable 13 cm wavelength refraction and absorption profiles for a range of altitudes in the Venus atmosphere reaching down to 38 km and for latitudes ranging form equatorial to polar.

To show the usefulness of the 13 cm opacity data for inferring the nature of the gaseous  $H_2SO_4$  abundance, we compare in Figure 7 the measured absorp-

tivity profile from orbit 2801N with that absorptivity which would result from a saturation abundance of gaseous  $H_2SO_4$  (from Steffes, 1985) in the 43 to 55 km altitude range. It can be seen that for altitudes in the 49 to 51 km altitude range (the nominal altitude range of the Venus lower cloud - see Ragent and Blamont, 1980), absorptivity values close to those caused by a saturation abundance of gaseous  $H_2SO_4$  are seen. At lower altitudes, most values for absorptivity are below those caused by a saturation abundance. It should be noted that error bars for this preliminary absorptivity data have not yet been computed, but are expected to be on the order of  $\pm 0.001$  dB/km ( $\pm 0.00023$  km<sup>-1</sup>).

Figure 8 shows the full extent to which application of our laboratory results can be carried. The abundance of gaseous  $H_2SO_4$  (derived from the absorptivity profile in Figure 7 by using laboratory results from Steffes, 1985) is plotted as a function of altitude, along with a plot of the saturation abundance of gaseous  $H_2SO_4$ , for comparison. However, this plot can only be considered valid if the expression for  $H_2SO_4$  opacity at 13 cm is correct. While we have high confidence in the laboratory measurements made by Steffes (1985 and 1986), the question of a scale factor error due to the dissociation of gaseous  $H_2SO_4$  must be resolved, and will be proposed for a laboratory measurement in the next grant year. As our work in the Pioneer-Venus Guest Investigator Program yields more absorptivity profiles for a wide range of locations in the Venus atmosphere, we hope to be able to well characterize the abundance, structure, and spatial variations of gaseous  $H_2SO_4$  in. the Venus atmosphere and to make comparative studies with earlier radio astronomical and radio occultation measurements in order to detect possible temporal variations in H<sub>2</sub>SO<sub>4</sub> abundance and structure.

### E. Publications and Interaction with Other Investigators

In the first half of the current grant year, a paper was revised and accepted for publication in <u>Icarus</u>, describing results and applications of some experiments performed during the previous year of Grant NAGW-533 (Joiner <u>et al.</u>, 1989). This paper describes the results and applications of the laboratory measurements of the millimeter-wave opacity of ammonia between 7.5 and 9.38 mm, described in Section C of this report. In addition, we completed a paper describing observations and interpretive studies of the 1.3 to 3.6 cm Venus emission spectrum (Steffes <u>et al.</u>, 1989). We also submitted updated summaries of our most recent laboratory measurements for inclusion in the twenty-third issue of the <u>Newsletter of Laboratory Spectroscopy for Planetary</u> Science.

We attended the 20th Annual Meeting of the Division for Planetary Sciences of the American Astronomical Society the week of October 30 through November 4, 1988. In addition to presenting our work involving the Venus studies (Jenkins and Steffes, 1988), our latest laboratory results for the millimeter-wave opacity from ammonia were presented by GRA Joanna Joiner (Joiner et al., 1988).

In addition to the radio astronomical observations of Venus and analysis work conducted jointly with Dr. Michael J. Klein of JPL, we have also worked with Dr. Michael A. Janssen of JPL regarding models for the Venus atmosphere, interpretation of microwave emission measurements, and theoretical models for the absorption spectrum of  $H_2SO_4$ . We have also worked with Dr. Arvydas J. Kliore of JPL on the reduction and interpretation of data from recent Pioneer-Venus Radio Occultation Studies as part of our involvement in the Pioneer-

Venus Guest Investigator Program. More informal contacts have been maintained with groups at the California Institute of Technology (Dr. Duane O. Muhleman and his students, regarding interpretation of radio astronomical measurements of Venus and the outer planets), at the Stanford Center for Radar Astronomy (V. R. Eshleman, G. L. Tyler, and T. Spilker, regarding Voyager results for the outer planets, and laboratory measurements), and at JPL (Drs. Robert Poynter and Samuel Gulkis, regarding radio astronomical observations of the outer planets and Venus). We have also worked with Dr. Imke de Pater (University of California-Berkeley) by using our laboratory measurements of atmospheric gases in the interpretation of radio astronomical observations of Venus and the outer planets. We have also studied possible effects of the microwave opacity of cloud layers in the outer planets atmospheres. In this area, we have worked both with Dr. de Pater and with Dr. Paul Romani (Goddard SFC).

Dr. Steffes has also been active in the review of proposals submitted to the Planetary Atmospheres Program at NASA (both as a "by-mail" reviewer and as a member of the March 1988, September 1988, and February 1989 review panels) and as a reviewer of manuscripts submitted to <u>Icarus</u> and the <u>Journal of Geophysical Research</u>, for which Dr. Steffes is an Associate Editor. We have also continued to serve the planetary community through the distribution of reprints of our articles describing our laboratory measurements and their application to microwave and millimeter-wave data from planetary atmospheres. The results of these measurements have been used in the mission planing for radio and radar systems aboard the Galileo and Magellan missions, and more recently, for proposed experiments for the Cassini mission. Dr. Steffes also participated as a member of the International Jupiter Watch (IJW) Laboratory/

Theory Discipline Team. Another source of close interaction with other planetary atmospheres principal investigators has been Dr. Steffes' membership in the Planetary Atmospheres Management and Operations Working Group (PAMOWG). Travel support for Dr. Steffes' attendance at PAMOWG meetings, as well as the November AAS/DPS meeting, has been provided by Georgia Tech in support of Planetary Atmospheres Research. Also in support of Planetary Atmospheres Research, Georgia Tech has provided support for required repairs and maintenance to microwave equipment and the ultra-cold freezer system used in the outer planets atmospheric simulator.

As in the past, we have maintained contact with members of the Georgia congressional delegation, keeping them aware of our work and aware of our continued support for the solar system exploration program. We were especially pleased with the support received from Senator Wyche Fowler for the CRAF/Cassini "new start," after briefing his staff on this issue.

### F. Conclusion

In the first half of this grant year, we have continued our work with laboratory measurements of the millimeter-wave properties of atmospheric gases under simulated conditions for the outer planets. Significant improvements in our system have made it possible to accurately characterize the opacity from gaseous NH<sub>3</sub> at wavelengths as short as 3 mm. The preliminary results of these measurements have been significant in that they indicate that the large opacities predicted by a number of workers at these wavelengths are indeed incorrect and that a form of the modified Ben-Reuven formalism for computing the millimeter-wave opacity from ammonia is correct. In the next six months of the grant, we will complete this study. We will also pursue further

analysis and application of our laboratory results to microwave and millimeter-wave absorption data for the outer planets, such as results from Voyager Radio Occultation experiments and earth-based radio astronomical observations. We also intend to pursue the analysis of available multispectral microwave opacity data from Venus, including data from our most recent radio astronomical observations in the 1.3 to 3.6 cm wavelength range and newly obtained Pioneer-Venus Radio Occultation measurements at 13 cm, using our laboratory measurements as an interpretive tool.

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H. Report Figures

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Figure 1: Diagram of Fabry-Perot Resonator

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Figure 2: Block Diagram of Atmospheric Simulator (30-40GHz)

FIGURE 3: DIAGRAM OF FABRY-PEROT RESONATOR (94GHZ)









Figure 6: Absorptivity of gaseous mixture (5.07% NH<sub>3</sub>, 9.37% He, and 85.56% H<sub>2</sub>) under simulated Jovian conditions (Total pressure: 2 Bars, Temperature: 210 K). Shown for comparison (horizontal lines), are the theoretically computed values from the Van Vleck-Weisskopf formalism (top line), the modified Ben Reuven formalism (middle line), and the Gross formulation (bottom line).

Figure 7 : Comparison of absorptivities measured with radio occultation technique (circular points -- from Pioneer-Venus Orbit 2801entry occultation) with absorptivity which would result from saturation abundance of H<sub>2</sub>SO<sub>4</sub> (from Steffes, 1985). The absorption coefficient scale is logarithmic (exponents of 10). All measurements were made at the 13-cm wavelength (2.293 GHz).



Figure <sup>8</sup>: Abundances of gaseous H<sub>2</sub>SO<sub>4</sub> inferred from Pioneer-Venus 13-cm absorptivity profiles (circular points) compared with the saturation abundance profile of gaseous H<sub>2</sub>SO<sub>4</sub> (from Steffes, 1985). The mixing ratio scale is logarithmic (exponents of 10).

