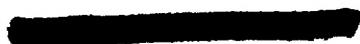


A LISTING OF AVAILABLE EXPERIMENTAL DATA
ON THE FORMATION AND DESTRUCTION OF EXCITED
STATES BY COLLISIONS BETWEEN ATOMIC SYSTEMS

TECHNICAL REPORT NO. 1



By E. W. Thomas

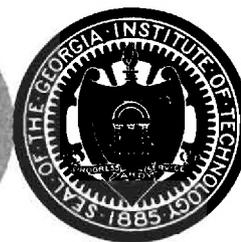
Report No. ORO-2591-22

Contract No. AT-(40-1)-2591

U.S. ATOMIC ENERGY COMMISSION
OAK RIDGE, TENNESSEE



30 October



School of Physics
GEORGIA INSTITUTE OF TECHNOLOGY
Atlanta, Georgia

A LISTING OF AVAILABLE EXPERIMENTAL DATA
ON THE FORMATION AND DESTRUCTION OF EXCITED STATES
BY COLLISIONS BETWEEN ATOMIC SYSTEMS

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E. W. Thomas

Technical Report No. 1.

Project No. B-2021

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Oak Ridge, Tennessee

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Technical Report No. 1, Project No. B-2021

TABLE OF CONTENTS

	Page
INTRODUCTION.....	1
1 The Formation of Excited Atoms and Ions by Impact Between Two Ground State Atomic or Molecular Structures Under Single Collision Conditions.....	4
2 Differential Scattering Associated with the Formation of an Excited State.....	27
3 Collisional Quenching of Excited States Under Single Collision Conditions.....	31
4 The Formation of Excited States in a Beam of Particles Traversing a Gas Cell or Plasma Under Multiple Collision Conditions.....	35
5 The Formation of Excited States by the Passage of a Beam of Particles Through Thin Solid Films.....	40

Technical Report No. 1, Project No. B2021

A Listing of Available Experimental Data
on the Formation and Destruction of Excited States
by Collisions Between Atomic Systems

A listing has been made of the sources of experimental data concerning the formation and destruction of excited states by atom-atom and ion-atom collisions. All processes occurring at impact energies less than 10 eV have been excluded. This compilation is believed to be complete as of 30 October 1966.

The listing is in five sections, each of which includes tabulations of reactants, range of impact energy, assessment of data accuracy and references. The five separate sections are as follows:

Section 1.

The formation of excited states by the collision of two ground state atomic structures under single collision conditions. Such data will in general be expressed in the form of cross sections either on an absolute or relative basis.

Section 2.

Differential inelastic scattering with the formation of an excited state, by the collision of two atomic structures under single collision conditions. Such data will in general be expressed in the form of a cross section.

Section 3.

Collisional quenching of excited states under single collision conditions. Such data will in general be expressed in the form of cross

sections, either on an absolute or relative basis.

Section 4.

The formation of excited states in beams of particles passing through a gas or plasma, under multiple collision conditions. Such data can only be expressed in the form of the relative population of various states in the emergent beam.

Section 5.

The formation of excited states in beams of particles passing through a solid target, under multiple collision conditions. Such data can only be expressed in the form of the relative population of various states in the emergent beam.

Each section is separately accompanied by an explanation of the symbols used and full references. No data obtained prior to 1940 is included since these have been adequately summarized by Maurer and Massey (see references below.).

Reviews

The following reviews are particularly important in this subject and are listed in historical order.

W. Maurer. "The Excitation of Light by Ionic and Atomic Collisions," Phys. Zeits. 40, 161, 1939

H. S. W. Massey and E. H. S. Burhop. "Electronic and Ionic Impact Phenomena." (London, Oxford University Press) 1952, p. 533

S. N. Ghosh and B. N. Srivastava. "Exciting Particles for Auroral Spectra." Zeits. für Astrophysik 53, 186, 1961

S. N. Ghosh and B. N. Srivastava. "Excitation of Spectra by Ion Bombardment." Proc. Nat. Acad. Sci. India A32, 231, 1962

F. J. de Heer. "Experimental Studies of Excitation in Collisions Between Atomic and Ionic Systems." Advances in Atomic and Molecular Physics (Ed Bates and Estermann) Volume II, p. 328, 1966

N. V. Fedorenko, V. A. Ankudinov, and R. N. Il'in. "Lorentz Ionization of Highly Excited Hydrogen Atoms". Soviet Physics, J.E.T.P. 10, 461, 1966

Section 1

The Formation of Excited Atoms and Ions by Impact Between Two

Ground State Atomic or Molecular Structures

Under Single Collision Conditions

Most of the experiments in this listing employed optical detection techniques. Investigations on the formation of metastable and other long lived excited states using alternative techniques are also included. Experiments involving differential inelastic scattering leading to the formation of definite excited states are covered by Section 2.

Column 1 gives the incident fast particles, arranged in order of increasing molecular weight and state of ionization (where applicable).

Column 2 gives the target particle in order of increasing molecular weight.

Column 3 gives energy range in KeV.

Column 4 gives excited state investigated (spectroscopic notation).

Column 5 gives information on data obtained.

Q.....Denotes measurements expressed as cross sections for the excitation of levels, rather than emission functions.

P.....Denotes that the polarization fraction is also measured.

S.....Applies to cases where the excited state may have come from either the target or projectile and denotes excited fast particles have been investigated separately from the target.

Column 6 gives an assessment of the usefulness of the measurements. A series of results is classified as poor on the grounds of error, poorly determined beam composition or energy, or low accuracy.

A.....Denotes good quality absolute measurements.

B.....Denotes poor quality absolute measurements.

C.....Denotes good quality relative measurements.

D.....Denotes poor quality relative measurements.

In cases where the classification D has been assigned, the published information generally consists of little more than an optical emission spectrum with no quantitative measurement of relative intensity of the various spectral lines.

Column 7 comments.

Column 8 reference to published work.

Where a number of papers on one collision combination have been published by the same group, the reference symbols are all included on the same line.

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
H	He	0.5-12	HeI, HI		D	Canal ray poorly defined	En 39
H	He	10-35	HeI	Q P	A		Ec 62, 64i and ii Ec 63 ii
H	H ₂	15	HI		B	2s state fast particle	Se 64
H	N ₂	2.4	N ₂		D	Qualitative no data	Ca 57
H	N ₂	100	N ₂ , N ₂ ⁺		D	Qualitative no data	Br 54
H ⁺	H		HI	S P		Lyman alpha only	St 65
H ⁺	H ₂	2	HI		D		Di 56
H ⁺	H ₂	10	HI	S	B	2s state fast particle	Ma 59
H ⁺	H ₂	7-40	HI	S	C	2s state fast particle	Co 62, Cr 63
H ⁺	H ₂	0.5-3.0	HI		A	Lyman alpha only	Du 62
H ⁺	H ₂	40	HI		A		Ba 62
H ⁺	H ₂	0.15-0.6	HI		D	Upper limit to cross section only	Al 63
H ⁺	H ₂	5-140	HI	S	A	Considerable error on Doppler shifted lines (See	Hu 63 Ph 64)
H ⁺	H ₂	25-100	HI		D	Optical techniques not used n = 9 to 18 states	Ri 63
H ⁺	H ₂	15	HI		B	2s state fast particle	Se 64
H ⁺	H ₂	60-180	HI	S	A	n = 10 state	Il 65i
H ⁺	H ₂	5-115	HI	Q S	A	3s state	Hu 66

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
H ⁺	He	2	HI		D		Di 56
H ⁺	He	2-20	HeI		D		Ha 56
H ⁺	He	40-200	HeI		D		St 61
H ⁺	He	5-35	HeI, II, and HI	Q P	A		Ec 62, 63ii, 64i and ii
		5-100	HeI				He 63ii and 66i
H ⁺	He	200	HeI, II, and HI	Q	A		Hu 61i
H ⁺	He	7-40	HI		C		Co 62, Cr 63
H ⁺	He	1-25	HI		A	Lyman alpha only (2 p state)	Ja 65i
H ⁺	He	20-130	HeI, II	Q	A		Dod 64
H ⁺	He	5-30	HI		A	Balmer series n = 2 to 5 (See corrections)	Bo 64 Bo 65ii)
H ⁺	He	60-180	HI		A	n = 10 state only	Il 65i
H ⁺	He	150-1000	HeI, HeII		C		Th 65
H ⁺	He	150-1000	HeI	Q	A		Th 66
H ⁺	He	25-100	HeI	Q P	A		He 66i
H ⁺	He	5-115	HI	Q	A	3s state	Hu 66
H ⁺	He	10-40	HI		A	2s and 2p states	An 66
H ⁺	Li	10-180	HI		C	Little Data	Il 65ii

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
H ⁺	Ne	5-35	NeI, II, HI		A		He 63i,ii, Ec 63i
H ⁺	Ne	1-25	HI		A	Lyman alpha only (2 p)	Pr 63i, Pr 63ii
H ⁺	Ne	1-25	HI		A	2 s Quenched to 2 p	Ja 65i
H ⁺	Ne	5-30	HI		A	Balmer series n = 2 to 5 (See corrections	Bo 64 Bo 65ii)
H ⁺	Ne	50	HI		C	Highly excited states	Il 65i
H ⁺	Ne	5-115	HI	Q	A	3s state	Hu 66
H ⁺	Ne	10-40	HI		A	2s and 2p states	An 66
H ⁺	Na	10-180	HI		C	Little Data	Il 65ii
H ⁺	N ₂	230	N ₂ ⁺ , N ₂		D	No useful data	Me 52
H ⁺	N ₂	40-230	N ₂ ⁺ , NII, HI		D	Little data, qualitative	Fa 53
H ⁺	N ₂	100	N ₂ ⁺		D	No data, qualitative	Br 54
H ⁺	N ₂	20, 205	N ₂ ⁺		C	No data, qualitative	Fa 56
H ⁺	N ₂	2.4	N ₂ ⁺ , HI		D	No data, qualitative	Ca 57
H ⁺	N ₂	10-30	N ₂ ⁺		D	No data, qualitative	Ro 58
H ⁺	N ₂	1-5	N ₂ ⁺ , HI, NI		B		Ca 58
H ⁺	N ₂	500-1500	N ₂ , N ₂ ⁺		D	No data, qualitative	Ni 59
H ⁺	N ₂	500-1500	N ₂ ⁺		C		Re 60
H ⁺	N ₂	500-1500	N ₂ ⁺		D	Rotational Temperature	Re 61
H ⁺	N ₂	3-30	N ₂ ⁺ , HI		A		Sh 61
H ⁺	N ₂	200	N ₂ ⁺ , NII, HI		A	HI probably in error	Hu 61ii
H ⁺	N ₂	40	HI		A		Ba 62
H ⁺	N ₂	0.5-5	HI		A	Lyman alpha only	Du 62

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
H ⁺	N ₂	5-140	N ₂ ⁺ , NII, HI	Q	A	HI probably in error	Ph 64
H ⁺	N ₂	5-30	HI	Q	C	Population of 3 s, 3 p, 3 d determined separately	Da 65
H ⁺	N ₂	10-65	N ₂ ⁺ , HI	Q	A		Sh 65
H ⁺	N ₂	60-180	HI		A	n = 10 state only	Il 66i
H ⁺	N ₂	500	N ₂ ⁺ , NI, NII		D	Qualitative, no data	Du 65ii
H ⁺	N ₂	30-600	N ₂ ⁺ , NI, NII HI		A		Du 65iii
H ⁺	N ₂	2-35	HI	Q P	A		Mu 66
H ⁺	N ₂	5-115	HI	Q	A	3s state	Hu 66
H ⁺	O ₂	20, 205	O ₂ ⁺		D	No data, qualitative	Fa 65
H ⁺	O ₂	500-1500	OI, OII		D	No data, qualitative	Ni 59
H ⁺	O ₂	40	O ₂ ⁺		D		He 61
H ⁺	O ₂	40	HI		A	2 s state of fast particle	Ba 62
H ⁺	O ₂	5-130	O ₂ ⁺ , OII		A		Hu 64
H ⁺	O ₂	500	O ₂ ⁺ , OI, OII		D	Qualitative, no data	Du 65i
H ⁺	O ₂	30-600	O ₂ ⁺ , OII, HI		A		Du 65iii
H ⁺	O ₂	20-100	OII, OI, O ₂ ⁺ HI	Q P	A		La 65
H ⁺	O ₂	5-115	HI	Q	A	3 s state	Hu 66
H ⁺	Ar	10-35	ArII		A		He 63iii
H ⁺	Ar	1-25	HI		A	Lyman alpha only (2 p)	Pr 63i, Pr 63ii
H ⁺	Ar	1-25	HI		A	2 s Quenched to 2 p	Ja 65i
H ⁺	Ar	50	HI		C	Highly excited states	Il 65i

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
H ⁺	Ar	5-115	HI	Q	A	3 s state only	Hu 66
H ⁺	Ar	10-40	HI		A	2s and 2 p states	An 66

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
H ⁺	K	10-180	HI		C	Little Data	Il 65ii
H ⁺	CO ₂	40	HI		A	2 s state of fast particle	Ba 62
H ⁺	CO ₂	60-180	HI		A	n = 10 state only	Il 65i
H ⁺	C ₂ -HeO	40	HI	S	A	2 s state of fast particle	Ba 62
H ⁺	Kr	10-35	KrII		A		He 63iii
H ⁺	Kr	1-25	HI		A	Lyman alpha only (2 p)	Pr 63i
H ⁺	Kr	10-40	HI		A	2 s and 2 p states	An 66
H ⁺	C ₇ H ₈	40	HI	S	A	2 s state of fast particle	Ba 62
H ⁺	Xe	1-25	HI		A	Lyman alpha only (2 p)	Pr 63i
H ⁺	Xe	1-25	HI		A	2 s Quenched to 2 p	Ja 65i
H ⁺	Xe	10-40	HI		A	2 s and 2 p states	An 66
H ⁺	Cs	0.2-4	HI		B	2 s state only	Don 64
H ⁺	Cs	10-180	HI		C	Little data	Il 65ii
H ⁺	Hg	5-30	HI, HgI		A		Bo 65i

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
H ₂ ⁺	H ₂	2	HI		D		Di 56
H ₂ ⁺	H ₂	5-130	HI	S	B	Doppler shifted lines in error (See	Ha 63 Ph 64)
H ₂ ⁺	H ₂	130, 200	HI	S	B	Probably in error	Hu 61iii
H ₂ ⁺	H ₂	0.5-3	HI		A	Lyman alpha only	Du 62
H ₂ ⁺	H ₂	20 MeV	HI	S	D	n > 5 states	Be 64
H ₂ ⁺	He	2	HI		D		Di 56
H ₂ ⁺	He	2-20	HI, HeI		D		Ha 56
H ₂ ⁺	He	130, 200	HI	S	B	Probably in error	Hu 61iii
H ₂ ⁺	He	0.5-3	HI		A	Lyman alpha only	Du 62
H ₂ ⁺	He	0-25	HI		A	Lyman alpha only	Zy 64
H ₂ ⁺	He	5-30	HI		A	Balmer series (See corrections	Bo 64 Bo 65ii)
H ₂ ⁺	He	1-25	HI		A	2 s state only	Ja 65ii
H ₂ ⁺	Ne	0-25	HI		A	Lyman alpha only	Zy 64
H ₂ ⁺	Ne	5-30	HI		A	Balmer series (See corrections	Bo 64 Bo 65ii)
H ₂ ⁺	Ne	1-25	HI		A	2 s state only	Ja 65ii
H ₂ ⁺	N ₂	500-1500	N ₂ ⁺		C	Rotational Temperature	Re 60
H ₂ ⁺	N ₂	0.505	HI		A	Lyman alpha only	Du 62
H ₂ ⁺	N ₂	500	N ₂ ⁺ , NI, NII		D	Qualitative, no data	Du 65ii
H ₂ ⁺	N ₂	30-600	N ₂ ⁺ , NII		A		Du 65iii

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
H ₂ ⁺	O ₂	500	O ₂ ⁺ , OI, OII		D	Qualitative, no data	Du 65i
H ₂ ⁺	O ₂	30-600	HI		A		Du 65iii
H ₂ ⁺	Ar	1-25	HI		A	Lyman alpha only	Zy 64
H ₂ ⁺	Ar	1-25	HI		A	2 s state only	Ja 65ii

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
H ₂ ⁺	Kr	1-25	HI		A	Lyman alpha only	Zy 64
H ₂ ⁺	Xe	1-25	HI		A	Lyman alpha only	Zy 64
H ₂ ⁺	Xe	1-25	HI		A	2 s state only	Ja 65ii
H ₂ ⁺	Hg	5-25	HI, HgI		A		Bo 65i
H ₃ ⁺	H ₂	200	HI	S	B	Probably in error	Hu 61iii
H ₃ ⁺	H ₂	0.5-6	HI		A	Lyman alpha only	Du 62
H ₃ ⁺	H ₂	5-130	HI	S	B	Doppler shifted lines in error (See	Ha 63 Ph 64)
H ₃ ⁺	He	2-20	HI, HeI		D		Ha 56
H ₃ ⁺	He	200	HI	S	B	Probably in error	Hu 61iii
H ₃ ⁺	He	0.5-4	HI		A	Lyman alpha only	Du 62
H ₃ ⁺	He	5-30	HI		A	Balmer series (See corrections	Bo 64 Bo 65ii)
H ₃ ⁺	Ne	5-30	HI		A	Balmer series (See corrections	Bo 64 Bo 65ii)
H ₃ ⁺	N ₂	500-1500	N ₂ ⁺		C	Rotational Temperature	Re 60
H ₃ ⁺	N ₂	500-1500	N ₂ ⁺		C	Rotational Temperature	Re 61
H ₃ ⁺	N ₂	0.5-5	HI		A	Lyman alpha only	Du 62

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
D ⁺	He	40-200	HeI		C		St 61
D ⁺	He	0.5-25	DI		A	Lyman alpha only	Pr 63ii
D ⁺	He	1-25	DI		A	2 s Quenched to 2 p	Ja 65
D ⁺	Ne	0.5-25	DI		A	Lyman alpha only	Pr 63ii
D ⁺	Ne	1-25	DI		A	2 s Quenched to 2 p	Ja 65
D ⁺	N ₂	75-320	DI, N ₂ ⁺ , NII		D		Fa 53
D ⁺	N ₂	10-65	DI, N ₂ ⁺		A	2 s state only	Sh 65
D ⁺	Ar	0.5-25	DI		A	Lyman alpha only	Pr 63ii
D ⁺	Ar	1-25	DI		A	2 s Quenched to 2 p	Ja 65
D ⁺	Kr	0.5-25	DI		A	Lyman alpha only	Pr 63ii
D ⁺	Xe	0.5-25	DI		A	Lyman alpha only	Pr 63ii
D ⁺	Xe	1-25	DI		A	2 s Quenched to 2 p	Ja 65
D ⁻	H ₂	20 MeV	DI		D	n > 5 states	Be 64
D ₂ ⁺	He	0-25	DI		A	Lyman alpha only	Zy 64
D ₂ ⁺	Ne	0-25	DI		A	Lyman alpha only	Zy 64
D ₂ ⁺	Ne	1-25	DI		A	2 s state only	Ja 65

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
D_2^+	Ar	0-25	DI		A	Lyman alpha only	Zy 64
D_2^+	Kr	0-25	DI		A	Lyman alpha only	Zy 65
D_2^+	Xe	0-25	DI		A	Lyman alpha only	Zy 64

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
He ⁺	H ₂	2-20	HeI, HI		D		Ha 56
He ⁺	H ₂	0.1-2	HI		A	Lyman alpha only	Du 62
He ⁺	H ₂	5-35	HeI	Q	A		He 63ii
He ⁺	H ₂	5-35	HI		A	Balmer series	An 64
He ⁺	D ₂	2-20	HeI, DI		D		Ha 56
He ⁺	He	5-35	HeI	Q S	A		He 63ii, He 64
		5-100	HeI	Q P S	A		He 65i He 66ii
He ⁺	He	20-120	HeI	Q S	A		Hea 65
He ⁺	He	0.04-0.8	HeI	S	B		Sm 66
He ⁺	Ne	5-35	HeI, NeI, II	Q P	A		He 63i,ii, Ec 63i
He ⁺	Ne	0-0.5	?		D	Broad Spectrum detected	Li 66
He ⁺	N ₂	150-450	N ₂ ⁺ , N ₂		D		Fa 53
He ⁺	N ₂	150,450	N ₂ ⁺		C		Fa 56
He ⁺	N ₂	1-3.5			B	"Countable U-V radiation"	Du 62
He ⁺	N ₂	20-120	HeI	Q	A		Hea 65
He ⁺	N ₂	10-65	N ₂ ⁺	Q	A		Sh 65
He ⁺	O ₂	20-120	HeI	Q	A		Hea 65
He ⁺	Ar	10-35	ArII		A		Ec 63iii

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
He ⁺	Ar	100-400	ArII		A		Th 64
He ⁺	Ar	0-0.5	ArII		D		Li 66
He ⁺	Kr	5-35	HeI, KrII		A		He 63ii, Ec 63i
He ⁺	Kr	0-0.5	?		D	Broad spectrum detected	Li 66
He ⁺	Xe	0-0.5	?		D	Broad spectrum detected	Li 66
He ⁺⁺	N ₂	5.3 MeV	N ₂		D	Po _α ²¹⁰ source used	Or 52
He ⁺⁺	N ₂	5.3 MeV	N ₂ ⁺ , N ₂		D	Po _α ²¹⁰ source used	Ni 57
He ⁺⁺	O ₂	5.3 MeV	O ₂		D	Po _α ²¹⁰ source used	Or 52

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
Li ⁺	H ₂	5-35	HI, LiI		A		Ec 61
Li ⁺	He	5-35	HeI, LiI		A		Ec 61
Li ⁺	N ₂	2-4	LiI, NI, NII NIII, N ₂ ⁺		D	No data	Ni 56
Li ⁺	N ₂	3	N ₂ ⁺		C	Rotational Temperature	Lo 65i
Li ⁺	N ₂	4-10	N ₂ ⁺		C	Rotational Temperature	Lo 65ii
Li ⁺	O ₂	2-4	LiI, OI		D	No data	Ni 56
Li ⁺	Ar	2-4	LII, ArI, ArII		D	No data	Ni 56
Li ⁺	Hg	0.2-1.6	HgI		D		Jo 33
Li ⁺	Hg	0.1-1.0	HgI		D		Cl 48
N ⁺	N ₂	10-65	N ₂ ⁺		A		Sh 65
Ne ⁺	H ₂	5-35	HI		A	Balmer series	An 64
Ne ⁺	He	100-400	HeI, II		A		Th 64
Ne ⁺	N ₂	400	N ₂ ⁺ , NII		D	No useful data	Fa 53
Ne ⁺	N ₂	0.4-2	N ₂ ⁺ , NII, NeI		A		Ne 64
Ne ⁺	N ₂	10-65	N ₂ ⁺		A		Sh 65

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
Ne ⁺	Ar	100-400	ArII		A		Th 64
Na ⁺	N ₂	0.4-2	NII, NaI, N ₂ ⁺		A		Ne 64
Na ⁺	N ₂	6-10	N ₂ ⁺		C	Rotational Temperature	Lo 65ii
Na ⁺	Hg	0.1-1	HgI		D		Cl 48
Mg ⁺	N ₂	0.4-2	MgI, II		A		Ne 64
N ₂ ⁺	CO	0.005-0.2	CO ⁺		D		Ut 65
N ₂ ⁺	N ₂	0.5-3	N ₂ ⁺		A		Ne 64
N ₂ ⁺	N ₂	1-10	N ₂ ⁺ , NII		A		Doe 64
N ₂ ⁺	Ar	0.5-3	N ₂ ⁺		A		Ne 64
Ar ⁺	He	5-24	ArII		B		Sl 59i
Ar ⁺	He	100-400	ArII, HeI		A		Th 64
Ar ⁺	He	30-100	ArII		A		Th 64
Ar ⁺	Ne	5-24	ArII		B		Sl 59i
Ar ⁺	Ne	100-400	ArII		A		Th 64
Ar ⁺	Ne	30-100	ArII		A		Th 64
Ar ⁺	N ₂	0.5-3	N ₂ ⁺		A		Ne 64

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
Ar ⁺	Ar	5-25	ArI		D		Sl 59ii
Ar ⁺	Ar	5-24	ArII		B		Sl 59i, Sl 59ii
Ar ⁺	Ar	0.5-3	ArII		A		Ne 64
Ar ⁺	Ar	100-400	ArII		A		Th 64, Th 63
Ar ⁺	Ar	30-100	ArII		A		Th 64
Ar ⁺	Kr	5-24	ArII		B		Sl 59
Ar ⁺	Kr	100-400	ArII		A		Th 64
Ar ⁺	Xe	5-24	XeI		B		Sl 59
Ar ⁺⁺	H ₂	0.1-0.4	HI		B	Lyman alpha only	Du 62
K ⁺	N ₂	0.4-12	N ₂ ⁺ , KI		A		Ne 64
K ⁺	N ₂	5-10	N ₂ ⁺		C	Rotational Temperature	Lo 65ii
Ca ⁺	N ₂	0.4-2	CaI, II		A		Ne 64
Kr ⁺	He	100-200	KrII		A		Th 64
Kr ⁺	Ne	100-200	KrII		A		Th 64
Kr ⁺	Ar	100-200	KrII		A		Th 64
Rb ⁺	N ₂	5-10	N ₂ ⁺		C	Rotational Temperature	Lo 65ii

Incident	Target	Energy Range KeV	Measured Excited States	Absolute and Polarization	Classification	Comments	References
1	2	3	4	5	6	7	8
Cs ⁺	N ₂	5-10	N ₂ ⁺		C	Rotational Temperature	Lo 65ii

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Section 2

Differential Scattering Associated with the Formation of an Excited State

This section lists the very limited data concerning differential scattering associated with the formation of an excited state. Only data where the excited state is specifically identified is included. In principle the excited state may be formed in either the fast particle or the target. The technique currently employed for experimental measurement involves measuring the differential scattering of fast particles which have suffered the specific energy loss corresponding to that energy required to excite the level of interest.

Column 1 gives the incident fast particles, arranged in order of increasing molecular weight.

Column 2 gives the target particle in order of increasing molecular weight.

Column 3 gives the energy range in KeV.

Column 4 gives the scattered particle whose angular distribution is measured. (Present data always considers the scattering of the fast particle).

Column 5 gives the excited particle whose specific state is determined. This may be either the incident or target particle, spectroscopic notation is used.

Column 6 gives an assessment of the usefulness of the measurements. A series of results is classified as poor on the grounds of error, poorly determined beam composition or energy, or low accuracy.

A.....Denotes good quality absolute measurements.

B.....Denotes poor quality absolute measurements.

C.....Denotes good quality relative measurements.

D.....Denotes poor quality relative measurements.

In cases where the classification D has been assigned the data consists only of an inelastic energy loss spectrum without positive identification of specific excited states.

Column 7 comments.

Column 8 references to published work.

Incident	Target	Energy Range KeV	Scattered Particle	Excited Particle	Classification	Comments	References
1	2	3	4	5	6	7	8
He ⁺	He	0.6	He ⁺ (Incident)	HeI (Target)	B		Lo 66
He ⁺	Ne	0.4,0.6	He ⁺	HeII, NeI	D	Qualitative	Ab 66
He ⁺	Ar	0.4	He ⁺	ArI	D	Qualitative	Ab 66
Ar ⁺	Ar	0.4,0.6	Ar ⁺	ArII, ArI	D	Qualitative	Ab 66

References for Section 2

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Section 3

Collisional Quenching of Excited States

Under Single Collision Conditions

This section lists experiments which measure cross sections for destruction of specific excited states of atoms by collision with atomic and molecular structures under single collision conditions. Experiments where the energy of impact of one particle on the other is less than 10 eV have been excluded.

Collisional processes which lead to the removal of excited states include excitation to a higher level, ionization (i.e. excitation to an unbound state), and collisional de-excitation. Most experimental work has been concerned with the measurement of an aggregate cross section for all processes leading to the destruction of a particular excited state in a beam of excited particles. Often no data are available on the state of the atom after de-excitation or on the process involved.

The very limited amount of experimental work is listed in a form most suitable to the type of data being obtained at present.

Column 1 gives incident fast particle and its state of excitation (spectroscopic notation). The listing is in order of increasing atomic weight, increasing state of ionization, and increasing level of excitation.

Column 2 gives target particle listed in order of increasing atomic weight.

Column 3 gives energy of impact in KeV.

Column 4 gives final state of fast particle after quenching collision. Often this information is not available.

Column 5 gives an assessment of the usefulness of the measurements.

A series of results is classified as poor on the grounds of error, poorly determined beam composition or energy, or low accuracy.

A.....Denotes good quality absolute measurements.

B.....Denotes poor quality absolute measurements.

C.....Denotes good quality relative measurements.

D.....Denotes poor quality relative measurements.

Column 6 comments.

Column 7 reference to published work.

Incident	Target	Energy Range KeV	Final State of Excitation	Classification	Comments	References
1	2	3	4	5	6	7
H(n= 4 to 6)	H ⁺ and e	10,000	H(n= 6 to 9)	A	Target, hydrogen plasma	Be 65 i
H(2 s)	H ₂	0.135		B		Fi 59
H(2 s)	H ₂	15		B		Se 64
H(n= 4 to 6)	H ₂	10,000	H(n= 6 to 9)	A		Be 65
H(n = 14)	H ₂	20 - 100	H ⁺	A		Ri 63
H(n < 6)	H ₂	20 MeV	H(n= 6 and 7)	D		Be 64
H(2 s)	H ₂ O	0.135		B		Fi 59
H(2 s)	N ₂	0.135		B		Fi 59
H(2 s)	O ₂	0.135		B		Fi 59
H(n < 6)	Ar	20 MeV	H(n= 6 and 7)	D		Be 64

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Section 4

The Formation of Excited States in a

Beam of Particles Traversing a Gas

Cell or Plasma Under Multiple Collision Conditions

This section lists experiments which give information on the production of excited states in a beam of particles traversing a gas cell. Such experiments involve multiple collision conditions and information on cross sections for specific populating and de-populating processes and can only be obtained indirectly. Where such information has been obtained, it is listed as appropriate under Section 1 or 2. This section includes cases where a "plasma" has been used as the target cell.

In most of these experiments, the gas cell pressure is high enough to ensure equilibrium between the various charge state components of the fast beam but not between the excited states.

Column 1 gives incident fast particle listed in order of increasing molecular weight.

Column 2 gives target particle listed in order of increasing molecular weight.

Column 3 gives energy of impact in KeV.

Column 4 gives the emerging fast particle and the state of excitation investigated (spectroscopic notation).

Column 5 gives information on data obtained

Q.....data giving a quantitative measurement of excited state population in terms of "thickness" of the gas cell.

E..... data shows that equilibrium was established between the excited state population and de-population processes.

Column 6 gives an assessment of the usefulness of the measurements.

A series of results is classified as poor on the grounds of error, poorly determined beam composition or energy, or low accuracy.

A.....Denotes good quality absolute measurements.

B.....Denotes poor quality absolute measurements.

C.....Denotes good quality relative measurements.

D.....Denotes poor quality relative measurements.

At the present time, data for these processes are generally expressed as the ratio of the number of excited particles produced in the target to the number of particles incident on the target or emerging from the target. Consequently, all data are in the form of a relative probability for producing the excited state and have therefore been classified under C or D.

Column 7 comments.

Column 8 references.

Incident	Target	Energy Range KeV	State of Excitation	Quantitative Equilibrium	Classification	Comments	References
1	2	3	4	5	6	7	8
H ⁺	H ₂	20-100	HI	Q E	C	Highly excited states	Ri 63i
H ⁺	H ₂	15	HI	Q E	C	2 s state	Se 64
H ⁺	He	10-30	HI	Q	C		An 65
H ⁺	Li	35 , 42	HI		C	Highly excited states	Fu 63
H ⁺	H ₂ O	35 , 42	HI		C	Highly excited states	Fu 63
H ⁺	Ne	10-30	HI	Q	C		An 65
H ⁺	Ar	30	HI		D	Highly excited states	Sw 62
H ⁺	CO ₂	120	HI	Q E	C	Highly excited states	Il 65
H ⁺	Arc	750			D	Highly excited states	Lu 63
H ₂ ⁺	He	10-30	HI		C		An 65
H ₂ ⁺	Ne	10-30	HI		C		An 65
H ₃ ⁺	He	10-30	HI		C		An 65
H ₃ ⁺	Ne	10-30	HI		C		An 65

Incident	Target	Energy Range KeV	State of Excitation	Quantitative Equilibrium	Classification	Comments	References
1	2	3	4	5	6	7	8
D ⁺	H ₂	100	DI	E	C	Highly excited states	Ri 63ii
D ⁺	He	100	DI	E	C	Highly excited states	Ri 63ii
D ⁺	Ne	100	DI	E	C	Highly excited states	Ri 63ii
D ⁺	Ar	100	DI	E	C	Highly excited states	Ri 63ii
D ⁺	Kr	100	DI	E	C	Highly excited states	Ri 63ii
D ⁺	Xe	100	DI	E	C	Highly excited states	Ri 63ii

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1963 (Pub. SERMA Paris, 1964) p. 105
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Section 5
The Formation of Excited States
by the Passage of a Beam of Particles Through
Thin Solid Films

This section lists experiments which give information on the production of excited states in a beam of particles traversing a thin solid film. Such experiments involve multiple collision conditions and information on cross sections for specific populating and de-populating processes are not obtained.

Column 1 gives incident fast particle listed in order of increasing molecular weight.

Column 2 gives target material listed in order of increasing molecular weight.

Column 3 gives energy of impact in KeV.

Column 4 gives the emerging fast particle and the state of excitation investigated (spectroscopic notation).

Column 5 gives information on data obtained

Q.....data giving a quantitative measurement of excited state population in terms of "thickness" of the film.

E.....data shows that equilibrium was established between the excited state population and de-population processes.

Column 6 gives an assessment of the usefulness of the measurements.

A series of results is classified as poor on the grounds of error, poorly determined beam composition or energy, or low accuracy.

A.....Denotes good quality absolute measurements.

B.....Denotes poor quality absolute measurements.

C.....Denotes good quality relative measurements.

D.....Denotes poor quality relative measurements.

At the present time, data for these processes are generally expressed as the ratio of the populations of various excited states. Consequently, all data are in the form of a relative probability for producing the excited state and have therefore been classified under C or D.

Column 7 comments.

Column 8 references.

Incident	Target	Energy Range KeV	State of Excitation	Quantitative Equilibrium	Classification	Comments	References
1	2	3	4	5	6	7	8
H ⁺	C	157	HI		D		Go 66
N ⁺	C	200-2,000	NII, NIII, NIV		D		Ka 65
N ⁺	C	2,000	NI to NV CI to CIII		D	Spectra only	Ba 64

References for Section 5

- Ba 64 Bashkin. Astrophysical Journal, 139, 413, 1964
Go 65 Goodman and Donahue. Phys. Rev. 141, 1, 1966
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March 1, 1965 to November 30, 1965

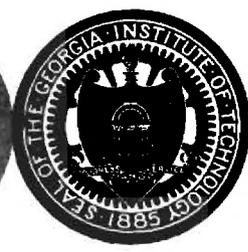
EMISSION AND EXCITATION
CROSS SECTIONS

By E. W. Thomas
G. D. Bent

Contract No. AT-(40-1)-2591

U.S. ATOMIC ENERGY COMMISSION
OAK RIDGE, TENNESSEE

1 December



Engineering Experiment Station
GEORGIA INSTITUTE OF TECHNOLOGY
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TECHNICAL STATUS REPORT NO. 1

PROGRESS REPORT

Project No. B-176-002

Covering the Period

March 1, 1965 to November 30, 1965

EMISSION AND EXCITATION CROSS SECTIONS

By

E. W. Thomas

G. D. Bent

Contract No. AT-(40-1)-2591

U.S. Atomic Energy Commission
Oak Ridge, Tennessee

December 1, 1965

Technical Status Report No. 1, Project No. B-176-002

TABLE OF CONTENTS

	Page
LIST OF FIGURES	iii
I. TITLE.	1
II. INTRODUCTION	1
III. OBJECTIVE AND METHOD	1
IV. STATUS OF THE EXPERIMENTAL WORK.	3
(i) Summary	
(ii) Experimental Apparatus	
(iii) Problems Associated with the Measurement of Target Gas Pressure	
V. EXPERIMENTAL RESULTS	21
VI. PROGRAM FOR THE REMAINDER OF THE CONTRACT YEAR	30
VII. PROGRAM FOR THE FUTURE	31
VIII. PUBLICATIONS AND TRAVEL.	33
IX. PERSONNEL.	33
X. ACCELERATOR USAGE.	33
XI. INCIDENT REPORT.	34

Progress Report

LIST OF FIGURES

Figure	Page
1. Schematic diagram of the experimental arrangement	5
2. Schematic diagram of the modified McLeod gauge, after Meinke and Reich ⁴	16
3. Enlarged diagram of capillary tubes	16
4. Ratio of Argon pressure measured in "normal" mode of McLeod gauge operation to pressure measured in modified gauge.	16
5. Photograph of the emission of visible light from the path of a proton beam passing through a nitrogen target	22
6. Comparison of cross sections for the emission of the 5047 Å HeI ($4^1S \rightarrow 2^1P$) spectral line for protons and electrons of equal velocity incident on helium.	28
7. Comparison of cross sections for the emission of the 4437 Å HeI ($5^1S \rightarrow 2^1P$) spectral line for protons and electrons of equal velocity incident on helium.	28
8. Comparison of cross sections for the emission of the 4686 Å HeII($n=4 \rightarrow n=3$) spectral line for protons and electrons of equal velocity incident on helium.	29
9. Samples of the spectra produced when protons are incident on helium and nitrogen	29

Progress Report

I. Title

Emission and Excitation Cross Sections

II. Introduction

This report summarizes the work performed on excitation phenomena as part of contract AT-(40-1)-2591 for the U.S. Atomic Energy Commission. The work was initiated on March 1, 1965 as a result of Modification No. 7 increasing the scope and financing of the existing contract. This report covers the period 1 March 1965 to 30 November 1965 which corresponds to the first 9 months of the 12 month period covered by the extension to the contract.

The program for measurement of ionization cross sections which was the subject of the original contract and subsequent modifications 1 through 6, has been continued and is reported separately.

III. Objective and Method

The objective of this research is the measurement of cross sections for production of particles in excited states as the result of the impact of protons on targets of hydrogen and other gases. The energy of the incident particle ranges from 0.15 to 1.0 MeV. Previous work in this area has been confined to incident particle energies below 0.15 MeV, with the exception of one series of measurements¹ involving heavy incident ions. Thus the present work utilizing protons as projectiles represents an extension into a region which is largely unexplored. Theoretical predictions are expected to be reasonably accurate over the upper half of this energy range and useful correlations with the experimental results should be

1. Thomas, E. W. and Gilbody, H. B., Proc. Phys. Soc. (London) 85, 363, 1965

Progress Report

possible.

The source of the energetic protons is a 1 MeV Van de Graaff positive ion accelerator, which is equipped with a beam analyzing and stabilizing system. The ion beam is passed through a series of small apertures to provide fine collimation before entering a collision chamber containing the target gas. A quartz window situated in the wall of the chamber allows the photons, emitted by atoms which have suffered collisional excitation, to be spectrally analyzed and detected. The ion beam passes through the observation region and is monitored with a Faraday cup. The chamber dimensions and gas pressure are such that the target is "thin" in the sense that only a small fraction of the incident particles undergo any collisions at all. Also the path traveled by the photons before leaving the gas filled collision chamber has been arranged to be small to reduce absorption.

The photons emitted from the region of the ion beam are focused onto the entrance slit of a grating spectrometer by a simple lens. A low noise photomultiplier at the exit slit serves to detect the photons in the dispersed spectrum. Enhancement of the signal-to-noise ratio is obtained by chopping the light signal with a vibrating reed before it enters the spectrometer and applying a narrow band, phase sensitive detection system to the output of the photomultiplier.

The detection efficiency of the optical system is calibrated by using a substitution technique involving a standard lamp whose emissive power is known in terms of the black body radiator.

In general, the atomic and molecular processes which occur when fast atoms or ions collide with the molecules of the target gas involve the rearrangement of the electrons associated with the two systems. We may use the term "charge rearrangement" to refer to cases where electrons are

Progress Report

ejected from either of the colliding systems or transferred between them. Charge rearrangement may be investigated by observing the change of the ionic composition of the fast beam and the target region. In addition to the gross removal or transfer of charge, there is a probability that one or more of the electrons remaining bound to the nucleus after the collision may be in an excited state. Quantitative measurement of the photon flux emitted as a result of subsequent decay to the ground state allows the determination of the cross section for the formation of excited states.

In general, the work which has been carried out in this field, both on excitation and charge rearrangement, involves observation of the electron rearrangement in only one of the two colliding atomic systems. The detection of changes of the electron arrangement in both colliding atoms, simultaneously, requires a coincidence type of experiment. Such a technique has recently been applied to charge rearrangement processes, but as yet has not been extended to include the formation of excited states. Progress in this direction is urgently needed.

IV. Status of the Experimental Work

(i) Summary

The primary objective of the work proposed for the period covered by this report was to design, construct and test an experimental facility to be used for measuring the cross section for the formation of excited states of atoms as a result of collisions between atomic systems. This objective has been achieved.

A secondary objective was to use this equipment for a broad general survey of the principal excitation processes occurring in various cases and to make a few absolute measurements on a simple collision system for

Progress Report

comparison with theoretical predictions. The broad survey has been completed and the major part of the analysis has already been carried out. Some measurements on the variation of excitation cross sections with energy have been made, but absolute values have not yet been determined. It is anticipated that these secondary objectives will be completed in the remaining part of the contract year (3 months).

As part of the development of equipment for this experiment, a study was undertaken to determine the best means of measuring target gas pressure. Some experimental work was carried out to reduce the effects of the streaming of mercury vapor from the reservoir to the cold trap. In general the results were inconclusive and further consideration will be given to this problem.

(ii) Experimental Apparatus

Figure 1 gives a schematic diagram of the apparatus with the relevant analytical and electronic equipment indicated in block form. There now follows a point by point discussion of the most important features of the apparatus.

a. The Incident Beam Source

The beam of fast ions in the energy range 0.15 to 1.0 MeV is provided by a Van de Graaff accelerator; type JN of the High Voltage Engineering Corporation. The accelerator is mounted vertically and the beam is deflected into a horizontal trajectory by a large electromagnet, which also serves to mass analyze the ions. The proton energy is stabilized by electronic regulation of the accelerator voltage to maintain equal currents on the edges of two slits placed symmetrically about the ion beam path, which amounts to demanding a constant deflection in the regulated

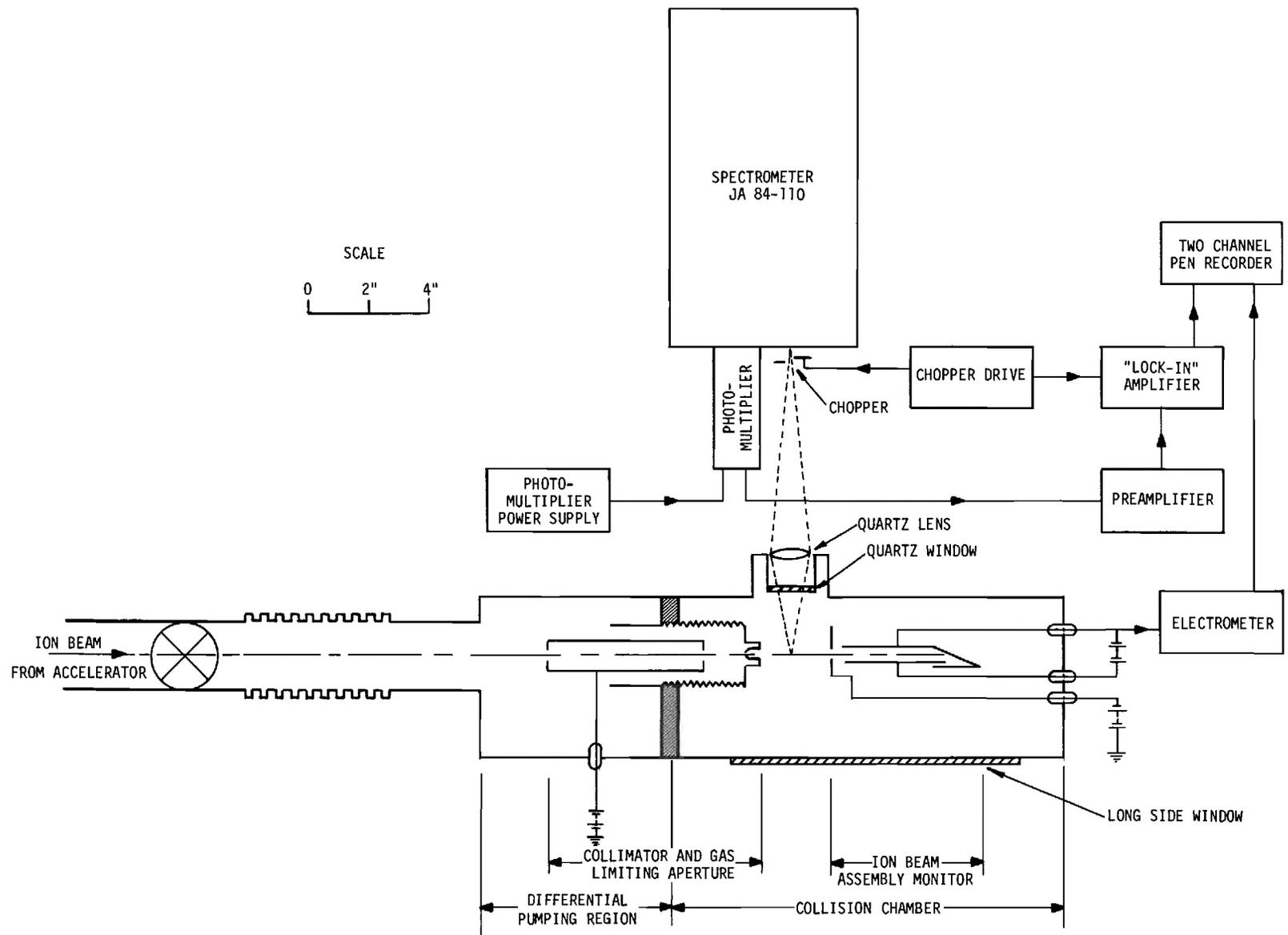


Figure 1. Schematic diagram of the experimental arrangement.

Progress Report

magnetic field. The nominal energy spread of the beam is ± 2 Kev at 1 MeV, the particle energy being determined by the value of the magnetic field and measured by measuring that field. For this purpose, we use a Harvey Wells model G-501 Nuclear Magnetic Resonance precision gaussmeter, which, as used in our experiments, has relative and absolute accuracies of one part in 10^3 . The deflection geometry has been calibrated empirically by measuring the magnetic field corresponding to the 1.019 MeV threshold of the nuclear reaction $\text{He}^3(p, n)\text{He}^3$, using a tritium-zirconium target.

As shown in Figure 1, the ion beam enters the collision chamber through three circular knife edge apertures, mounted co-axially to form a collimator. The first two apertures are of $1/16$ " diameter, mounted at opposite ends of a 12 cm tube, and, therefore, electrically connected. The third aperture is $3/32$ " in diameter and mounted on a tube which projects through the end plate of the collision chamber. The tube carrying the first two apertures is mounted inside the tube carrying the third slit and is insulated from it. The diameters and separations of the apertures are arranged so that the first two provide the collimation of the beam and no ions should impinge on the third. A positive voltage may be applied to the collimating apertures to suppress secondary electrons and to prevent them passing into the collision chamber. The third aperture has the function of defining the physical limit of the gas cell, and it is provided with a long snout of $1/4$ " diameter, to inhibit the pumping of gas into the collimator. A pressure differential of a factor of one hundred can be established between the collision chamber and the collimator region. The collimator lies in a chamber that can be differentially pumped with respect to the rest of the system, and is constructed with a

Progress Report

large number of pumping holes in the tubes so that any gas that does pass through the final slit is rapidly removed from the ion beam path.

A screw thread is cut into the outer wall of the tube carrying the third slit. This allows the collimator assembly to be screwed through the end wall of the collision chamber to vary the separation of the entrance point to the collision chamber and the observation region.

The differential pumping chamber is constructed from type 304 stainless steel, and the collimator assembly is of brass, teflon being used to insulate the two basic components. Good alignment of the three apertures was obtained by constructing the collimator as a single unit and cutting the apertures with the assembled device mounted on a lathe.

b. The Collision Chamber

The collision chamber was constructed from non-magnetic type 304 stainless steel to cut down stray fields. It is intended at some later date to investigate the polarization of the light emitted from this experiment, and it is known that small stray fields can appreciably disturb the relative populations of the magnetic quantum number states of the excited atom, which governs the polarization fraction.

One end of the chamber is blanked off by a plate carrying the ion beam current monitoring assembly, and the opposite end is closed by the collimator tube. A port is provided beneath the beam monitor which connects to a 2" oil diffusion pump used to evacuate the chamber. The usual precautions of using a refrigerated trap to prevent oil streaming into the collision chamber have been observed. A baffle valve is provided to isolate the pump from the system, and a direct line to the mechanical pump may be used for roughing out the chamber.

Progress Report

A port on top of the chamber is used for both pressure measuring devices and the introduction of target gas.

Each side of the chamber is provided with a long port which may be used for the introduction of windows to permit optical observation. One side is designed to carry a plate glass window extending the complete length of the chamber which will be used at some later date to monitor the variation of emitted light intensity with penetration of the fast particles through the target gas. At the present time this window is not in place and a steel plate has been substituted. The opposite side of the chamber has a somewhat smaller port which is designed to accommodate the small circular windows through which observations are made with the spectrometer. Two quartz windows are planned for observation at 90° and 60° to the ion beam path, respectively. These are separately mounted on individual plates and can be interchanged. The 60° window assembly was seriously damaged while being welded to the mounting plate and is currently not available for use.

A considerable amount of ingenuity was involved in trying to keep the diameter of the collision chamber as small as possible and reducing the optical path between the window and the region of the ion beam path under observation. Absorption of resonance photons will tend to repopulate states which are optically coupled with the ground state of the atom, and therefore alter not only the intensity of the resonance radiation but also the intensities of all other spectral lines emitted from this level. It is unlikely that the apparatus in use at the moment will be capable of dealing with many resonance spectral lines since they generally lie in the far U.V., but emission of other lines from the excited states which are

Progress Report

optically coupled with the ground state, will be observed. It was originally intended to mount both 60° and 90° windows together, but the necessity of keeping optical paths short required that they be mounted separately and we must live with the inconvenience of breaking into the vacuum system whenever a change of viewing angle is desired.

The collision chamber is now complete except for the damaged 60° window, and background pressures of 10^{-5} mm Hg are being achieved. Although this background pressure is not quite as low as expected, we consider it to be adequate for the present program.

c. Monitoring of the Ion Beam

After passing through the region of the collision chamber from which optical measurements are made, the fast particle beam passes into a deep Faraday Cage structure where it may be monitored. As shown in Figure 1, the Cage is formed from two plates which taper together to form a blind end. The ion beam impinges on one plate; any secondary electrons are drawn to the second plate by a positive potential, and are therefore prevented from returning towards the observation region. The total current to the two plates is equal to the ion beam current and is monitored by a Keithley Model 410 Electrometer which drives one channel of a two-channel pen recorder. Additional safeguards against loss of ions and secondary electrons from the cage include giving the collection plates overlapping walls to enclose completely the cage region, and providing an aperture plate in front of the system to which a negative potential may be applied to return electrons to the plates.

Tests to ensure that the suppression fields are adequate will be carried out in the near future. For the purposes of the general survey of

Progress Report

collision combinations which provides the bulk of the work reported here, the loss of a few ions or electrons from the detection region is not too serious. Voltages are currently being maintained at values which previous experience has shown to be reasonable for this purpose.

d. Gas Handling and Pressure Measurement

The gas samples used in the present work were admitted to the chamber through a mechanical leak. A cold trap, refrigerated with dry ice and acetone or liquid nitrogen, was used to remove any condensible materials (water vapor, oil, etc.). The gases are obtained from Matheson in high pressure cylinders and are stated to be 99.9% pure. No further purification procedures were used in the work reported at this time.

Three pressure measuring devices are provided on the present equipment; McLeod gauge, ionization gauge, and Pirani gauge. These are connected in parallel to a tube which projects through the upper port in the chamber, to within 1" of the observation region. Pressure gradients may be anticipated in the chamber and this projecting tube, sometimes called a "sniffer," ensures that the target density is determined in the region where optical measurements are made. The ionization gauge is only used for determining base pressure before the start of a day's operations. The Pirani gauge is used as the monitor of gas pressure and will be periodically calibrated against the McLeod gauge. As mentioned in a later section (IV, iii), we are still not entirely happy with the pressure measuring arrangement. A cold trap must be inserted between the McLeod gauge and the experimental region to prevent contamination of the target gas with mercury, and this technique is known to introduce errors. It seems clear that with reasonable precautions we may use this system to measure pressure

Progress Report

of light gases, which will be the subject of our initial program. However, it is expected that a capacitance manometer type of gauge employing a metal diaphragm (micro-manometer) will be more suitable and this possibility is being explored (see Section IV, iii).

e. Optical Equipment and Signal Processing

Light emitted from the collision region is focused by a quartz lens onto the entrance slit of an evacuable 1/2 meter grating spectrometer, Jarrel Ash Model 84-110. The optical system of the unit is of the Ebert-Fastie type, and photomultiplier detection is used at the exit slit. The wavelength range that the spectrometer is expected to cover at the present time is from 2,000 to 7,000 Å, the upper limit being set by the poor sensitivity of available detectors, and the lower limit being due to the photo-absorption by air and water vapor. There are plans to extend the lower limit to 1,000 Å at a later date, by evacuating the spectrometer.

Most of the experimental problems associated with this type of experiment are caused by the very low light levels which must be detected quantitatively. Under certain conditions the signal obtained from the photomultiplier will be comparable with the inherent noise current of the device and an A.C. detection technique has been adopted to enhance signal-to-noise ratios. A mechanical chopper, similar to a tuning fork, has been placed in front of the entrance slit of the spectrometer. The light signal is therefore modulated and may be separated from the inherent noise of the photomultiplier by utilizing its specific frequency and phase. The mechanical chopper, supplied by American Time Products, provides a reference signal bearing a constant phase relation to the chopper motion, which is used to drive the phase sensitive detector circuit of the Prince-

Progress Report

ton Applied Research Corporation "Lock in Amplifier." The output is displayed on one channel of a two-channel pen recorder, where it may be compared directly with the trace displaying the ion beam current.

Considerable care has been taken to ensure that the signal-to-noise ratio obtained from the output is as large as possible. The whole of the optical aperture of the spectrometer is filled with light, and a special wide slit system has been obtained which allows us to obtain a considerable increase in the transmitted light intensity, although this is achieved with a consequential loss of resolution. The photomultipliers available for use with the spectrometer are types 9502S and 6256B made by E. M. I. of England. The published specifications of these tubes indicate that they are the best types commercially available, and the units purchased were further specially selected to have the largest possible signal-to-noise ratio.

It is intended to calibrate the detection sensitivity of the optical system by comparing the observed emission with a standard lamp of known emissive power. This is not a very satisfactory procedure since the standard is up to five orders of magnitude more intense than the system under investigation. Obviously it is necessary to introduce a substantial attenuation, either optically or through the electronics, and the accuracy with which this large attenuation factor is known will not be very great. The calibration procedures will be carried out in the near future before a detailed program of measurement is commenced and, until that time, it would be hazardous to guess what accuracy will be achieved. However, the experience of others in this field has shown that an uncertainty of $\pm 15\%$ may not be an unreasonable figure.

There is some possibility of using the collision process itself to

Progress Report

calibrate the system, normalizing the observed signals to a case where the cross section may be predicted theoretically with reasonable certainty. Past experience² has shown that in cases where the Born approximation can be solved, it predicts ionization cross sections to within 5% above a collision energy of 500 Kev. There is a reasonable expectation that this will also be the case for excitation collisions, although no experiments have yet been done at a suitably high energy for this contention to be checked. Comparisons will be made between the experimental results and the calculated Born approximation, and it is hoped that sufficient agreement will be found to warrant discarding the standard lamp as the reference source and normalizing all measurements to a calculated cross section.

(iii) Problems Associated with the Measurement of Target Gas Pressure

The measurement of a cross section for a particular atomic collision process, using the experimental configuration of a particle beam passing through a static target gas, requires the determination of the number density of the gas. Typical target gas pressures in an atomic collision experiment range from 10^{-5} to 10^{-2} mm Hg, and the only device capable of making an absolute pressure measurement in this region is the McLeod gauge. A cold trap must be used with the gauge to prevent contamination of the experimental system with mercury. It has recently been shown that the streaming of mercury into this trap introduces a pumping effect that can seriously affect the accuracy of the pressure measurement.^{3,4,5}

2. Hooper, J. W., et al., Phys. Rev. 125, 2000, 1962.

3. Ishii, H. and Nakayama, K. I., Trans. 8 Nat. Vac. Symp., Pergamon Press, London (1962), p. 519

4. Meinke, C. and Reich, G., Vakuum Technik. 12, 79, (1963)

5. Takaishi, T., Trans. Faraday Soc., 61, 840 (1965)

Progress Report

It is vitally important to the present project and to similar programs that the pumping error be eliminated or a different type of gauge found for pressure measurement, and considerable thought has been given to the problem.

A decision was made to carry out the modification to the McLeod gauge suggested by Meinke and Reich.⁴ These have the advantages of involving relatively minor and low cost alterations to the standard McLeod gauge. A schematic diagram of the modified McLeod gauge is shown in Figure 2. The tube connecting the gauge to the mercury reservoir was replaced by a longer (8 cm) tube of smaller (1 mm) diameter. A ball valve operated by an external solenoid was inserted between the gauge and the cold trap.

In operation the modified McLeod gauge is used in the following manner. The gauge is connected to the experimental system through the cold trap in the normal way, and initially the ball valve is held open. The capillary tube connecting the mercury reservoir to the gauge proper introduces a considerable impedance to the flow of mercury vapor towards the cold trap, and the equilibrium pressure established in the gauge should be very close to that in the experimental system. The ball valve is now closed, the mercury column raised, and the pressure measurement made in the normal manner. The use of the ball valve ensures that no mercury streaming takes place while the mercury is being raised to cut off the sample volume of gas in the bulb of the gauge.

In order to get some idea of the magnitude of the errors involved in the use of the normal McLeod gauge, it is necessary to make some comparison of the modified gauge with one used in the normal manner. In principle, this could be done using a normal gauge and a modified gauge to make simul-

Progress Report

taneous measurements of the pressure of a suitable gas reservoir. However, it is also possible to make this comparison using the same gauge in two modes of operation and making successive measurements of pressure in the gas reservoir. Such a procedure eliminates problems associated with the mechanical differences that are often found between two nominally similar McLeod gauges.

In our work, the following procedure was adopted. A measurement of the pressure could be made in the "normal" manner by keeping the ball valve open, and the mercury level just below the "cut off" point. (Point C in Figure 2.) Mercury was then free to stream onto the cold trap in the manner expected, since the impedance of the connecting tubing is relatively small. A measurement of pressure made by raising the mercury in the conventional manner would then be affected by the streaming problem. The gas pressure in the reservoir was maintained constant, and a series of measurements made using alternately the "modified" and "normal" techniques. After each individual measurement, a period of about 20 minutes was allowed for the system to reach an equilibrium situation. There was no evidence that lengthening or shortening this time delay had any effect on the measurements. A series of eight measurements of the pressure was made alternately with each mode of operation. If the four measurements in each mode gave the same value of pressure, within the experimental error, it was assumed that the gas pressure in the reservoir had remained constant.

The actual measurement of gas pressure was made by taking the product of the volume of gas compressed in the closed capillary and the pressure of this gas measured in mm Hg from the height of the mercury column in the open capillary. The product of the two lengths H and Δh (see Fi-

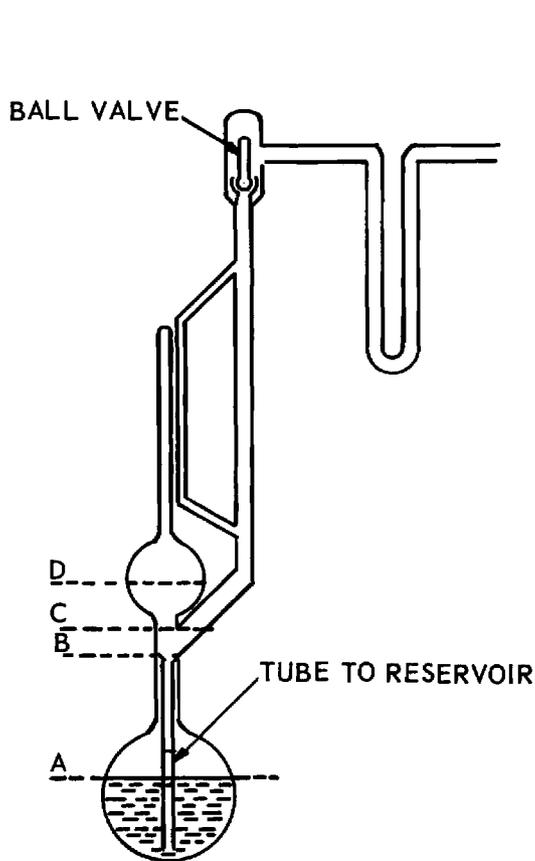


Figure 2. Schematic diagram of the modified McLeod gauge,⁴ after Meinke and Reich.

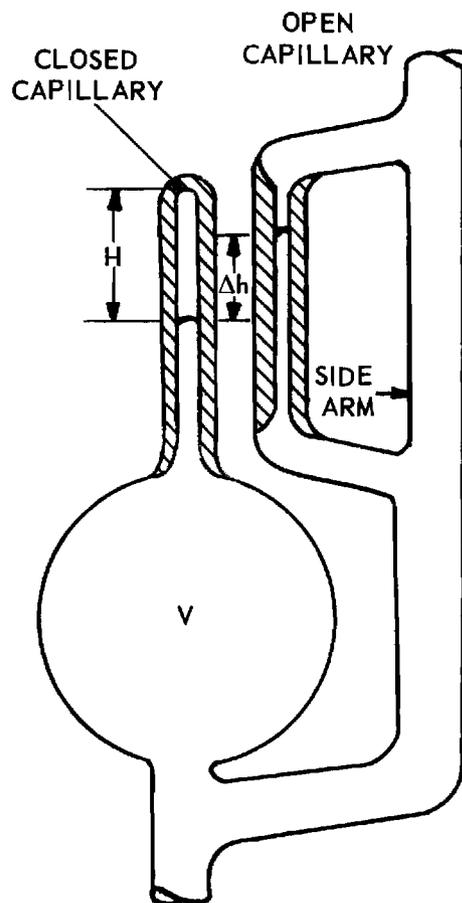


Figure 3. Enlarged diagram of capillary tubes.

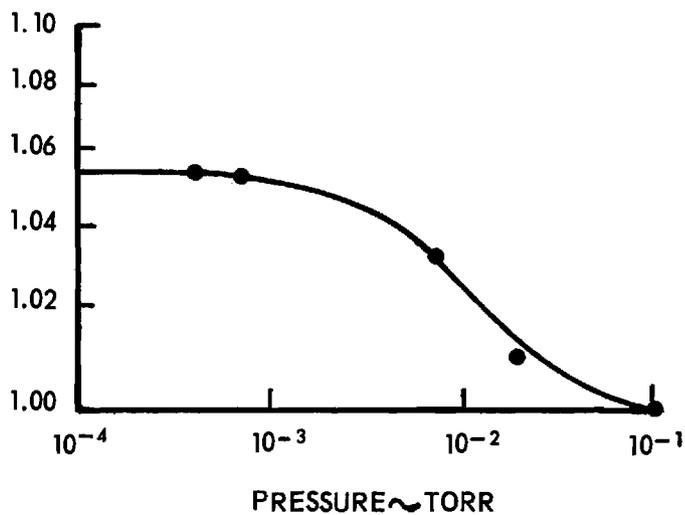


Figure 4. Ratio of Argon pressure measured in "normal" mode of McLeod gauge operation to pressure measured in modified gauge.

Progress Report

gure 3), is proportional to the pressure of the test gas before compression. Suitable normalizing factors involving the volume of the bulb and the bore of the capillaries can be derived and measured but in the present work where the comparison is made between the two modes of operation of the one McLeod gauge, this is unnecessary. For each determination of pressure, a series of measurements was made of the variation of H with the applied mercury column pressure Δh . By use of the techniques suggested by Carr,⁶ capillary depression errors in the closed capillary were eliminated and the proportionality of H to Δh^{-1} was taken as indicating that the adsorption of gas molecules on the gauge walls was negligible.

No special precautions were taken to ensure the cleanliness of the McLeod gauge. A fairly strong bake out was given to the closed capillary tube, and this was found to improve reproducibility, presumably by reducing the sticking of the mercury in the tube. The cold trap used was a symmetrical type⁶ in which thermal transpiration effects should be negligible.

The technique of measuring the volume of the compressed gas in the closed capillary (proportional to H) for various applied pressures (proportional to Δh) ensures that each measurement of gas pressure is the mean of about eight determinations.

The product of H and Δh was taken as being proportional to the reservoir pressure. The ratio of this product measured by the modified technique to that obtained with the "normal" mode of operation gives an indication of the error due to the pumping of mercury vapor onto the cold trap. If such error is negligible, the ratio would be unity.

Attempts were made to measure this error for helium, nitrogen, and

6. Carr, P. H., Vacuum, 14, 37, (1963)

Progress Report

argon gases. Since the error function is predicted to vary with pressure, gradually disappearing as the pressure regime changes from molecular flow to stream line flow, the error was measured at various ambient pressures. Calculations were made of the predicted error function using the simple formulae given by Meinke and Reich,⁴ and compared with experiment.

At pressures of about 10^{-3} mm Hg, the measured error for helium, nitrogen, and argon was found to be respectively, 2%, 2%, and 5%, with an uncertainty in each case of about $\pm 2\%$. In the case of argon, some variation with pressure was found (see Figure 4). No attempt was made to investigate the variation of error with pressure in the other cases since the accuracy was already so poor under the conditions where the effect was supposed to be a maximum that such measurements would have been meaningless. These experimental values must be compared with the predicted values of 2.8%, 11%, and 12%, respectively.

It is concluded that the errors due to the mercury streaming effect in our McLeod gauge were substantially below those predicted by simple theory, and in two of the three cases were of the same magnitude as the random uncertainty in the pressure measurement. When the additional uncertainties associated with bulb volume and capillary diameter are also considered, it must be concluded that the error due to the pumping effect was in no case greater than the estimated limits of accuracy of the gauge established by the other parameters (5%).

Since most of the publications which have sought to confirm the pumping error by experiment have reproduced the theoretical predictions fairly well, an explanation of our diverging conclusion must be found. Private conversations with others interested in this problem have brought

Progress Report

to light further instances in which experiment has not confirmed theory. Unfortunately these have not been published. It is suggested that the discrepancy may arise due to the use of "dirty" McLeod gauges. A film of dirt and chemical compounds on the surface of the mercury might appreciably inhibit the evaporation into the evacuated region above it. The streaming to the cold trap would, therefore, be considerably less than that predicted. In the cases where experimenters have sought to measure the effect accurately, they have taken considerable care to clean their gauges and mercury. Thus, the published data on the subject has probably all been taken under conditions where the effect is at a maximum.

We conclude from our own observations that the error in the McLeod gauge used in this experiment is less than five percent for the gases investigated, and hence by inference it is probably equally small for other similar gases. The technique used by Meinke and Reich seems to be fool-proof in that the pumping effect is eliminated, rather than measured and corrected for, as has been the case in certain other experiments.

The present line of investigation is not being pursued further, since the uncertainty in the measurements is often greater than the error function that is being investigated. A further instrumental problem that arises with the Meinke and Reich gauge, which was not mentioned in the original article, is that the small diameter of the tube connecting the gauge to the mercury reservoir considerably restricts the flow of mercury when the level is being raised and lowered. The time required to make a single measurement becomes inconveniently long, and considerable turbulence is experienced. A McLeod gauge modified in the Meinke and Reich manner is not a convenient instrument to use. It would also seem that the error in-

Progress Report

involved in the use of a dirty McLeod gauge is so small that it can be neglected in comparison to other problems. Since gauges used with experiments which do not of themselves require high purity techniques are often not subjected to extreme cleaning techniques, it would be a reasonable assumption that much of the published work on excitation phenomena has not been unduly affected by the problem.

In principle, the McLeod gauge should be a reasonably sensitive and accurate method of measuring gas pressure. However, recent experience in many laboratories⁶ has shown that the ultimate capabilities of the instrument are only achieved with the use of highly sophisticated observation techniques. This is a considerable disadvantage when the device is to be used for frequent routine pressure determinations. Recent reports^{7,8} suggest that a suitable alternative pressure gauge may be devised around the deflection suffered by a metal diaphragm when subjected to a pressure differential. The most detailed assessment of commercially available gauges of this type,⁸ shows that the "Baratron"⁹ is both highly stable and accurate. It is suggested that this device is certainly adequate as a routine instrument, and in principle could also be used as a primary standard.

We have come to the conclusion that the McLeod gauge is not suitable for the convenient measurement of low pressures, and it is hoped to acquire a "Baratron" in the near future. Until such time as funds are available for this purpose, a McLeod gauge is being built into our experimental system. The measurements planned for the immediate future will involve helium

7. Barnett, C. F., Private communication

8. Romy, P. R., UCRL-11218, TID-4500, AEC Contract No. W-7405-eng-48

9. M. K. S. Instruments, Inc., Burlington, Massachusetts

Progress Report

targets and it is predicted^{3,4,5} that the errors due to pumping of mercury and adsorption of gas on the capillary tube⁶ should be negligible in this case.

V. Experimental Results

The construction of apparatus for the excitation experiment was completed in late September. During the following month, the spectra induced by the impact of protons on He, Ar, H₂, and N₂ target gases were recorded and analyzed. From these data, a choice can be made of the transitions which are most suitable for emission cross section determinations. The spectra obtained from the impact of He⁺ ions on these same gaseous targets were also considered, but analysis of these cases was hampered by the very low ion beam currents obtained from the accelerator. Three lines were selected from the spectrum emitted as a result of proton impact on a helium target and measurements made of the variation of cross section with energy of the incident particle.

As is shown in Figure 1, provision is made for inserting a long window into one side of the collision chamber. During normal operating conditions when the spectrometer is in use, a steel plate covers this aperture. An attempt was made to observe visually the photon emission from the track of the ion beam through the target gas, by looking through this long window. Light emission could be seen in the cases of proton impact on nitrogen and He⁺ impact on nitrogen and argon. Figure 5 is a photograph showing the emission observed visually when nitrogen is bombarded by protons. The emission of light appears to come from a track approximately 1/4" wide, while the ion beam entering from the left of the photograph is restricted to 1/16" diameter by the collimator assembly.

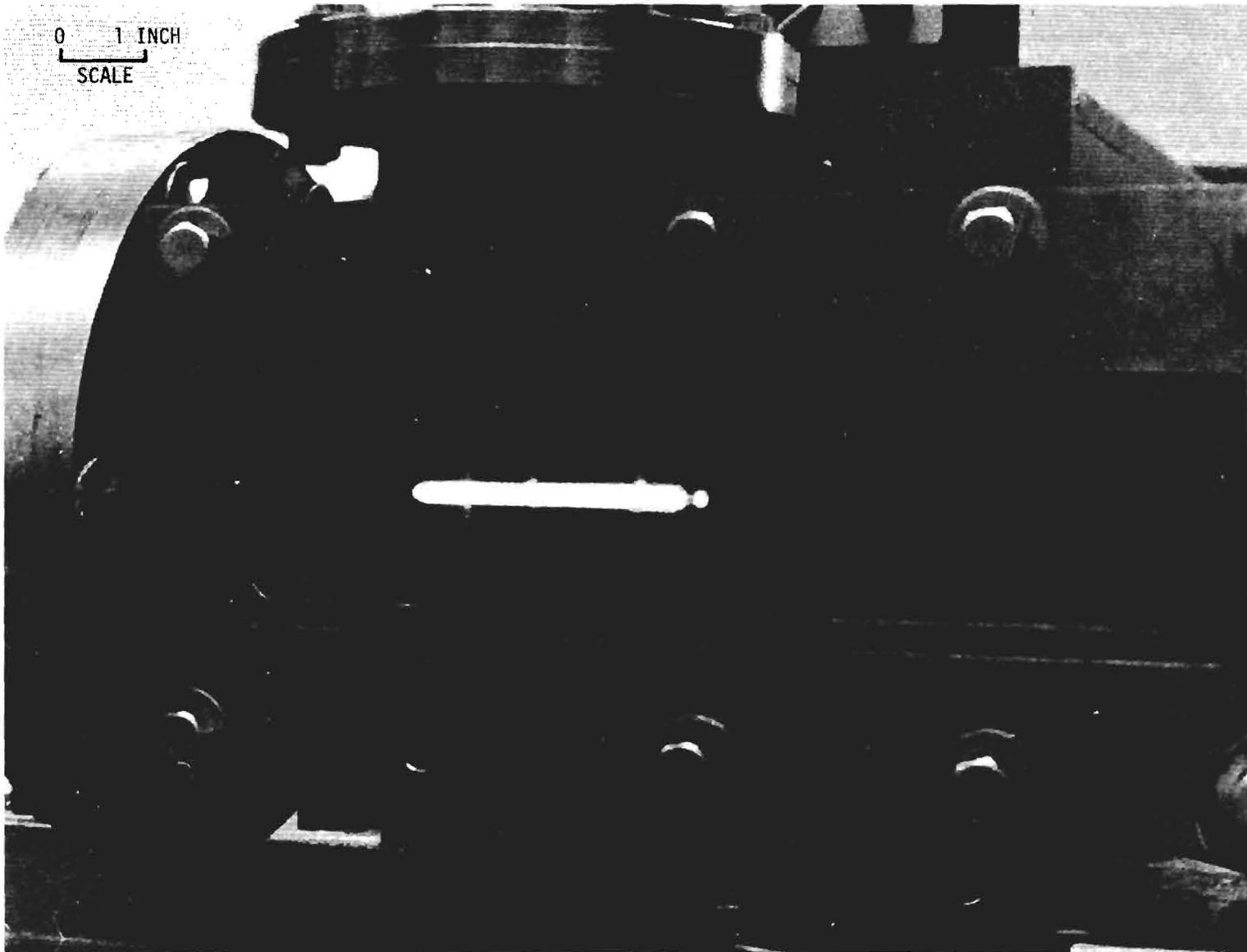


Figure 5. The emission of visible light from the path of a proton beam passing through a nitrogen target. The beam enters the chamber through the collimator at left.

Progress Report

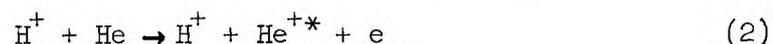
Analysis of the spectra produced in this case shows that the emission comes primarily from excited target particles, generally N_2^+ ions. We believe that the excited particles acquire an appreciable kinetic energy as a result of the collision and may move out of the ion beam path before decaying to the ground state and emitting a photon. Using this model, simple considerations suggest that the decrease of intensity with distance from the ion beam path should be exponential and knowledge of the lifetime of the excited state should allow a very rough assessment of the average velocity of the excited target particle.

Figure 9 shows typical spectra obtained for the impact of protons on helium and nitrogen. In the case of helium, the light emission was very weak and the noise level has been removed from the trace shown. The emission from nitrogen was about two orders of magnitude more intense and noise was negligible.

The impact of protons on helium is observed to produce spectral lines from both the neutral (HeI) and singly ionized (HeII) helium systems. Neutral helium lines must arise from the direct excitation process.



The ionized helium lines arise from either an ionization process,



or a charged transfer process.



At the energies used in this experiment, process 2 is expected to predominate over 3 for the formation of excited helium ions.

The impact of protons on nitrogen shows primarily the N_2^+ first

Progress Report

negative system with less intense traces of excited N^+ and H. The formation of excited neutral hydrogen will be by the process



that is to say, by charge transfer leaving the fast neutral atom in an excited state.

The impact of protons on argon showed a very complex spectrum which was difficult to analyze due to the poor signal-to-noise ratio. No wavelengths of ArI were definitely identified, but the ArII spectrum was very prominent, particularly the 4610 Å transition which has shown an odd behavior in previous measurements on excitation processes.¹ There was also some evidence for the formation of excited doubly ionized atoms through the tentative identification of seven ArIII spectral lines.

The impact of protons on molecular hydrogen gave rise to the emission of the Balmer alpha and beta lines. Without the use of the 60° viewing position, we were unable to decide whether this emission came from the incident fast particle or from the target.

Table I summarizes the most prominent spectral features observed for the impact of protons on various targets.

Table I

<u>Collision</u>	<u>Observed Spectra</u>
$H^+ + He$	HeI HeII
$H^+ + Ar$	ArII ArIII
$H^+ + H_2$	HI
$H^+ + N_2$	N^+ , N_2^+ , HI

A number of spectra were obtained for the impact of He^+ ions on He,

Progress Report

Ar, H₂, and N₂ targets, the identified lines being entirely from the excited target. However, a very poor current of He⁺ ions was obtained from the accelerator and analysis of the spectra was therefore hampered by the high noise level. An improvement in the operation of our ion source will be necessary before detailed measurements could be made on such cases. There are no plans to carry out such measurements in the immediate future.

From the spectrum emitted as a result of the impact of protons on a helium target, three spectral lines were chosen for a brief study of the variation of emission cross section with energy. The transitions were:

HeI 5047 Å (4¹S - 2¹P); HeI 4437 Å (5¹S - 2¹P); HeII 4686 Å (n=4 - n=3).

The processes leading to these spectral lines have already been discussed above. The data we present must be regarded as preliminary; the main object of this study was to confirm the satisfactory operation of the equipment.

For this series of measurements, the target gas pressure was maintained at about 5×10^{-3} mm of Hg. Previous work at lower energies¹⁰ has shown that single collision conditions exist up to substantially higher pressures than this. The spectrometer was set on the wavelength of interest and the resolution adjusted to reject any adjacent spectral lines.

For thin target conditions, the number of photons, n_λ , of wavelength λ , emitted from 1 cm of the ion beam path through the chamber is related to the cross section for the emission of the spectral line, σ_λ , by the equation

$$n_\lambda = \sigma_\lambda \rho \frac{I}{e},$$

10. Van Eck, J., De Heer, F. J., Kistemaker, J., *Physica* 30, 1171, (1964)

Progress Report

where ρ is the number of atoms/cc in the target and I/e is the incident beam current measured in ions per second. Now, the signal obtained from the electronics, S_λ , is related to the photon emission n_λ ; the coefficient of proportionality, A_λ , is essentially the detection sensitivity of the system governed by optical, geometrical, and electrical parameters.

$$n_\lambda = A_\lambda S_\lambda$$

Therefore

$$\sigma_\lambda = \frac{S_\lambda}{I} \left(\frac{A_\lambda e}{\rho} \right)$$

In the work we present here, $A_\lambda e/\rho$ is constant and the ratio of output optical signal to ion beam current is proportional to the emission cross section of the transition of wavelength λ . The ratio of the signal from the optical apparatus to the ion beam current at various incident proton energies in the range 0.15 to 1.0 MeV allows the determination of the variation of emission cross section with impact energy.

Strictly speaking, these cross sections are for the emission of a photon of one specific wavelength. However, it may be shown that cross sections for excitation to the upper level of the transition may be related to the emission cross section provided cascade contributions and branching ratios are known. Previous work by others^{10, 11} has shown that cascade into the n^1S levels is negligible at energies up to 150 keV. We may, therefore, assert that the emission cross section for the $n^1S - 2^1P$ line is proportional to the cross section for excitation of the n^1S level. This is not the case for the emission of the HeII ($n=4 - n=3$) transition where cascade contributions have not been assessed. We have not made absolute determinations of the cross sections but have normalized our data

11. Dodd, J. G., Hughes, R. H., Phys. Rev. 135, A618 (1964)

Progress Report

to the measurements of emission cross section made by Dodd and Hughes at 150 keV.¹¹ The present results and those by Dodd¹¹ are presented in Figures 6, 7, and 8, plotted on a log - log graph. Also shown on these graphs are emission cross sections for electron impact on helium plotted on the same velocity scale. The electron impact data for the 5047 Å and 4437 Å lines by St. John¹² are not absolute and have been normalized to our proton data at 0.9 MeV. The data for the 4686 Å line by Hughes¹³ are absolute and have not been re-normalized to our proton data.

The simple Born approximation predicts that at sufficiently high energies the cross sections for electron and proton impact excitation of any one level should have the same energy dependence.¹⁴ In the case of the excitation of the 5¹S and 4¹S levels of neutral helium, we observe that the curves of electron and proton impact data tend to the same slope at high impact velocities. The comparison is very similar to that observed when considering proton and electron ionization of helium.² In the case of the emission of HeII line 4686 Å (n=4 - n=3), the comparison between the electron and the proton impact data is not so good.

It is to be noted that our data normalize to the work of Dodd and Hughes¹¹ to give a very smooth curve. This gives us some confidence in the validity of our work.

The emission cross sections measured in our energy range can be represented by an equation

$$\sigma_{\lambda} = A E^{-C}$$

12. St. John, R. M., et al., Phys. Rev. 134, A888, (1964)

13. Hughes, R. H., Weaver, L. D., Phys. Rev. 132, 710, (1963)

14. McDaniel, E. W., "Collision Phenomena in Ionized Gases," Wiley, 1964 page 326.

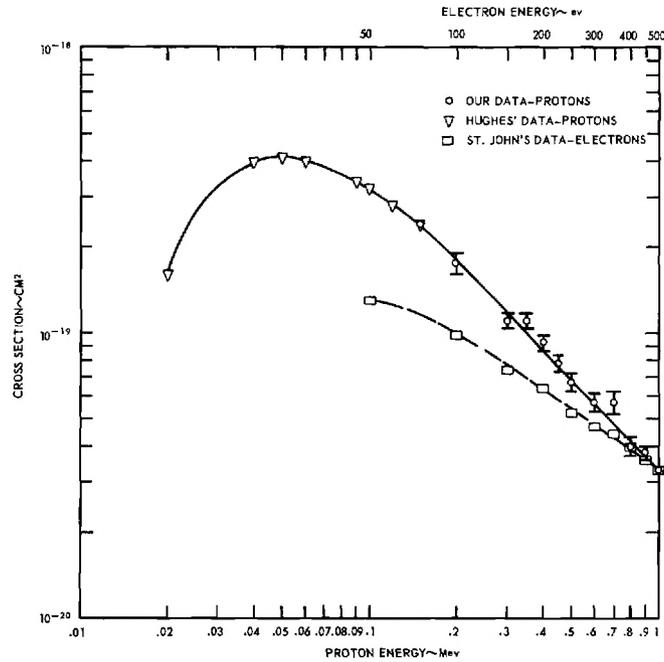


Figure 6. Comparison of cross sections for the emission of the 5047 Å He I ($4^1S \rightarrow 2^1P$) spectral line for protons and electrons of equal velocity incident on helium.

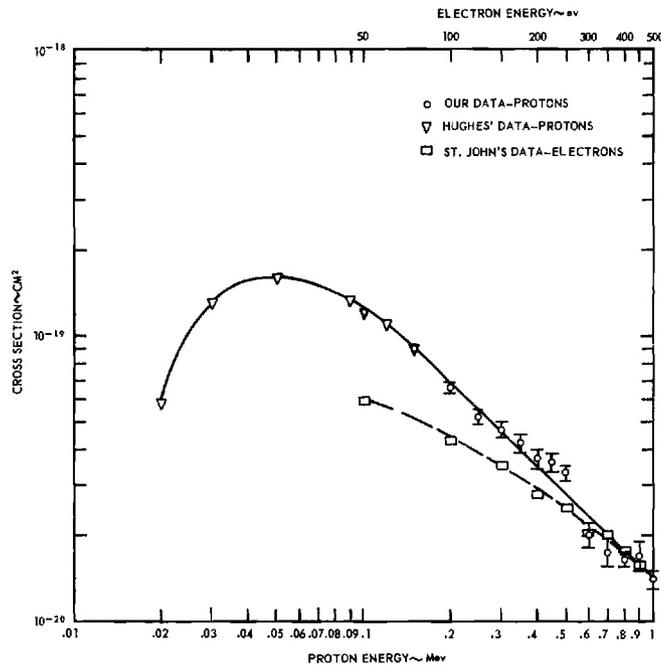


Figure 7. Comparison of cross sections for the emission of the 4437 Å He I ($5^1S \rightarrow 2^1P$) spectral line for protons and electrons of equal velocity incident on helium.

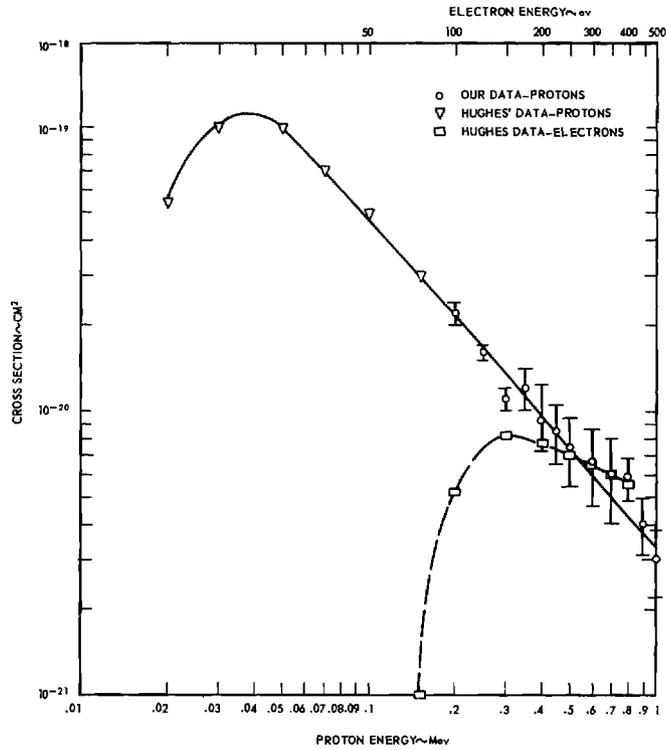


Figure 8. Comparison of cross sections for the emission of the 4686\AA HeII ($n = 4 \rightarrow n = 3$) spectral line for protons and electrons of equal velocity incident on helium.

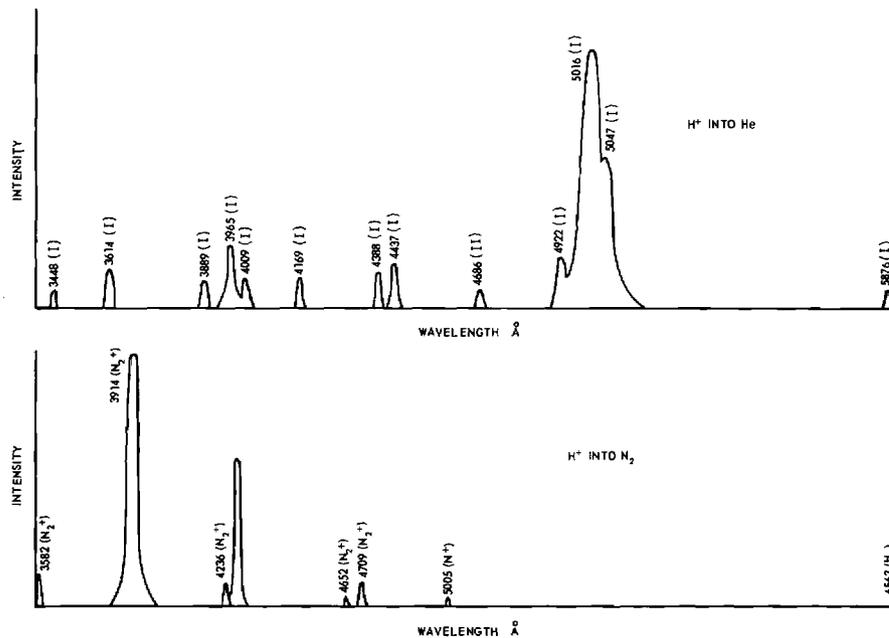


Figure 9. Samples of the spectra produced when protons are incident on helium and nitrogen. Target pressure is about 1μ , and beam current is about $1.5\mu\text{a}$.

Progress Report

where E is the incident proton energy and A and C are constants. Such a formula was also used by Hooper, et al.² to represent their proton ionization data in this energy range and it can be shown that over the relatively small impact velocity range available to our experiment this expression is close to the one predicted by the Born approximation,

$$\sigma = \frac{A \text{ Log } BE}{E}$$

the constants being different in the two expressions.

The curve on Figure 8 shows the data points for electron and proton impact cross sections for the emission of the HeII 4686 Å line coinciding over a range of energies from 0.4 to 0.8 MeV. In view of the normalization procedures used to establish our data in terms of an absolute cross section, and the considerable uncertainties in the absolute values of the electron and proton impact data by Hughes, this observation must be regarded as a coincidence.

As part of our future program, these measurements will be repeated and placed on an absolute basis with improved accuracy. At that time, an attempt will be made to fit the Born approximation to the data with the objective of determining the unknown constants in the expression.

VI. Program for the Remainder of the Contract Year

At the present time the program of construction is almost complete. Certain components of the system must be improved, and in particular the 60° viewing window must be made vacuum tight. A McLeod gauge will be built into the system to allow absolute measurements of pressure, and the detection efficiency of the optical system will be calibrated. It is expected that these improvements will be carried out before the next schedule of experimental observations begins in December.

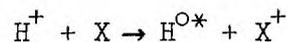
Progress Report

Continuing attention will be given to the problem of pressure measurement, the McLeod gauge being added to the system at the present time is regarded as only a temporary measure. Information from various sources^{7,8} suggests that a good alternative to the McLeod gauge may be found in the diaphragm-type gauge, particularly the unit sold under the name of the "Baratron."⁹ It is hoped that funds will be available in the coming contract year to purchase such a device.

Two one-month periods of experimental measurement are scheduled for the three months remaining in the current contract year. It is intended to restrict attention to the impact of protons on helium targets and to make absolute measurements of the cross section for the emission of a few prominent spectral lines from the helium target. Corrections for cascading effects will be made in the conventional manner,¹⁰ and the results presented as cross sections for formation of excited states in the target.

VII. Program for the Future

It is intended that a substantial proportion of our future effort should be devoted to investigating the formation of fast excited atoms in the incident beam. The process of interest will be charge transfer leaving the fast neutral particle in the excited state.



Special techniques must be adopted for this experiment since the lifetime of the excited state is of the order 10^{-8} sec, and particles with velocities of around 10^9 cm per sec will travel a considerable distance before decaying to the ground state with the emission of the relevant photon. The variation of intensity of emitted radiation with distance from the entry point to the collision chamber may be predicted in terms of the life-

Progress Report

time of the excited state and the velocity of the particle. It is intended to investigate this spatial variation of intensity and to derive cross section for specific processes which leave the fast neutral particle in an excited state.

This technique should also provide a means of finding the cross sections for the excitation of different angular momentum quantum number states. In the case of hydrogen, the 3s-2p, 3p-2s, 3d-2p transitions all emit photons of essentially the same wavelength. However, since the lifetimes of the 3s, 3p, and 3d states are different, the spatial variation of intensity will be the sum of three separate functions, which may in principle be separated by knowledge of the lifetimes and use of curve fitting techniques. In this manner, the cross sections for the formation of atoms in the 3s, 3p, and 3d states may be determined separately. A similar technique has already been employed for this purpose and is published in abstract form,¹⁵ but the work remains in an unsatisfactory state.

A program of measurement of cross sections for the formation of excited states in various target gases by proton impact will also be pursued. Rare gas targets such as neon and argon will be used and the emission functions of one or more spectral lines measured. These functions will be related to the excitation functions of the parent level of the transition, and a comparison made of the shape of the curve with the predicted shape from the Born approximation. Similar measurements will be carried out for molecular nitrogen targets with the additional intention of providing data for the interpretation of upper atmosphere phenomena and light emission seen in accelerator tubes and plasma devices.

15. Dawson, H. R., Doughty, B. M., Hughes, R. H., Bull. Am. Phys. Soc., 10, 170, (1965)

Progress Report

VIII. Publications and Travel

There have been no publications during this year. It is anticipated that the presentation of any of our results will await the completion of the detailed measurements on the excitation of a helium target by proton impact.

Dr. Thomas attended the "Fourth International Conference on the Physics of Electronic and Atomic Collisions," Quebec, Canada, August 2-6, 1965; and the 18th Annual Gaseous Electronics Conference held in Minneapolis, October 20-22, 1965. Periodic visits have been made by Dr. Thomas to Oak Ridge National Laboratory for the purpose of consultation with C. F. Barnett and others.

IX. Personnel

The work described in this report was carried out as part of the A.E.C. Contract No. AT-(40-1)-2591 and has been under the jurisdiction of Dr. Thomas. In June, 1965 Mr. Gary Bent joined the project as a Graduate Assistant on the project. He passed his comprehensive examination in the Fall of 1965 and this project will provide the basis of his Ph.D. thesis problem.

X. Accelerator Usage

Use of the Van de Graaff accelerator is shared with a project to measure ionization cross sections, also under AEC Contract No. AT-(40-1)-2591. It is intended that the facility should be shared equally on a time basis with the two experiments alternating at one month intervals. The excitation experiment has used the machine through the month of October and will also operate during the months of December and February. No difficulty has been experienced in switching the ion beam from one experi-

Progress Report

ment to the other, an operation which involves disconnecting the vacuum system and analyzer magnet. A period of one day suffices to connect and realign the ion beam tube.

XI. Incident Report

There have been no incidents for which a report is required during the performance of the research under this contract in the present reporting period.

~~B-2021~~

ORO-2591-23

EMISSION AND EXCITATION CROSS SECTIONS

PROGRESS REPORT NO. 2

Covering the Period

December 1, 1965 to November 30, 1966

By E. W. Thomas

G. D. Bent

J. L. Edwards



Report No. ORO-2591-23

Contract No. AT-(40-1)-2591

U.S. ATOMIC ENERGY COMMISSION

OAK RIDGE, TENNESSEE

1 December



School of Physics
GEORGIA INSTITUTE OF TECHNOLOGY
Atlanta, Georgia

ERRATA

"Emission and Excitation Cross Sections "

AEC Report No. ORO-2591-22

Page 25 Equation (6), has a negative sign missing. Equation should read:-

$$Q_j = Q_{jk} \frac{\sum_k A_{jk}}{A_{jk}} - \sum_i Q_{ij} \quad (6)$$

Page 32 Line 19. Replace ($Q_{ij} \times E$) by ($Q_i \times E$)

Page 54 Line 3 Replace $X^+ + Ar = X^+ + Ar^{+1} + e$
by $X^+ + Ar = X^0 + Ar^{+1}$

Line 5 Replace $X^+ + Ar = X^0 + Ar^{+1}$
by $X^+ + Ar = X^+ + Ar^{+1} + e$

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Contract No. AT-(40-1)-2591

U.S. Atomic Energy Commission

Oak Ridge, Tennessee

30 November 1966

Progress Report

TABLE OF CONTENTS

	Page
<u>List of Figures</u>	iii
I <u>Title</u>	1
II <u>Introduction</u>	1
III <u>Objective and Method</u>	2
IV <u>Summary of Progress Made During This Reporting Period</u>	7
V <u>Experimental Apparatus and Procedures</u>	10
a The Source of the Incident Beam.....	11
b The Collision Chamber.....	13
c Gas Handling and Pressure Measurement.....	16
d Optical Detections Systems.....	17
e Calibration of the Optical Detection System.....	18
VI <u>Experimental Results</u>	22
a Excitation of a Helium Target by Protons.....	24
HeI $1^1S \rightarrow n^1S$ and $1^1S \rightarrow n^1D$	27
HeI $1^1S \rightarrow n^1P$	35
HeII	38
b Excitation of a Helium Target by H^+ , H_2^+ and H_3^+ ...	39
c Excitation of a Nitrogen Target by Protons.....	43
d Excitation of an Oxygen Target by Protons.....	50
e Excitation of an Argon Target by Protons.....	53
VII <u>Program for the Remainder of the Contract Year</u>	54
VIII <u>Program for the Future</u>	55
IX <u>Publications and Travel</u>	57
X <u>Personnel</u>	58
XI <u>Accelerator Usage</u>	59
XII <u>Incident Report</u>	60
<u>Appendix I</u>	60

Progress Report

LIST OF FIGURES

<u>Figure</u>	<u>Page</u>
1. Schematic diagram of the experimental arrangement.....	15
2. Cross sections for the formation of n^1S excited states of neutral helium by the impact of protons and deuterons....	28
3. Comparison of the cross sections for the formation of the 4^1S excited state of neutral helium by the impact of protons, deuterons, and electrons.....	29
4. Cross sections for the formation of n^1D excited states of neutral helium by the impact of protons and deuterons.....	30
5. Comparison of the cross sections for the formation of the 4^1D excited state of neutral helium by the impact of protons, deuterons, and electrons.....	31
6. The product of cross section and energy on an arbitrary scale, as a function of energy, for selected levels.....	33
7. Cross sections for the formation of n^1P excited states of neutral helium by the impact of protons and deuterons.....	36
8. Comparison of the cross sections for the formation of the 3^1P excited state of neutral helium by the impact of protons, deuterons, and electrons with theoretical predictions.....	37
9. Cross section for the emission of the HeII 4686 \AA ($n = 4 \rightarrow n = 3$) spectral line, induced by proton impact on a neutral helium target.....	40
10. Cross sections for the formation of the 4^1S excited state of neutral helium by the impact of H^+ , H_2^+ , H_3^+ ions at equal velocities.....	42
11. Cross section for the $v' = 0$ progression emission in the N_2^+ first negative system, induced by the impact of protons on a molecular nitrogen target.....	45
12. Population cross section for the $v' = 0$ level of the $B^2 \Sigma_u^+$ state of N_2^+ , induced by proton impact on a molecular nitrogen target.....	46
13. Cross sections for the emission of the NII 5005 and 5680 \AA lines induced by proton impact on a molecular nitrogen target	49

Progress Report

<u>Figure</u>		<u>Page</u>
14.	Cross sections for the population of the $v' = 0$ and $v' = 1$ levels of the $b^4\Sigma_g^+$ state of O_2^+ , and for the emission of the OII 4415 and 4417 Å lines, induced by proton impact on a molecular oxygen target.....	52
15.	Schematic diagram of Baratron sensing head.....	63
16.	Block diagram of vacuum system test rig.....	63
17.	Comparison of McLeod and Baratron gauge readings for helium.....	67
18.	Comparison of McLeod and Baratron gauge readings for argon.....	68
19.	Comparison of McLeod and Baratron gauge readings for nitrogen.....	69
20.	Comparison of McLeod and Baratron gauge readings for oxygen.....	70

Progress Report No. 2, Project No. B-2021

I. Title

Emission and Excitation Cross Sections

II. Introduction

This report summarizes the work performed on excitation phenomena as part of contract AT-(40-1)-2591 for the U.S. Atomic Energy Commission. The work was initiated on March 1, 1965, as a result of Modification No. 7, increasing the scope and financing of the existing contract. The present report covers the period 1 December 1965 to 30 November 1966 which corresponds to the first 9 months of the 12 month period covered by Modification No. 8 to this contract, plus the final three months of the preceding contract period.

Prior to the period covered by this report, the contract was administered through the Engineering Experiment Station at the Georgia Institute of Technology. The previous progress report entitled "Emission and Excitation Cross Sections", was issued through that division of the Institute.¹

A program for the measurement of ionization cross sections which was the subject of the original contract and subsequent Modifications 1 through 8 has been continued and is reported separately.²

-
1. E. W. Thomas and G. D. Bent, "Emission and Excitation Cross Sections" Progress Report No. 1. 1 December 1965, Project B176-002, Georgia Institute of Technology. U.S. Atomic Energy Commission Contract No. AT-(40-1)-2591.
 2. D. W. Martin, L. J. Puckett, and G. O. Taylor. "Ionization and Charge Transfer Cross Sections", Progress Report No. 19. 30 November 1966. U.S. Atomic Energy Commission Report Number ORO-2591-19.

Progress Report

III. Objective and Method

The objective of this research is the measurement of cross sections for production of particles in excited states as the result of the impact of protons on various gaseous targets under single collision conditions. The energy of the incident particles ranges from 0.15 to 1.0 MeV. A survey has been carried out during the present reporting period³ which indicates that the present program of research is still the only large scale effort directed toward the investigation of excitation processes at a sufficiently high energy of impact that theoretical predictions are expected to be accurate. It is of the utmost importance that the approximations made in theoretical predictions should be tested at every available opportunity, in order to establish the validity of calculations in situations where experimental verification is at the present time impossible for technical reasons. A prime objective in the present program is the acquisition of a body of data which can be used (a) for comparison with accurate quantum mechanical calculations where these are available, or (b) in the more complex situations, to indicate systematics in behavior useful in predicting basic characteristics and in assisting with the formulation of a phenomenological basis for semi-empirical and statistical theories. A second objective is the provision of data which is of direct value for design and diagnostic applications in thermonuclear, upper atmospheric, and other practical situations.

3. E. W. Thomas, Technical Report No. 1. Project No. B-2021, School of Physics, Georgia Institute of Technology, 30 October 1966. Contract No. AT-(40-1)-2591 U.S. Atomic Energy Commission. U.S. Atomic Energy Commission Report Number ORO-2591-22.

Progress Report

In general, the atomic and molecular processes which occur when fast atoms or ions collide with the molecules of the target gas, involve the rearrangement of the electrons associated with the two systems. We may use the term "charge rearrangement" to refer to cases where electrons are ejected from either of the colliding systems or transferred between them. Charge rearrangement may be investigated by observing the change of the ionic composition of the fast beam and the target region. In addition to the gross removal or transfer of charge, there is a probability that one or more of the electrons remaining bound to the nucleus after the collision may be in an excited state. Quantitative measurement of the photon flux emitted as a result of subsequent decay to the ground state allows the determination of the cross section for the formation of excited states. Clearly such a technique cannot be used for investigating the formation of metastable or other long-lived states, and these are not considered in the present work.

The source of the energetic protons is a 1 MeV Van de Graaff positive ion accelerator, which is equipped with a beam analyzing and stabilizing system. The ion beam is collimated by a series of small apertures, traverses a collision chamber containing the target gas, and is monitored on a Faraday cup. A quartz window situated in the wall of the chamber allows spectral analysis and detection of emission from the collision region. The chamber dimensions and gas pressure are such that the target is "thin" in the sense that only a small fraction of the incident particles undergo any collisions at all. Also the path traveled by the photons before leaving the gas-filled collision chamber has been arranged to be short to reduce absorption.

Progress Report

In general separate techniques must be utilized for measuring the formation of excited states in the static target and in the fast beam. In the former case the excited target atom will acquire only a small momentum as a result of the collision and will decay to the ground state with the emission of photons at essentially the same point as it was excited. However in the case of the fast incident particles, the velocities used in the present work are so high that the excited particle will move an appreciable distance during the finite lifetime of the excited state. Consequently emission from the fast particle will vary with the position of observation in the collision chamber. In either case the measurement technique consists of determining the rate of photon emission per unit length of the ion beam path in the collision chamber, per unit incident beam flux, per unit target density. This apparent cross section for the emission of a photon of a particular wavelength will be dependent on the position of observation in the case of the excited fast particles, but in the case of target molecules will be invariant with respect to position along the beam.

For the measurement of the formation of excited target particles, the photons emitted from a definite fixed part of the ion beam path in the chamber are focused onto the entrance slit of a grating spectrometer, by a simple lens. A low noise photomultiplier at the exit slit serves to detect the photons in the dispersed spectrum. Enhancement of the signal-to-noise ratio is obtained by chopping the light signal with a vibrating reed before it enters the spectrometer and applying a narrow band, phase sensitive detection system to the output of the photomultiplier. The determination of the detection efficiency of the system is made quite simply by substituting a source of emission of known output in place of

Progress Report

the collision experiment. The ratio of the emissions of the experiment and the standard will be equal to the ratio of observed signals. If E_λ is the emissive power of the standard at the wavelength of interest λ , S_λ the measured signal, S'_λ the signal from the excitation experiment, and n and N respectively the particle density of the target and particle flux of the incident beam, the cross section is given by:

$$Q_\lambda = \frac{S'_\lambda}{S_\lambda} \frac{E_\lambda}{n N} \quad (1)$$

Simple considerations show that when using a substitution technique where the standard is of the same geometrical shape as the source of experimental emission, then knowledge of the actual length of particle beam path observed by the detection equipment, and of the solid angle from which light is collected, is unimportant. The emission cross section is essentially the cross section for a single depopulating transition. Any one excited state will in general have a number of possible paths by which it may decay radiatively, and, in addition, there will be a series of cascade populations of the level due to transitions from higher excited states. In principle if the emission functions for all the radiative populating and depopulating transitions associated with a particular level are known, then the cross section for the formation of the excited state may be determined. If cascade is neglected, and the transition probability for the decay $j \rightarrow k$ corresponding to the wavelength λ , is A_{jk} , the sum of all decays to states k lower than j is $\sum_k A_{jk}$, then the cross section for the excitation of the level j is given simply by

$$Q_j = Q_{jk} \frac{\sum_k A_{jk}}{A_{jk}} \quad (2)$$

Progress Report

Knowledge of the relevant transition probabilities, and measurement of a single transition may therefore lead directly to the collisional excitation cross section. Transition probabilities are only known for the atomic hydrogen and helium cases, and therefore the techniques used for overcoming the problems of branching and cascade in a particular experimental investigation will be indicated in the discussion of specific results in section VI.

Simple consideration of the lifetime τ of the excited state and the velocity v of the fast particle show that the emission from excited fast particles will build up exponentially toward some asymptotic value as the beam traversed the chamber. If one again defines the cross section for photon emission in the same way as for target excitation this quantity will now vary with penetration x through the chamber. Neglecting for the moment cascade transitions, if Q_j is the cross section for the collisional excitation of the state j , A_{jk} is the transition probability for the $j \rightarrow k$ decay, and $\sum_k A_{jk}$ is the sum of all the radiative decay transition probabilities out of the level j to all levels k , then the emission cross section will vary as

$$Q_{jk} = Q_j \frac{A_{jk}}{\sum_k A_{jk}} \left(1 - \exp - \frac{x}{v\tau}\right) \quad (3)$$

This is to be compared with equation (2). Measurement of the emission cross section at one or more values of x , with knowledge of the transition probabilities and lifetime allows the evaluation of Q_j . The problem of assessing cascade contributions may be severe and will have to be treated separately for each specific case. This new phase of the experiment requires

Progress Report

the extension of the length of the collision chamber and the provision of long side windows, allowing the investigation of emission from all points along the track of the fast particle in the chamber. Detection will be achieved using a photomultiplier fitted with an interference filter to isolate the wavelength of interest. Pulse counting techniques will be used, and the effect of the background thermal noise in the photomultiplier reduced by cooling the photocathode. It is our intention to apply this technique first to the formation of excited hydrogen atoms by charge transfer, as protons traverse various target gases. This technique will allow us to separately measure the cross sections for the formation of the 3s, 3p, 3d states of hydrogen, since the decay of these states to the 2s and 2p states will involve the emission of photons of essentially the same wavelength, but by decays of different lifetimes. The measured intensity of the Balmer Alpha line as a function of penetration x is then expected to consist of three exponentially increasing functions. In principle the separate curves may be identified and the cross section for the formation of these three states separately determined.

IV. Summary of Progress Made During This Reporting Period

The objectives of the work proposed for the period covered by this report were twofold. Firstly, the equipment designed to investigate excitation of the target system, which was constructed in the previous reporting period¹, was to be utilized for a widely ranging series of measurements. Secondly, new equipment was to be designed to allow investigation of emission from fast particles traversing the collision chamber. Progress has fallen a little behind schedule, due primarily to our having undertaken a small number of subsidiary investigations which

Progress Report

seemed to us to be of importance but which were not provided for in the original proposal.

In this section of the report we will summarize briefly the progress that has been achieved, so providing an introduction to the detailed discussions of the various facets of our operation which are to be found in subsequent sections.

The equipment designed for the investigation of the formation of excited states in the gaseous target has continued to function well and has been subjected to no important modification. Certain problems associated with the calibration of the detection sensitivity of the system have come to notice recently and are discussed more fully in Section V f. A considerable quantity of data has been obtained with this equipment, and is discussed below. The equipment for measuring the formation of excited states in the fast particle beam has been constructed and at present preliminary tests are in progress. Due to various delays no data have been obtained from this device but it is hoped that successful operation will be achieved before the end of the contract year. The equipment is described more fully in Section V e.

Our investigation of the formation of excited states of helium by the impact of protons on a helium target has been more extensive than originally envisioned. We believe that since this is one of the few cases where exact quantum mechanical calculations are likely to be possible, there is justification for building up a considerable body of data in one self-consistent set. Cross sections have been measured for four each of the $1S$, $1P$ and $1D$ levels of neutral helium, providing a body of data over a range of principal and angular momentum quantum

Progress Report

numbers. Useful comparisons have been made between theoretical predictions and our observations, confirming for the first time the expected high energy behavior of excitation cross sections. Measurements have also been made on a single line emitted from the helium ion. This data is discussed in Section VI a.

Two subsidiary investigations were carried out using helium targets with different incident ions. It is generally expected that deuterons will give the same cross sections for the excitation of a target as protons at the same velocity. Assuming this to be the case, we have utilized beams of deuterons to extend downward the effective velocity range of our experiment for a few cases to allow comparison with existing low energy data. We also made a brief comparison of the cross sections for excitation of the helium target by H^+ , H_2^+ , and H_3^+ ions. These subsidiary investigations are discussed in Sections VI a and VI b.

Our next investigations were on the excitation of a nitrogen target by proton impact. Cross sections for the emission of some nine spectral lines in the first negative system of N_2^+ were measured, allowing the calculation of the cross sections for the formation of N_2^+ ions in the two lowest vibrational levels of the $B^2\Sigma_u$ excited state. The apparent rotational temperature of the N_2^+ excited state has also been determined. The cross section of a single NI (neutral atomic nitrogen) spectral line was also determined. Beams of deuterons were again used to extend the effective velocity range of our data in certain cases. The results of our investigations with a nitrogen target are presented in Section VI c.

Progress Report

Investigation of the formation of excited states in an oxygen target has also commenced. Excited states of molecular ions, neutral atoms and atomic ions are found and have been investigated. Preliminary assessments of the data are presented in section VI d.

Surveys have been carried out on the emission spectrum obtained from argon and hydrogen targets. A short program of cross section measurements for these gases will be undertaken before the end of the contract year, as mentioned in our proposal.

The modifications to the existing system to allow measurement of the formation of excited states in the fast particle system, have been completed. Preliminary tests are now being carried out and it is expected that preliminary measurements will be attained before the end of the contract year, using nitrogen as the target.

A continuing interest has been maintained in the problem of pressure measurement, and we have acquired, tested and brought into use a capacitance manometer type of pressure gauge. A detailed report on the procedures utilized to confirm the gauge calibration, incidentally providing additional information on the operating characteristics of the McLeod gauge, is included as an appendix to this report.

During this work a continuing record of published data on excitation and emission functions has been maintained and a listing of sources of data, with basic information as to the processes investigated and energy ranges covered has been prepared as a separate document.³

V. Experimental Apparatus and Procedures

The basic experimental system for the measurement of target excitation remains little changed from our previous report,¹ but a description will be repeated here for completeness, with particular emphasis on the

Progress Report

improvements that have been made. The modifications being made to allow the investigation of the excitation of the fast particle will also be discussed. Separate descriptions will also be given of the optical calibration techniques and the procedures utilized for determining excitation cross sections from the measured emission functions. A schematic diagram of the original apparatus used for measuring the excitation of the target is shown as fig. 1. The new equipment for the second phase of the experiment is not shown but is described in the text.

a. The Source of the Incident Beam

The beam of fast ions in the energy range 0.15 to 1.0 MeV is provided by a Van de Graaff accelerator, type JN of the High Voltage Engineering Corporation. The accelerator is mounted vertically and the beam is deflected into a horizontal trajectory by a large electromagnet, which also serves to mass analyze the ions. The proton energy is stabilized by electronic regulation of the accelerator voltage to maintain equal currents on the edges of two slits placed symmetrically about the ion beam path, a method which demands a constant deflection in the regulated magnetic field. The nominal energy spread of the beam is ± 2 Kev at 1 MeV, the particle energy being determined by the value of the magnetic field and measured by measuring that field. For this purpose, we use a Harvey Wells model G-501 Nuclear Magnetic Resonance precision gaussmeter, which, as used in our experiments, has relative and absolute accuracies of one part in 10^3 . The deflection geometry has been calibrated empirically by measuring the magnetic field corresponding to the 1.019 MeV threshold of the nuclear reaction $\text{He}^3(p,n)\text{He}^3$, using a tritium-zirconium target.

As shown in Figure 1, the ion beam enters the collision chamber

Progress Report

through three circular knife edge apertures, mounted co-axially to form a collimator. The first two apertures are of $1/16$ " diameter, mounted at opposite ends of a 12 cm tube, and, therefore, are electrically connected. The third aperture is $3/32$ " in diameter and mounted on a tube which projects through the end plate of the collision chamber. The tube carrying the first two apertures is mounted inside the tube carrying the third slit and is insulated from it. The diameters and separations of the apertures are arranged so that the first two provide the collimation of the beam, and no ions should impinge on the third. A positive voltage may be applied to the collimating apertures to suppress secondary electrons and to prevent them from passing into the collision chamber. The third aperture defines the physical limit of the gas cell, and it is provided with a long snout of $1/4$ " diameter, to inhibit the pumping of gas into the collimator. A pressure differential of a factor of one hundred can be established between the collision chamber and the collimator region. The collimator lies in a chamber that can be differentially pumped with respect to the rest of the system, and is constructed with a large number of pumping holes in the tubes so that any gas that does pass through the final slit is rapidly removed from the ion beam path.

A screw thread is cut into the outer wall of the tube carrying the third slit. This allows the collimator assembly to be screwed through the end wall of the collision chamber to vary the separation of the entrance point to the collision chamber and the observation region.

The differential pumping chamber is constructed from type 304 stainless steel, and the collimator assembly is of brass, teflon being used to insulate the two basic components. Good alignment of the three apertures was obtained by constructing the collimator as a single unit and cutting the apertures with the assembled device mounted on a lathe.

Progress Report

During the present reporting period considerable effort has been expended towards improving the operation and versatility of the accelerator. In the first place the proton beam currents available in the collision chamber after passage through the collimation system have been raised from one to eight microamps under normal conditions, with beams of some 15 microamps immediately after a new source is installed. Deuterium has also been used as a source gas and will in general yield a beam of deuterons of approximately one half the current observed with protons. Beams of H_2^+ and H_3^+ have also been produced at currents up to 4 microamps and have been used for experimental measurements. Recently neon gas has been installed in the source and excellent operation is observed with total ion beam current only a little less than with hydrogen in the source. Since the analyzer magnets should be able to handle beams of Ne^+ at energies up to 1 MeV, a short survey will soon be initiated to investigate whether this beam might provide some interesting excitation data.

b. The Collision Chamber

The collision chamber was constructed from non-magnetic type 304 stainless steel to cut down stray fields. It is intended at some later date to investigate the polarization of the light emitted from this experiment, and it is known that small stray fields can appreciably disturb the relative populations of the magnetic quantum number states of the excited atom, which govern the polarization fraction.

One end of the chamber is blanked off by a plate carrying the ion beam current monitoring assembly, and the opposite end is closed by the collimator tube. A port is provided beneath the beam monitor which connects to a 2" oil diffusion pump used to evacuate the chamber. The usual precautions of using a refrigerated trap to prevent oil streaming into the

Progress Report

collision chamber have been observed. A baffle valve is provided to isolate the pump from the system, and a direct line to the mechanical pump may be used for roughing out the chamber.

After passing through the region of the collision chamber from which optical measurements are made, the fast particle beam passes into a deep Faraday Cage structure where it may be monitored. As shown in figure 1, the Cage is formed from two plates which taper together to form a blind end. The ion beam impinges on one plate; any secondary electrons are drawn to the second plate by a positive potential, and are therefore prevented from returning towards the observation region. The total current to the two plates is equal to the ion beam current and is monitored by a Keithley Model 410 Electrometer which drives one channel of a two-channel pen recorder. Additional safeguards against loss of ions and secondary electrons from the cage include giving the collection plates overlapping walls to enclose completely the cage region, and providing an aperture plate in front of the system to which a negative potential may be applied to return electrons to the plates.

A port on top of the chamber is used for both pressure measuring devices and the introduction of target gas.

The original collision chamber, shown in figure 1, is provided with two long side ports to allow the use of windows to permit optical observation. For target excitation measurements, the long side window is blanked off, and only the opposite small circular quartz window used. All the present measurements were carried out using optical detection systems viewing the collision region at an angle of 90° to the ion beam path. However a second window permitting a 60° angle is also available, and will be particularly valuable whenever emission from target and fast particle

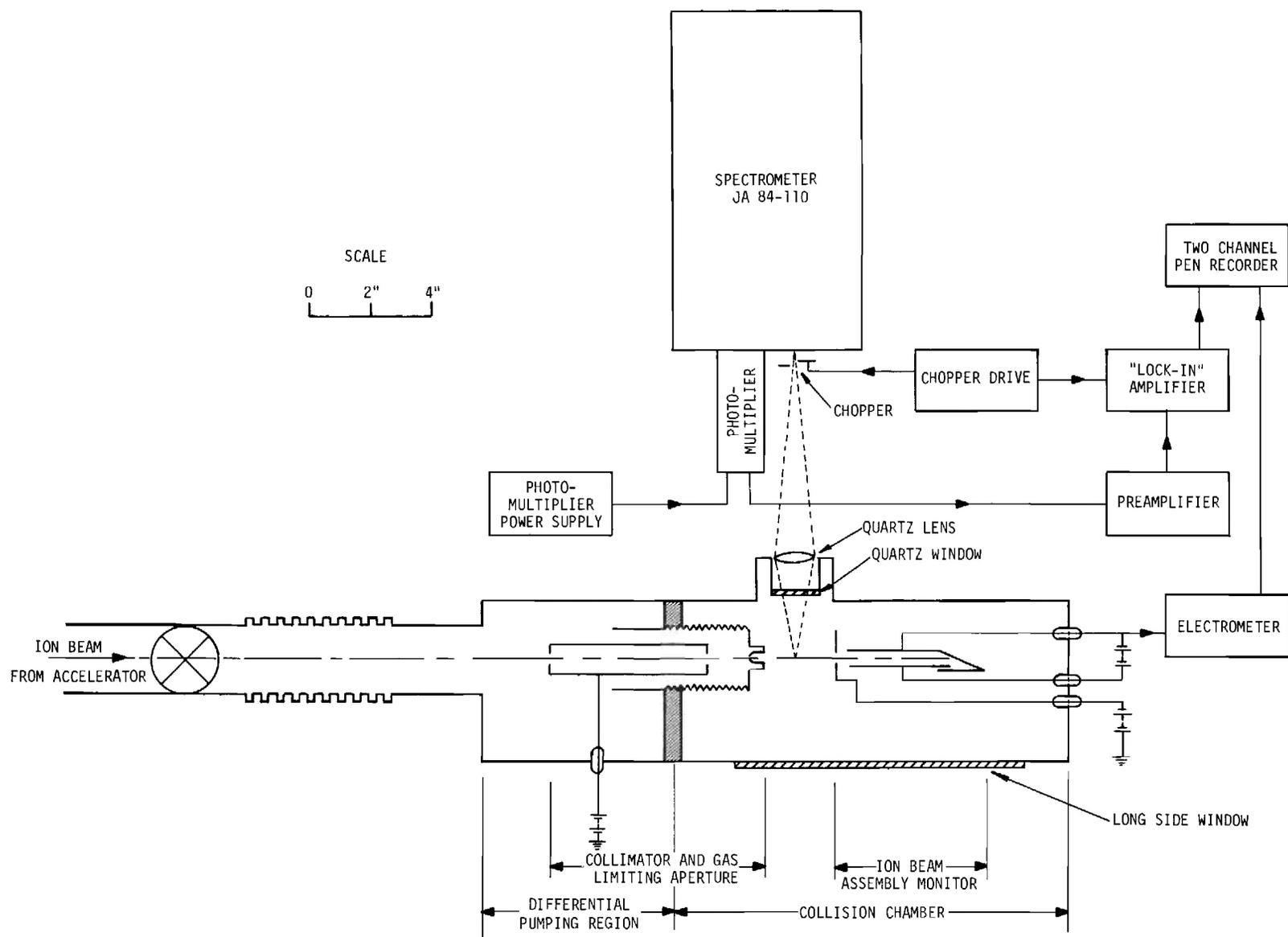


Figure 1. Schematic Diagram of the Experimental Arrangement.

Progress Report

have to be separated utilizing the Doppler shift of the radiation from the latter system. A considerable amount of ingenuity was used in trying to keep the diameter of the collision chamber as small as possible, with the object of reducing the possibility of photoabsorption. Resonance photons emitted by the decay of excited target systems will be readily reabsorbed. Such absorption influences the observed emission from any state which is optically coupled to the ground state.

The second phase of the program, designed to investigate excitation of the fast particle, requires the measurement of the emission from the ion beam path at a large number of points along a relatively long path through the target gas. An extension chamber has been built to provide the required length, and a long side window provided in both chambers allows the scanning of the emission over the whole length of the combined chamber.

c. Gas Handling and Pressure Measurement

The gas samples used in the present work were obtained from Matheson in high pressure cylinders and are stated to be 99.9% pure. A cold trap refrigerated with dry ice and acetone was utilized on the gas feed line to remove any condensible vapors before the gas entered the collision chamber through a mechanical leak.

The collision chamber was provided with Pirani and ionization gauges to give continuous estimates of pressure. We commenced the present program using a McLeod gauge for making the accurate pressure measurements needed in cross section determinations. Since the work involved only helium as a target gas the operation of the gauge was not influenced appreciably by the "mercury pumping" error which is described in Appendix 1. However the gauge was tedious to use, and sticking of mercury in the columns was

Progress Report

a considerable problem. We purchased a capacitance manometer gauge⁴ and carried out a series of tests to insure its linearity and accuracy. A complete description of the new gauge, its operation, and the checking procedures used, is given in Appendix 1. Having confirmed the successful operation of the capacitance manometer, it was utilized in all subsequent work, particularly that involving the heavier gases, oxygen and nitrogen.

d. Optical Detection Systems

In the work on emission from the target system, the light is observed through the small circular window, and focused by a quartz lens onto the entrance slit of a 1/2 meter Ebert Fastie scanning monochromator. Detection is by a photomultiplier, located at the exit slit of the instrument, and the limits of the sensitivity of the available tubes define the effective range of operation of our equipment as 2000 to 6500 Å. Although the tubes have been selected for their low noise characteristics, additional enhancement of the signal to noise ratio is achieved by chopping the light with a vibrating reed before it enters the spectrometer, utilizing a phase sensitive amplifier, and identifying the required signal by its specific frequency and phase. The output from the optical detection system is displayed on a two pen recorder, parallel to a trace of the fast ion beam current.

The original choice of D.C. detection systems for the optical analysis was dictated by the necessity of scanning through a whole spectrum to provide a broad analysis of the types of emitting systems that were important. The newer equipment, designed to measure emission from the fast beam, will not require that capability, and will consist of a photomultiplier, provided with suitable wavelength defining interference filters and operated

4. The "Baratron", M.K.S. Instruments, Burlington, Mass.

Progress Report

in a pulse counting mode. Background noise will be reduced by utilizing a thermoelectric cooling system, and preliminary tests indicate that this will reduce the noise count rate to about two pulses per second. Final noise elimination will be accomplished by counting signal and signal plus noise separately and taking the difference. Preliminary investigations of the Balmer Alpha emission using the spectrometer, which essentially allows the measurement of the emission at one penetration depth only, has given the basic design criteria of required pass bands of the interference filter, and of detection sensitivity. The whole photomultiplier and filter assembly are located on an accurately machined track parallel to the collision chamber. A series of locating points are provided allowing the detector to be placed at any one of 75 different positions. The use of a series of fixed positions rather than a continuous scan will insure that positional settings are reproducible.

e. Calibration of the Optical Detection System

The problem of calibrating the detection sensitivity of the optical equipment continues to be the greatest single hindrance and source of trouble to the successful pursuit of this program. In previous reports we have repeatedly contended that the difficulties and uncertainties in such procedures must be the explanation of the wide discrepancies observed between data obtained by different research groups. The requirement is for a standard of photon emission with a reasonably wide spectral range and an accurately known emissive power or photon emission rate. The black body radiator would seem to be the only source available. The electron synchrotron radiation, often considered to be an alternative, would seem to be good in principle, but the experimental inaccuracies involved in

Progress Report

establishing the absolute value of the emission appear considerable.⁵ The black body is inconvenient for the average laboratory to use very frequently, and a transfer standard in the form of a tungsten strip filament lamp, of known geometrical construction is utilized. The emissive power per unit area of filament, per Å bandwidth, per unit solid angle, per unit time, is known as a function of wavelength and filament temperature from the work of De Vos.⁶ Each time the calibration is carried out the filament temperature must be measured and the optical alignment of the system confirmed. At the present time even with considerable care being taken, the measured values of excitation cross sections obtained by different workers are varying by up to 100%, which is far outside the quoted error brackets.

Our present calibration procedure consists of substituting for the experimental source of emission, the standard strip filament lamp. Our major identifiable source of error arises in the setting of the lamp filament temperature with the use of an optical pyrometer. Fortunately the emission spectrum of the lamp does not change its shape markedly with a change in temperature of the order 10 °C, and the relative accuracy of calibrations made at different wavelengths should be high. However the absolute values may be in error by a considerable amount.

It has recently been suggested⁷ that a further source of error can be found at certain wavelengths due to scattered light within the

5. K. Codling and R. P. Madden, *J. Applied Physics* 36, 380, 1965.

6. J. C. de Vos, *Physica* 20, 690, 1954.

7. R. J. Anderson, Private Communication.

Progress Report

spectrometer itself. Quite obviously this is not a new problem, but it would appear that its significance has been ignored when carrying out calibrations using black body sources particularly in the near ultraviolet region of the spectrum. A spectrometer set to transmit a bandpass of 16 \AA centered on 3500 \AA will transmit only one part in 10^6 of the total photon emission from the standard lamp integrated over all wavelengths up to 6500 \AA , which is the limit of sensitivity of the detector employed. It is clear that if a few parts in a million of the incident photon flux from the standard lamp are able to pass to the exit slit of the spectrometer without being dispersed by the grating, then a very considerable error could result. We have made a test for this effect and have indeed found it to be very important below about 4000 \AA . Various techniques can be envisaged for assessing this problem using broad band filters to cut out the visible portion of the light, which presumably is making the greatest contribution to any scattered intensity observed at the exit slit. The easiest conceptually and practically is to take a piece of ordinary clear glass, which does not transmit below 3900 \AA , place it in front of the standard lamp and observe the change in the transmitted intensity. The change in intensity is due to the true ultraviolet signal at that wavelength setting, which is not transmitted by the glass. Any residual signal with the glass in place is entirely due to visible light presumably arriving at the detector by some scattering path. A preliminary investigation of this problem shows that it is indeed very serious, producing errors which range from 100% at 3900 \AA to 600% at 3600 \AA . It would appear however that at wavelengths above 4000 \AA the problem is negligible.

Progress Report

Two basic points must be emphasized about this newly discovered problem. Firstly it seems to have an appreciable influence only at wavelengths below 4000 \AA where the light intensity from the lamp is exceedingly low. At higher wavelengths the error may remain of the same absolute magnitude, but relative to the much increased intensity of the direct transmission through the spectrometer, the problem is negligible. Furthermore although the scattered light is a fault of the spectrometer's construction or operation, the broad band nature of the lamp's emission, with a massive peak in the red but with very low intensities in the blue and ultraviolet, is really the main cause of the serious errors. A scattered light intensity of one part per million incident photons is not excessive, but is enough to seriously effect the calibration at low wavelengths. It is not expected that the scattered light problem will appreciably influence measurements made on the emission from the experimental region, since these are line emissions and the integrated intensity of the whole spectrum will be many orders of magnitude less than that of the standard lamp. Therefore the scattered light problem certainly does not influence a determination of relative variation of cross section with energy.

This latest problem in calibration was identified only during the week preceding the writing of this report and no great attention has yet been paid to eliminating it. An attempt has been made to measure the influence of the effect on our measured cross sections, and where the corrections could be accurately determined, these have been applied to the data in this report. It is likely that a permanent solution can be found through the use of filters of known or calibrated transmission to cut down the intensity of visible light incident on the spectrometer when making calibrations in the ultraviolet. It is also our opinion that some

Progress Report

additional baffling in the optical spectrometer might assist in removing the problem. The arrangement of components in the Ebert Fastie Spectrometer is such that scattered light cannot be suppressed very effectively with baffles. It is very likely that all workers using these instruments have produced erroneous data due to this problem, and specific cases where we believe this has occurred will be mentioned under the discussion of data.

VI. Experimental Results

We will present data on the formation of excited states in helium, nitrogen and oxygen by the impact of protons, with additional limited investigations of the excitation of the same targets by the impact of deuterons, H_2^+ , and H_3^+ ions. It is generally expected that cross sections for excitation and ionization by incident protons and deuterons at the same velocity of impact should be very similar. Existing data on excitation⁸ indicate that the two projectiles produce the same cross sections within experimental error, and our own investigations confirm this, no proton and deuteron data at the same velocity of impact differing by more than 5%. We have used deuterons to extend the effective velocity range of our experiment down to that velocity which corresponds to a proton at 75 KeV energy. The extension of our range of impact energies facilitates greatly a comparison with existing published data at low energies, and also extends our data into the region of the cross section maximum where a considerable divergence between theory and experiment might be expected.

8. D. Pretzer, et al. Proc. 3rd Conf. on Physics of Electronic and Atomic Collisions. (Published North Holland 1964) page 618.

Progress Report

At the present time we have not completed an accurate estimate of the limits of accuracy of this work. In general the random scatter in our measurements is about 5%, and we would anticipate that the systematic errors in absolute calibration, pressure measurement, etc., will be about 15%. Thus a figure of 20% would give a good indication of the limits of accuracy of this work. In general relative measurements will be better than this, and the relative magnitudes of two cross sections for exciting different levels in the same target will be less than 10%, due to the elimination of certain systematic errors in pressure measurement and absolute sensitivity calibration.

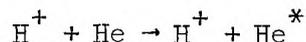
It was not the intention in this work to measure the polarization of the emitted radiation, but since this will affect the angular distribution of the emission and hence the relation of the present measurements at one specific angle to the total cross section for emission summed over 4π steradians, it was deemed advisable to make an estimate of the polarization factor. The relative intensities of the radiation polarized parallel and perpendicular to the ion beam path were obtained using our basic experimental arrangement with a polaroid filter inserted in the light path. The Ebert Fastie type of spectrometer utilized in this work is known to have different transmission factors for radiation polarized parallel and perpendicular to the axis of the entrance slit, and before attempting to measure the polarization of the emission from the experiment, this factor was evaluated using a source of unpolarized radiation. In none of the cases investigated did the polarization factor exceed 15%. A simple calculation indicates that for measurements made at 90° to the ion beam path, this order of polarization factor will introduce an error of no more than

Progress Report

7% in our total emission cross section. Although the measurements presented in this report are strictly speaking the emission cross sections measured at 90° to the ion beam path, the correction for polarization would introduce a change of no more than 7% in the worst case.

a. Excitation of a Helium Target by Protons

In the case of a helium target, the observed radiation results primarily from direct excitation of the target without associated charge rearrangement.



The emission of the HeII ion lines is also weakly observed.

The energy range covered by this work is sufficiently high that, drawing on experience with ionization cross sections,⁹ the general predictions of the first Born approximation are expected to be valid. The present work is the first comprehensive series of measurements of excitation cross sections in ion-atom collisions where the Born approximation predictions may be tested.

The helium ground state is 1^1S and therefore the excitation of the 1^1P states will be an optically allowed transition in which the cross section function would be of the form:

$$\sigma = \frac{AZ'^2}{E/M} \log (BE/M) \quad (4)$$

A involves the matrix element for the dipole transition and will vary with the level excited. B involves the energy required to excite the level and,

9. J. W. Hooper, et al., Phys. Rev. 125, 2000 (1962).

Progress Report

since all the 1P states considered are very close, should be almost invariant in this work. Z' is the effective charge and M the molecular weight of the incident fast particle, both of which will be unity for incident protons. The excitation of the 1S and 1D states proceeds by an optically forbidden transition and the cross section should be of the form

$$\sigma = \frac{CZ'^2}{E/M} \quad (5)$$

where C involves the quadrupole matrix element of the transition which varies with the excited level. The difference between the behavior of optically allowed and forbidden transitions will be clearly demonstrated by our data.

In section III we related the measurements of the cross section for the emission of a spectral line to the cross section for the formation of the excited upper level. If j is the excited state of interest, $j \rightarrow k$ is the radiative transition which is measured, then

$$Q_j = Q_{jk} \frac{\sum_k A_{jk}}{A_{jk}}$$

If cascade from higher states i is important then an additional term is introduced.

$$Q_j = Q_{jk} \frac{\sum_k A_{jk}}{A_{jk}} + \sum_i Q_{ij} \quad (6)$$

It is not necessary to measure Q_{ij} directly since if a transition to some other state m is measured (Q_{im}) and the relevant transition probabilities

Progress Report

are known then;

$$Q_j = Q_{jk} \frac{\sum_k A_{jk}}{A_{jk}} - \sum_i \frac{Q_{im}}{A_{im}} A_{ij} \quad (7)$$

In the case of helium it is fortunate that the transition probabilities are known accurately from theoretical predictions,¹⁰ and it is possible to evaluate these terms. As an example consider the measurement of the cross section for the formation of the 4^1S state. The $4^1S - 2^1P$ emission function can be measured directly as defined by equation (1). Although the cascade term for the transition $4^1P - 4^1S$ cannot be measured by our equipment the emission function of the $4^1P - 2^1S$ line can be determined directly, giving us essentially our term Q_{im} . This may be repeated for the 5^1P , 6^1P , etc., levels, allowing a summation over all relevant states i . The ratios of transition probabilities are known theoretically, and hence the cascade correction may be applied and the excitation function of the 4^1S level determined. In the case of the 1S levels, only cascade contributions from the 1P levels are optically allowed, but in the case of the measurement of the cross section for the formation of the 1P levels cascade contributions can occur from both the 1S and 1D levels. On this basis by making a series of measurements of the emission functions of the $n^1S - 2^1P$, $n^1P - 2^1S$ and $n^1D - 2^1P$ spectral lines for $n = 3$ to 7 , the excitation functions for a whole series of the singlet levels of neutral helium have been determined. It is found that only in the case of the n^1P levels were

10. A. H. Gabriel and D. W. O. Heddle, Proc. Phys. Soc. A, 258, 124, (1960).

Progress Report

cascade corrections appreciable.

Considerable experimental difficulty is involved in the measurement of n^1P-2^1S emission cross sections because of the repopulation of the 1P levels by absorption of resonance photons, and depopulation by collisional transfer. We attempted to use the methods developed by Phelps¹¹ and Heddle¹⁰ to correct for these effects. Considerable difficulty was experienced in applying the concept of an effective radius of our collision chamber for eliminating the effects of resonance absorption. Eventually it was decided to calculate an apparent cross section for the emission of the line at a series of target pressures from 0.3 to 5 μ , using equation (1) and extrapolating to zero target pressure to get the true emission cross section. We consider that less error is involved than if the effective radius concept were employed.

HeI $1^1S \rightarrow n^1S$ and $1^1S \rightarrow n^1D$

The experimental results for the excitation functions on the n^1S and n^1D levels are shown in figs. 2 and 4. Also shown are the measurements by De Heer¹², which would appear to lie 20 to 30% above the present data. In view of the combined estimated possible experimental error of 40% for the two sets of data this level of agreement is regarded as satisfactory. On figures 3 and 5 the cross sections for the 4^1S and 4^1D levels are plotted including additional comparisons with the work by Dodd and Hughes,¹³ Sternberg¹⁴

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11. A. V. Phelps, Phys. Rev. 110, 1362, 1958.
 12. F. J. De Heer and J. Van den Bos. Physica (to be published 1966)
(See also J. Van Eck, et al. Physica 30, 1171, 1964).
 13. J. G. Dodd and R. H. Hughes, Phys. Rev. 135, A618, 1964.
R. H. Hughes, et al. Phys. Rev. 122, 525, 1961.
 14. Z. Sternberg, P. Tomas, Phys. Rev. 124, 810, 1961.

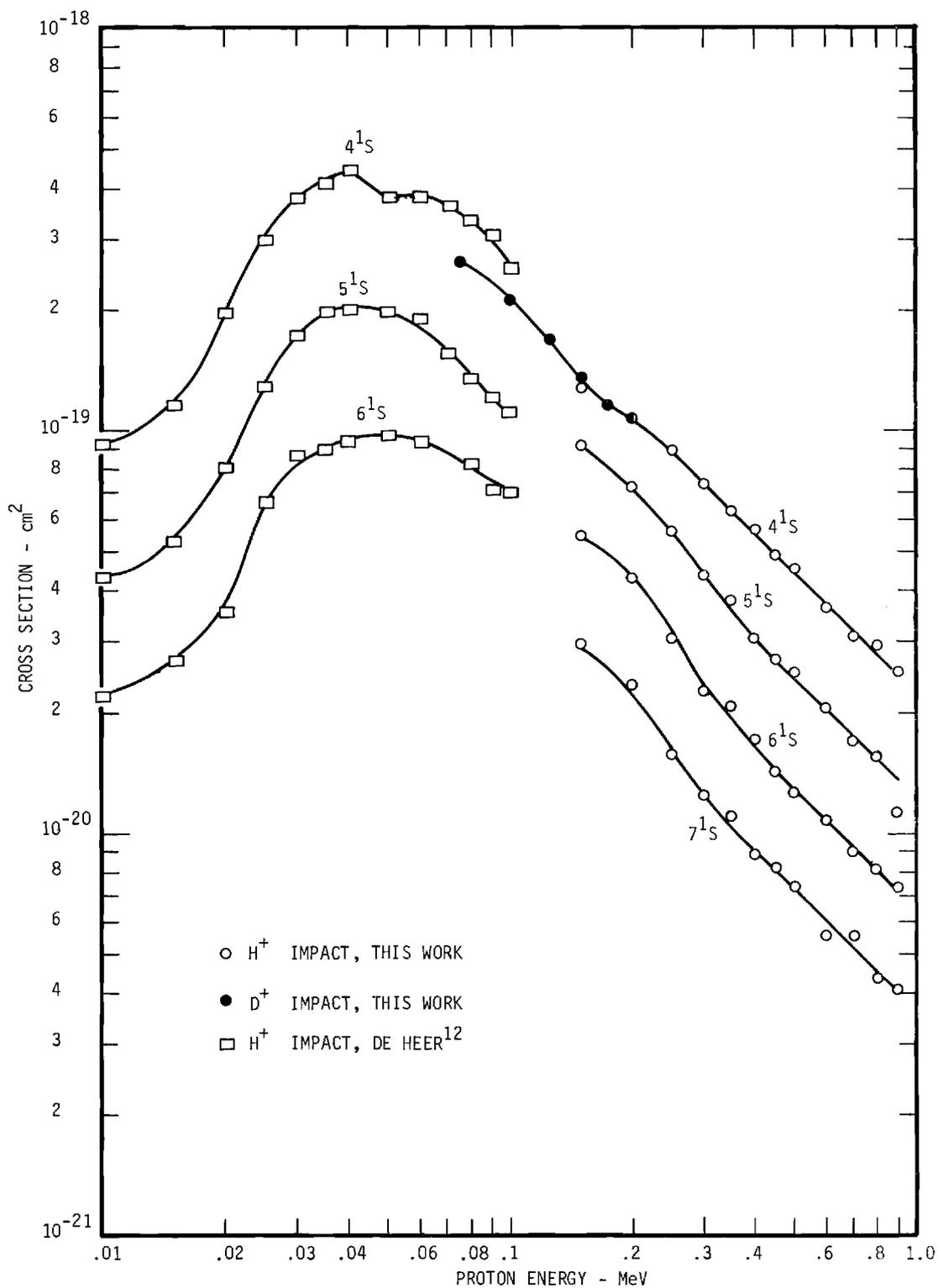


Figure 2. Cross Sections for the Formation of n^1S Excited States of Neutral Helium by the Impact of Protons and Deuterons.

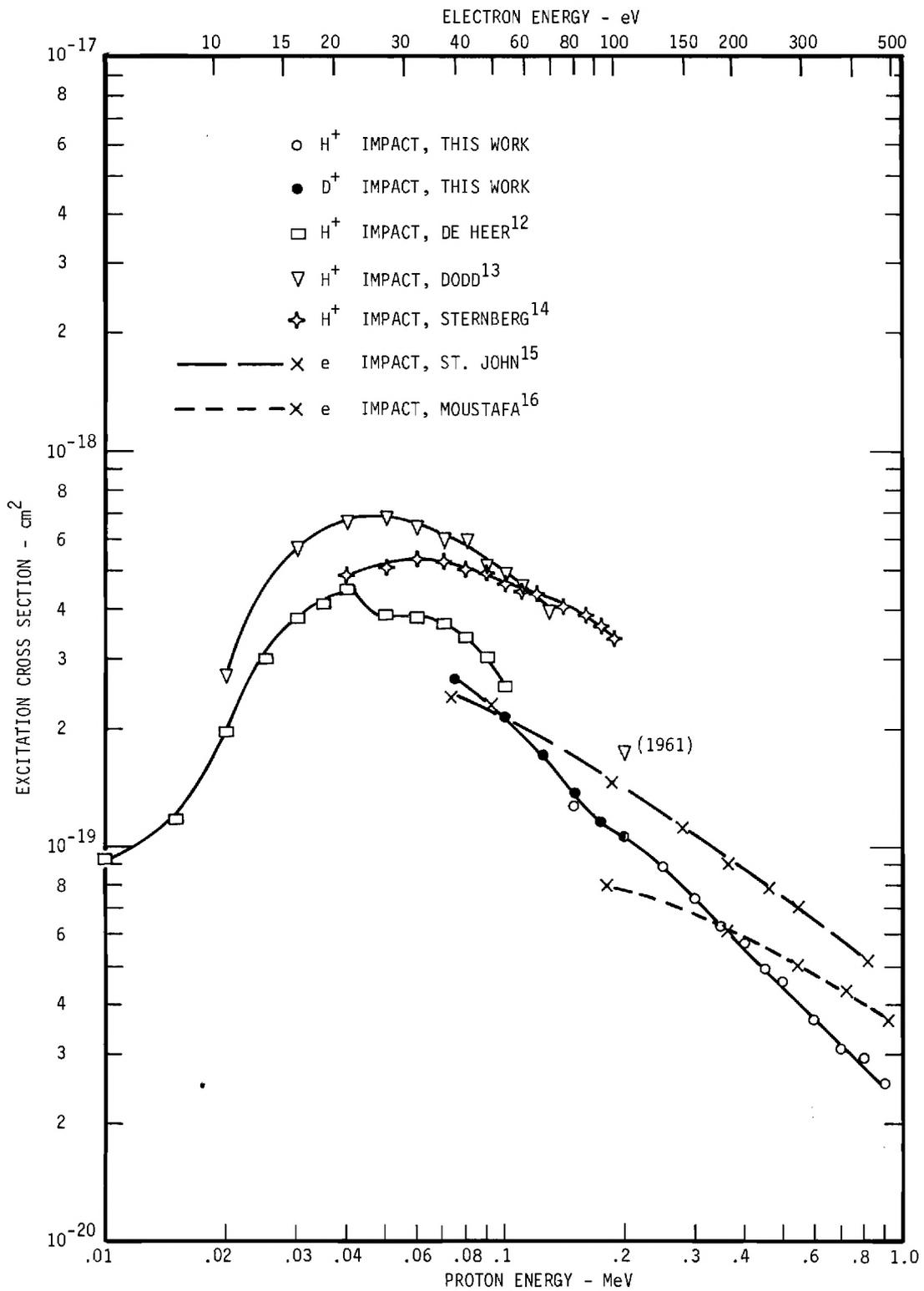


Figure 3. Comparison of the Cross Sections for the Formation of the 4^1S Excited State of Neutral Helium by the Impact of Protons, Deuterons, and Electrons.

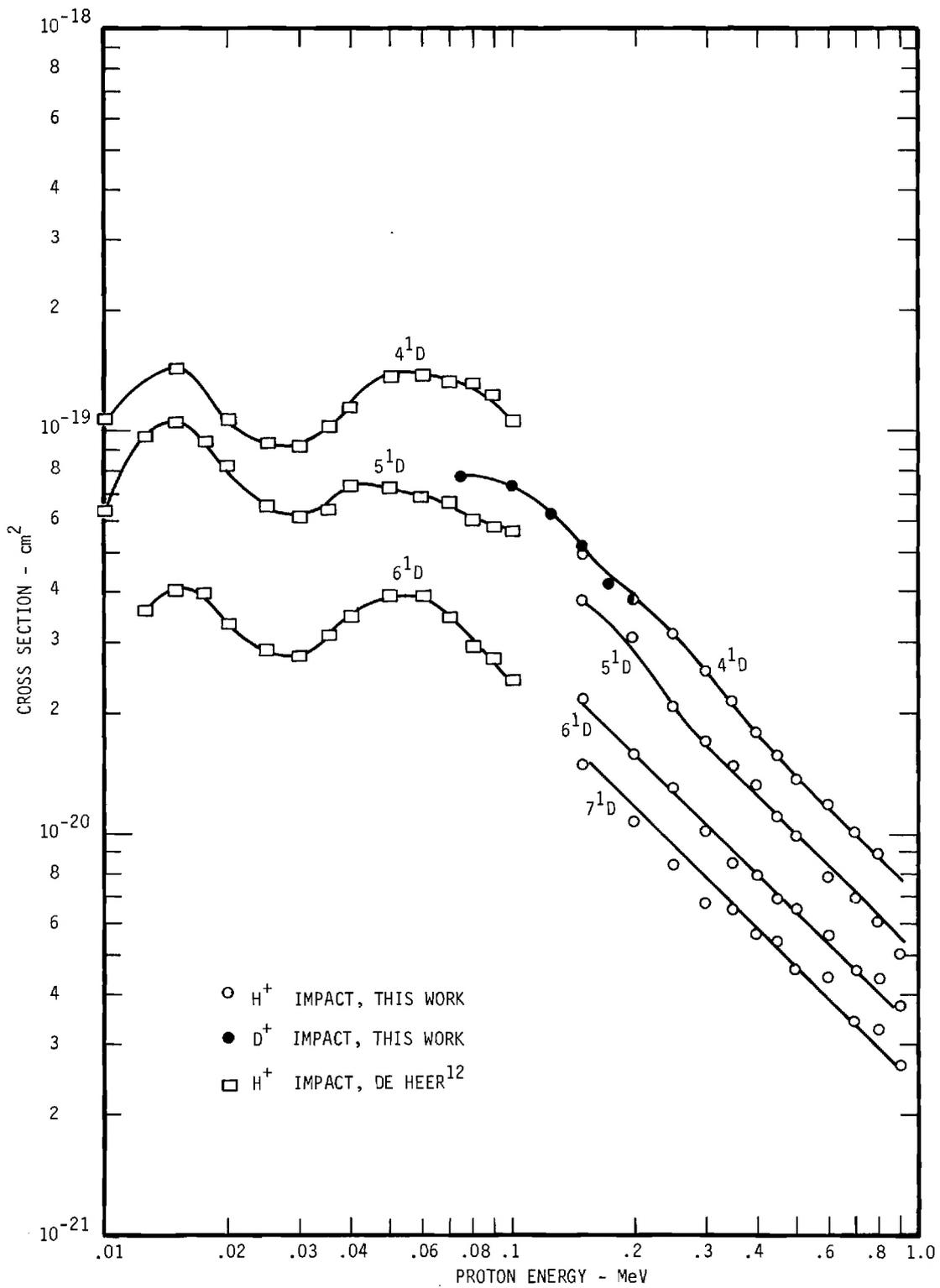


Figure 4. Cross Sections for the Formation of n¹D Excited States of Neutral Helium by the Impact of Protons and Deuterons.

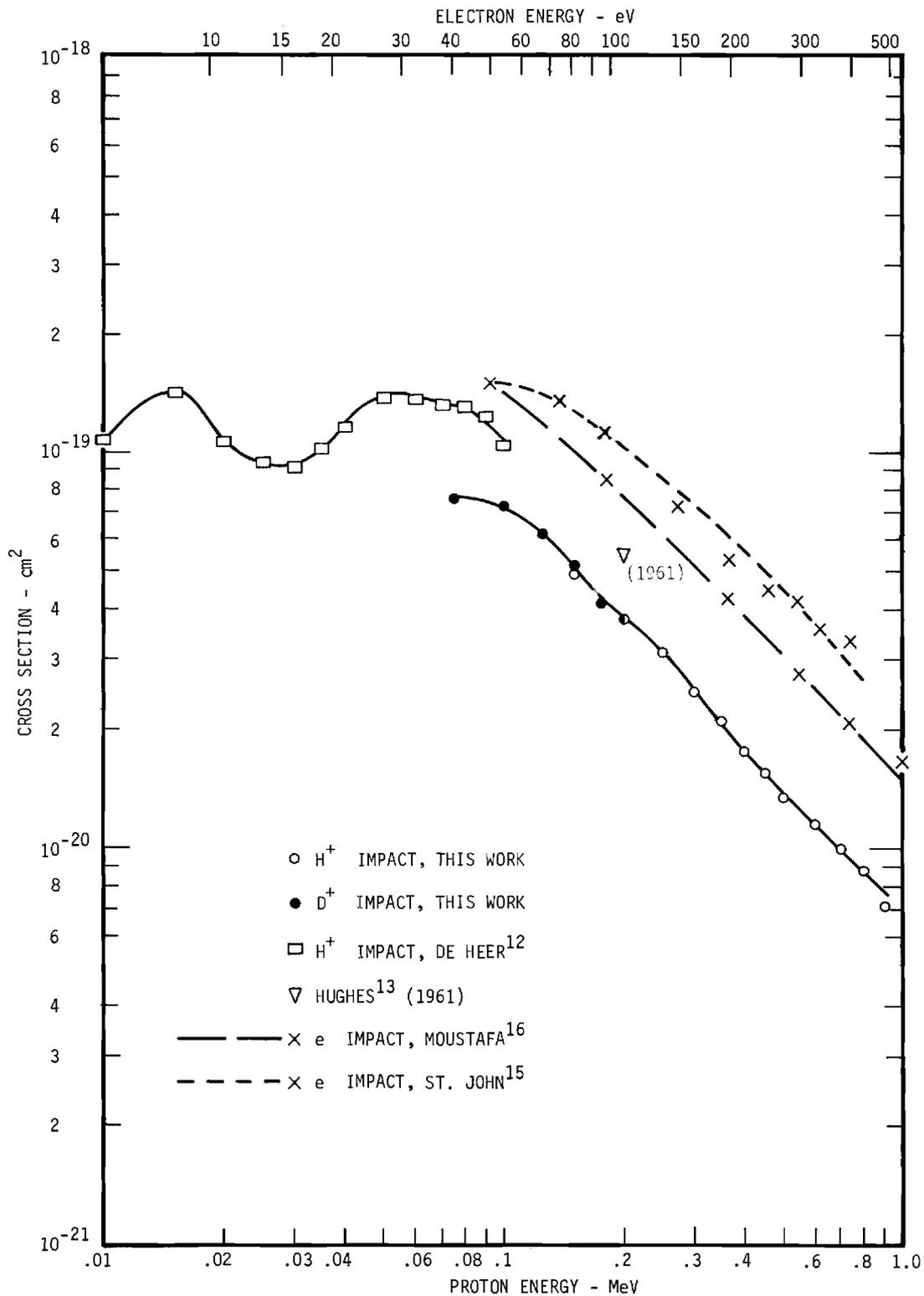


Figure 5. Comparison of the Cross Sections for the Formation of the 4^1D Excited State of Neutral Helium by the Impact of Protons, Deuterons, and Electrons.

Progress Report

and the electron impact data by St. John¹⁵ and Moustafa.¹⁶ Throughout the measurements presented in this report the data by Hughes and co-workers^{13,20,25} appear to lie consistently a factor of two higher than the present work, although the same dependence of cross section on energy of impact is indicated. Excellent agreement between our own work and the very careful work by De Heer coupled with the agreement of our own data with theoretical predictions for the 3^1P level, which we shall describe below, forces us to conclude that the absolute calibration of Dodd's,¹³ experimental data is incorrect. The data by Sternberg¹⁴ bear no very close relationship to any of the other work, and we conclude that these are in error, perhaps through the use of excessively high target pressures. The electron impact data by St. John¹⁵ and Moustafa,¹⁶ would seem to lie generally above our data, although their curves of cross section versus energy have the same shape at high velocities of impact.

It is to be expected that at sufficiently high velocities of impact the asymptotic high energy form of the Born Approximation given in equation (5) should apply for both the 1S and 1D functions. We consider that the most sensitive test for this dependence is to plot the product of excitation cross section and energy ($Q_{ij} \times E$) against energy and identify the region over which this becomes invariant with E . Sample data are shown on fig. 6, indicating the applicability of the Born Approximation for protons having energy of impact of 400 KeV or more. One would also expect that the cross sections for electron and proton impact at the same velocity of impact should be the same. The shapes of the functions are in agreement at high energies, but there is a considerable difference in magnitude between

15. R. M. St. John, et al. Phys. Rev. 134, A888, 1964.

16. Moustafa. Unpublished data, communicated by De Heer.

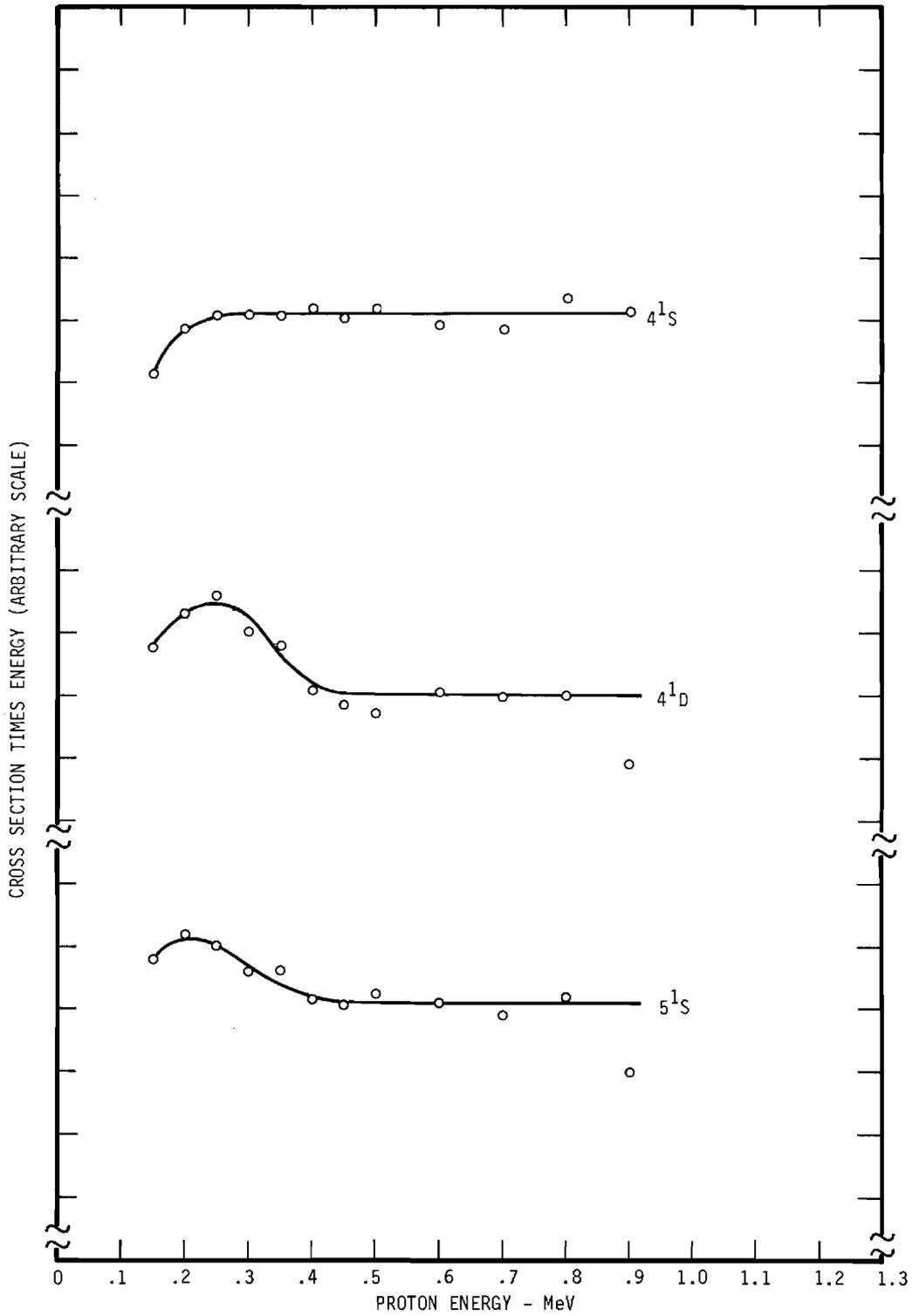


Figure 6. The Product of Cross Section and Energy on an Arbitrary Scale, as a Function of Energy, for Selected Levels. Constancy of the Product Indicated Validity of the Asymptotic High Energy Form of the Born Approximation.

Progress Report

all proton and electron impact data indicating perhaps discrepancies in absolute calibration.

Unfortunately there seem to be no exact theoretical predictions of the cross sections for these quadrupole transitions. We are able to locate only a reference by Seaton¹⁷ to unpublished work by Fox, which gives the first Born Approximation for the excitation of 1S states by impact of electrons on helium at 108 eV. The quoted values of 1.64, 0.81 and $0.46 \times 10^{-19} \text{ cms}^2$ for the excitation of the 4^1S , 5^1S and 6^1S levels respectively agree within experimental accuracy with our measurements for incident protons at the same velocity.

We may apply equation (3) to the data at energies above 400 Kev., where our experimental results show that it is applicable, and calculate values of the constant C for the 1S and 1D states. The results are shown in table I and indicate that in this region the value of C, and hence the magnitude of the cross section, varies as $n^{-3.4}$ for the 1S states and as $n^{-1.9}$ for the 1D .

TABLE I

Experimental Values of C

State	4^1S	5^1S	6^1S	7^1S	4^1D	5^1D	6^1D	7^1D
n	4	5	6	6	4	5	6	7
C x 10^{-14}	2.55	1.22	0.63	0.36	0.70	0.48	0.33	0.24

17. M. J. Seaton. "Atomic and Molecular Processes". (Ed. Bates, D. R., Pub. Academic Press 1962) page 410.

Progress Report

Using the above information we may suggest the following formulae for predicting the cross sections for quadrupole transitions at energies above 400 Kev. derived experimentally from our data. With E as the energy of impact in eV., and n as the principal quantum number,

$$Q(n^1S) = \frac{2.5 \times 10^{-12}}{E \times n^{3.4}} \text{ cms}^2$$

$$Q(n^1D) = \frac{1.0 \times 10^{-13}}{E \times n^{1.9}} \text{ cms}^2$$

It must be stressed that these equations are at best approximate, but it would be interesting to test their validity to higher levels and energies of impact.

HeI $1^1S - n^1P$

The cross sections for the excitation of four 1^1P states have been measured, but due to the recently discovered scattered light problem mentioned in section V e the measurements on the 5^1P and 6^1P levels have a considerable systematic error, the magnitude of which we have not yet determined accurately. We have therefore chosen only to present the 3^1P and 4^1P cross sections as shown in figures 7 and 8. It must be admitted that the absorption of resonance radiation influences the measurement of these lines considerably, and the accuracy of the present work is estimated to be no better than 30%. Figure 7 compares the excitation functions by De Heer¹² with our own work, and in both cross sections we appear to lie 50% below DeHeer. This difference is somewhat greater than for the quadrupole transitions, but still consistent with the combined experimental uncertainty of 50%. Figure 8 gives a more detailed comparison for the 3^1P state with the proton impact work of De Heer,¹² and Dodd¹³, the electron

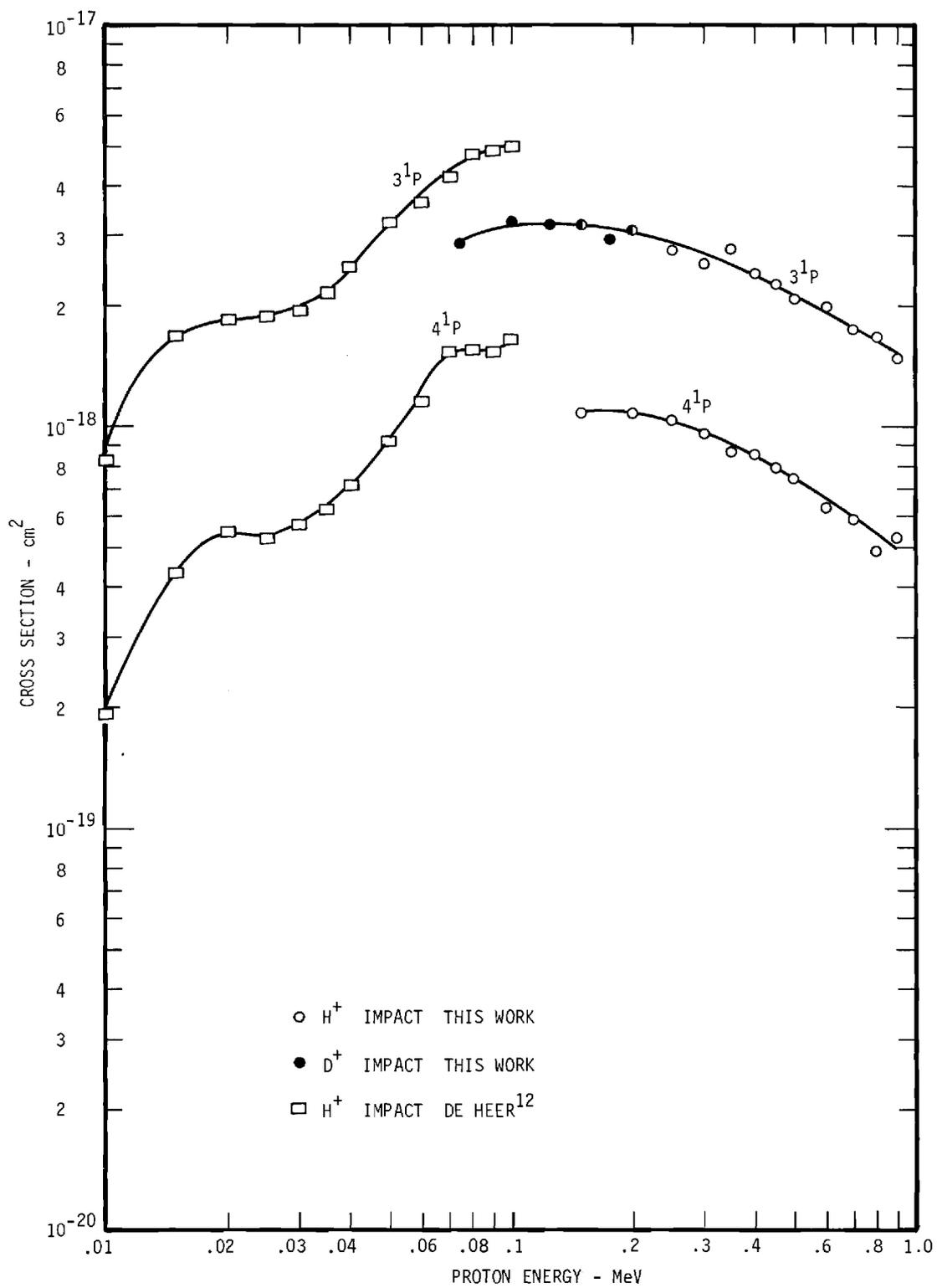


Figure 7. Cross Sections for the Formation of n^1P Excited States of Neutral Helium by the Impact of Protons and Deuterons.

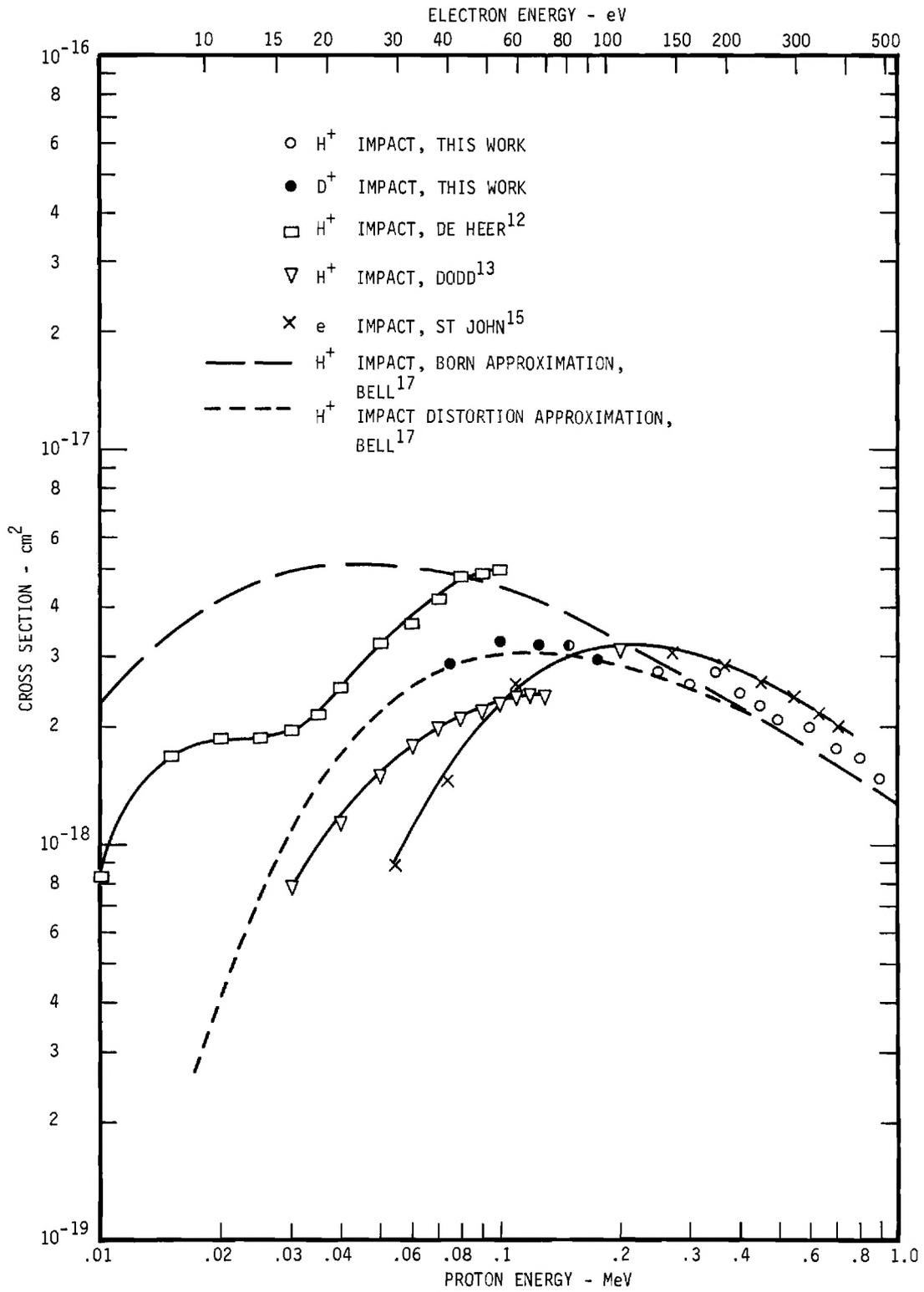


Figure 8. Comparison of the Cross Sections for the Formation of the 3^1P Excited State of Neutral Helium by the Impact of Protons, Deuterons and Electrons with Theoretical Predictions.

Progress Report

impact data of St. John,¹⁵ and the theoretical predictions of Bell¹⁸ using Distortion and Born Approximations.

The difference between the form of the excitation cross sections for quadrupole transitions and dipole transitions is obvious on comparison of figure 7 with 4 and 6. The function shows considerable curvature in our logarithmic plots, and decreases with increasing energy of impact far more slowly than for the 1S and 1P levels. Of most importance in these results is the excellent agreement between the experimental data and the Distortion Approximation prediction by Bell.¹⁸ Using the D^+ data to extend the effective velocity range of our experiment, we see that our data lie consistently about 10% above the theoretical prediction, which of course is well within the estimated error limits of our determination. The close agreement in the shape of the experimental and theoretical cross sections appears to be a substantial confirmation of the validity of the Distortion Approximation down to some 75 KeV. impact energy, which is below the cross section maximum. The Born Approximation is also shown and merges with the Distortion Approximation above 500 KeV, substantially agreeing with our measurements in this region.

HeII (n = 4)

We have measured the cross section for the emission of a single HeII (He^+) spectral line, the $n = 4$ to $n = 3$ transition.

It is to be expected that the formation of the excited state of the ion will occur primarily by the process of ionization leaving the target excited.



18. R. S. Bell, Proc. Phys. Soc. 78, 903, 1961.

Progress Report

rather than the alternative process of charge transfer which would be expected to show a rapid decrease with increasing energy of impact.



There are a total of four different transitions between the $n = 4$ and $n = 3$ states, which all have essentially the same wavelength and cannot be resolved with our equipment. Without knowledge of the separate emission functions for these transitions it is impossible to calculate with any degree of accuracy an excitation function for the upper level, and we therefore present our data as an emission function. If cascade can be neglected, equation (2) shows that the emission function measured should be proportional to the cross section for exciting the $n = 4$ level of the He^+ ion.

We have also indicated on figure 9, the emission function determinations by De Heer¹², which, due to the limited energy range cannot be compared directly with the present data, and also the work of Dodd,¹³ which appears to lie a factor of three higher. The agreement between the shapes of all three cross section determinations is good, although there does seem to be a considerable discrepancy in magnitudes.

At the present time the theoretical estimates for the simultaneous excitation and ionization of the helium target are limited to states $n \leq 3$,¹⁹ however the predictions are of the same basic form as the emission function obtained experimentally.

b. Excitation of a Helium Target by H^+ , H_2^+ , and H_3^+

A brief investigation was carried out to compare the cross section

19. R. A. Mapleton, Phys. Rev. 109, 1166, 1958.

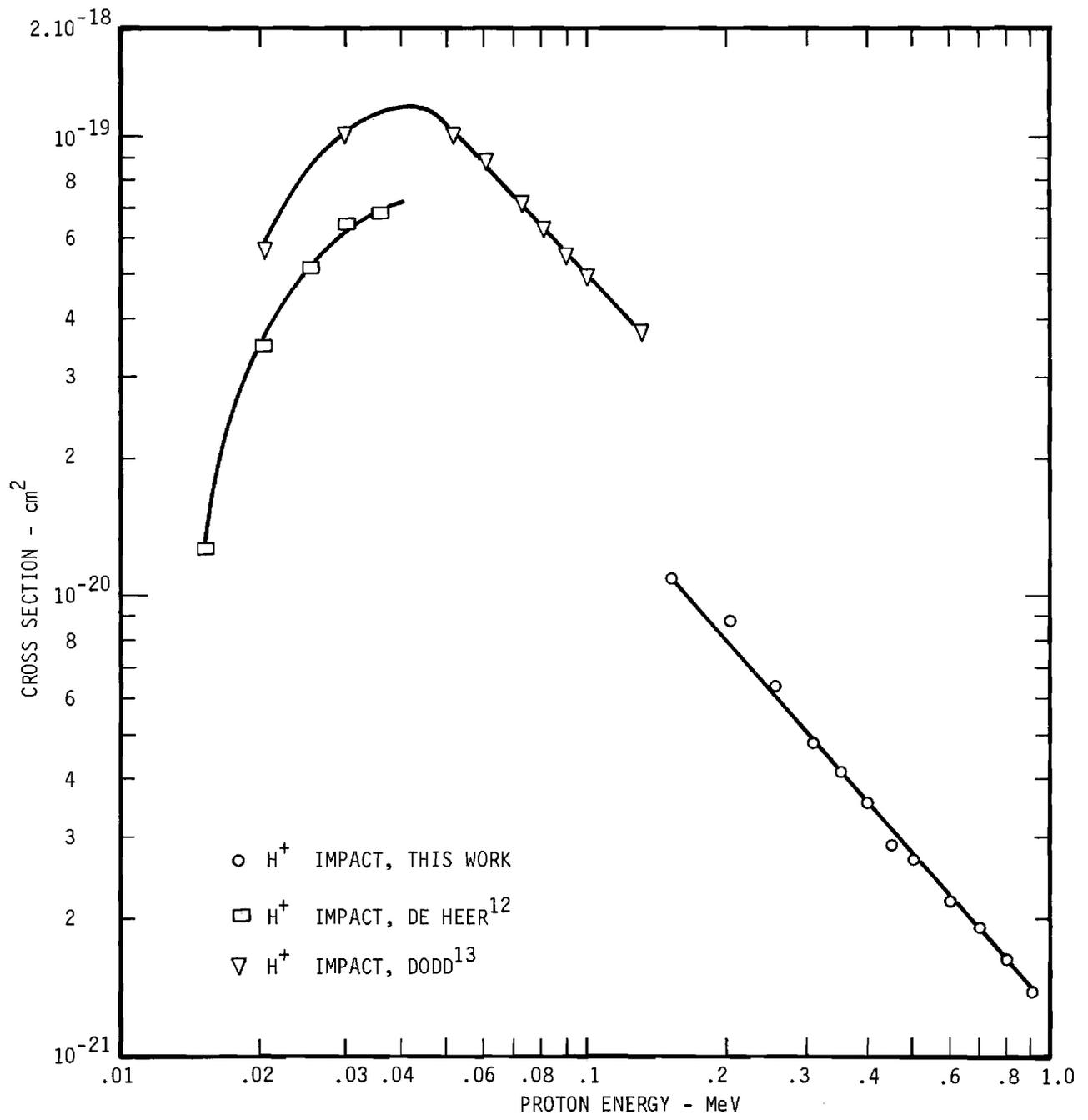


Figure 9. Cross Section for the Emission of the HeII 4686 Å (n=4→n=3) Spectral Line, Induced by Proton Impact on a Neutral Helium Target.

Progress Report

for the excitation of a helium target, by H^+ , H_2^+ and H_3^+ ions. Quite obviously the present experiment can give no information as to whether the incident molecular ions are dissociated by the impact. Our activities were restricted to the measurement of the cross section for the formation of the 4^1S state, and the results are displayed in figure 10, plotted on the same relative velocity of impact scale. Unfortunately the region of overlap between our three sets of data is fairly restricted, but we do observe that all three functions are parallel and that the cross section increases with the mass of the incident particle. Returning to equation (3) for the cross section of a quadrupole transition we notice that for a constant velocity of impact (i.e. constant value of E/M) the cross section varies as the square of the effective charge of the incident particle. Although the effective charge of the H_2^+ and H_3^+ ions at large distances from the nuclei will certainly be unity, there is no reason to suppose that the effective charge is the same in such high energy collisions as these, where the distance of closest approach to the target is of the same order of magnitude as the internuclear separation in the molecule. Assuming, just for the sake of argument, that the incident structure can be considered as a single particle, our data can be explained by applying equation (5) and assuming that the effective charge of the H^+ , H_2^+ and H_3^+ ions are respectively 1, 1.20 and 1.25. As will be mentioned in section c there are some data from our own work and that of Dufay on the excitation of N_2^+ ions in a N_2 target which can be treated the same way to give ratios of the effective charge of respectively 1, 1.14, 1.20. Within the admitted limitations of accuracy of the experiments, these two sets of ratios are approximately the same. This would suggest that as far as excitation of a target by fast ions is concerned the H_2^+ and H_3^+ molecular ions may be

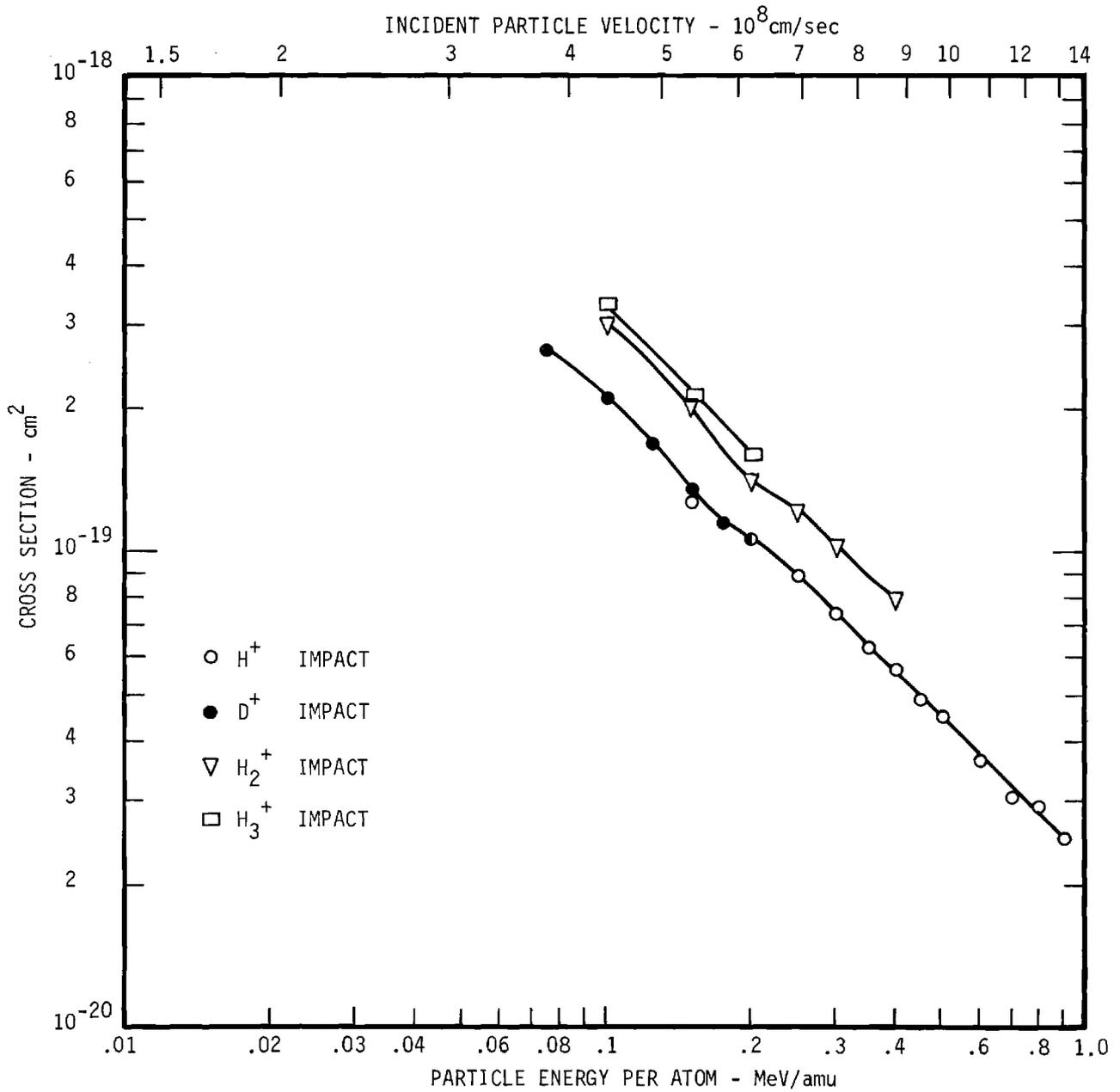


Figure 10. Cross Sections for the Formation of the 4^1S Excited State of Neutral Helium by the Impact of H^+ , H_2^+ , H_3^+ Ions at Equal Velocities.

Progress Report

considered as being single particles of effective charge slightly greater than unity, and increasing with the molecular weight of the ion. An alternative approach to explaining our data would be to consider the incident molecular ion as consisting of two or three separate particles. One might then expect the difference between H^+ and H_2^+ cross sections to be due to the additional H atom, and therefore the difference between H^+ and H_3^+ cross sections being due to two additional H atoms should be approximately twice the difference between the H^+ and H_2^+ data. This is certainly not the case; the addition of the second neutral hydrogen atom to the H_2^+ ion produces only a very small change in cross section. Thus the effective charge interpretation would seem to agree better with experimental data.

We readily admit that this interpretation of our data is entirely phenomenological, but the concept might be worth exploring to see whether it can be used as an approximate device for predicting the cross sections to be expected when using beams of high energy molecular ions.

c. Excitation of a Nitrogen Target by Protons

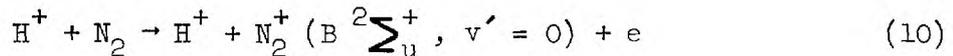
The emission from a nitrogen target bombarded by protons consists primarily of the N_2^+ first negative band system ($B^2 \sum_u^+ \rightarrow X^2 \sum_g^+$) with lesser contributions from the nitrogen ion, NII, and the Balmer Alpha and Beta lines produced by charge exchange leaving the fast particle in an excited state. Cross sections for various molecular transitions have been measured as well as for two NII lines.

The measurement of $v' = 0 \rightarrow v'' = 0, 1, 2, 3$, transitions has been completed, and the emission functions are shown in figure 11. Our data seem to lie consistently a factor of two below that of Philpot²⁰, a

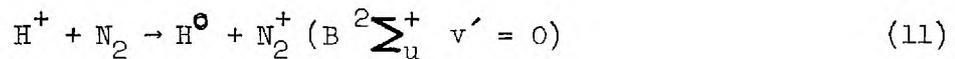
20. J. L. Philpot and R. H. H. Hughes, Phys. Rev. 133, A107, 1964
R. H. Hughes, et al. Phys. Rev. 123, 2084, 1961.

Progress Report

feature which as has been noted in our helium measurements occurs for all data produced by Hughes and co-workers^{13,20,25}, and presumably reflects a difference in calibration accuracy. By summing the cross sections for all the emissive transitions out of the $v' = 0$ level one can estimate the cross section for populating the $v' = 0$ level. There is essentially no cascade into this level and the first negative band system includes all transitions out of it. Since the $v' \rightarrow v''$ transitions have rapidly decreasing emission cross sections with increasing lower vibrational state v'' , we can confidently neglect states above $v'' = 3$, and arrive at the cross section for populating the $v' = 0$ level shown on figure 12. This is essentially the process;



It is expected that the formation of this state by charge transfer



is unlikely since the total cross section for the charge transfer cross section, integrated over all excited states of both particles, is approximately the same as our measured excitation cross section at 150 KeV., and some three orders of magnitude less at 1000 KeV. We conclude that the process shown in equation 10 predominates. Our measured population of the $v' = 0$ level is compared in figure 12 with the work of Philpot²⁰, Dufay,²¹ and the electron impact data of Stewart.²²

21. M. Dufay, et al. Comptes Rendos 261 1635, 1965.

22. D. T. Stewart, Proc. Phys. Soc. (London) A. 69, 437, 1956.

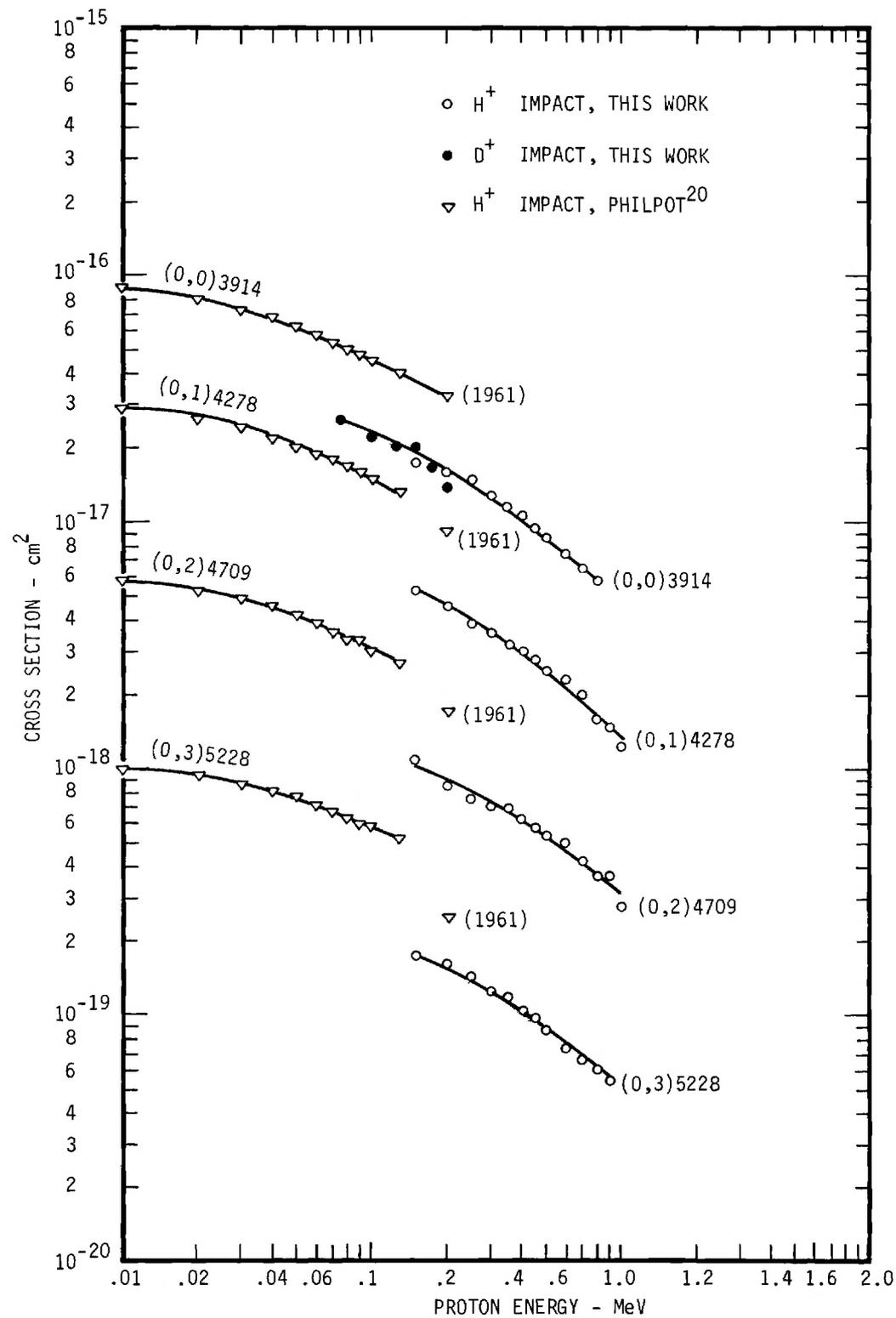


Figure 11. Cross Section for the $v'=0$ Progression Emissions in the N_2^+ First Negative System, Induced by the Impact of Protons on a Molecular Nitrogen Target.

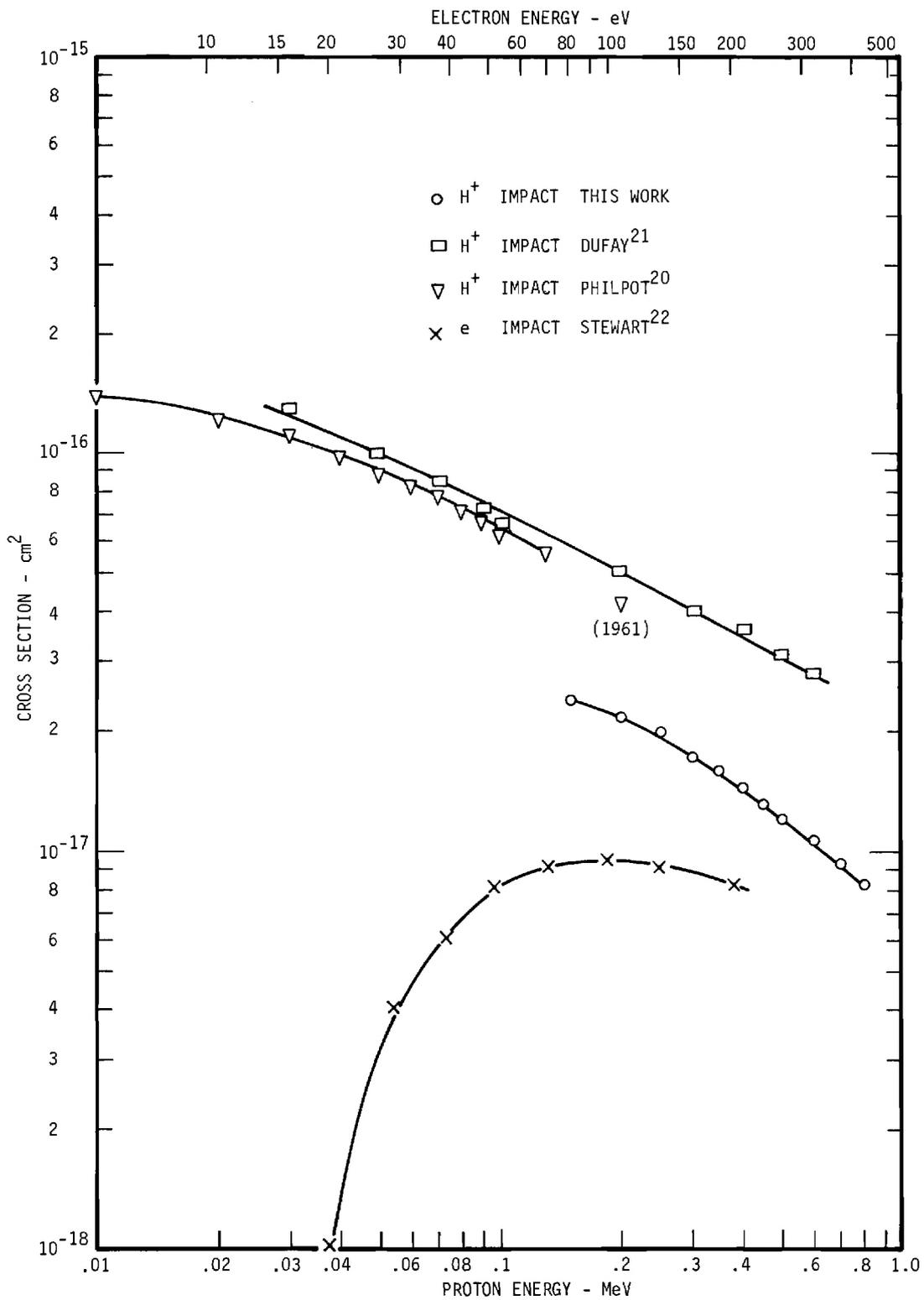


Figure 12. Population Cross Section for the $v'=0$ Level of the $B^2\Sigma_u^+$ State of N_2^+ , Induced by Proton Impact on a Molecular Nitrogen Target.

Progress Report

We have also attempted to measure the emission cross sections for the $v' = 1 \rightarrow v'' = 0, 1, 2, 3$, progression, but the wavelengths of the relevant transitions all lie below 4000 \AA and the absolute calibration has been affected by the scattered light problem mentioned in section V e. It would seem that the published data by Philpot²⁰ involves a similar defect since the ratio of the various intensities for these emissive decays certainly does not agree with the expected Franck-Condon factors and indeed two transitions appear to have the same emission cross section. The general problems of scattered light influencing our absolute calibrations is currently being investigated. When the corrections are assessed, they may be applied without remeasuring the emission from the experiment.

A brief investigation was carried out on the relative cross sections for excitation of the N_2^+ system by H^+ , H_2^+ , and H_3^+ at the same velocity of impact. The ratios of the cross sections were 1: 1.3: 1.44, which as mentioned in section VI a may be regarded as implying that the H^+ , H_2^+ , and H_3^+ ions behave as single particles with effective charges in the ratio of the square roots of the cross section ratios. The work of Dufay²¹ gives essentially the same ratio of cross sections for H^+ and H_2^+ excitation, as the present work.

If a Boltzmann distribution of excited atoms among the available rotational states is assumed, then one may utilize the measurement of the intensity of the peaks in the rotational spectrum to derive an apparent rotational temperature, as indicated by Herzberg.²³ We have carried out this procedure and find that the rotational temperature is approximately the same as room temperature, namely $310^\circ\text{K} \pm 30$. One would expect that

23. G. Herzberg. Spectra of Diatomic Molecules, Van Nostrand, 1950 pages 205 ff.

Progress Report

the distribution of molecules among the various rotational states in the neutral target gas should certainly be governed by the Boltzmann distribution appropriate to room temperature. The fact that this distribution remains unchanged after the collisional processes have taken place implies that the rotational state of the molecule is not influenced by the collision. This is reasonable since the time of interaction of the fast particle and the target is given approximately by the product of the target size and the incident particle velocity, about 10^{-17} secs compared with the time of one vibration of about 10^{-9} secs. One would expect that interaction time should be of the same order or greater than the vibrational period before a transition is likely to take place. This is confirmed by the observation of enhancement of upper rotational states for slow heavy particle impact.²⁴

The cross sections for the emission of the two NII lines 5005 \AA ($3p \ ^3D \rightarrow 3d \ ^3F^{\circ}$) and 5680 \AA ($3s \ ^2P^{\circ} \rightarrow 3p \ ^3D$) are shown in figure 13 compared with the data by Philpot,²⁰ and the measurement by Dufay²¹ on the 5680 \AA line only. The cross sections for the emission of these two lines are exceedingly close and indeed Philpot draws a single curve to represent both of them. Our own data are essentially the same for the two lines within the statistical fluctuations of our measurements, but are plotted with separate lines of best fit. Our data lie essentially a factor of two below Philpot's, the comparison having been made with the assistance of our D^+ data.

These emissions must arise as the result of one of the following processes:

24. R. P. Lowe, H. I. S. Ferguson. Proc. Phys. Soc. (London) 85, 813, 1965.

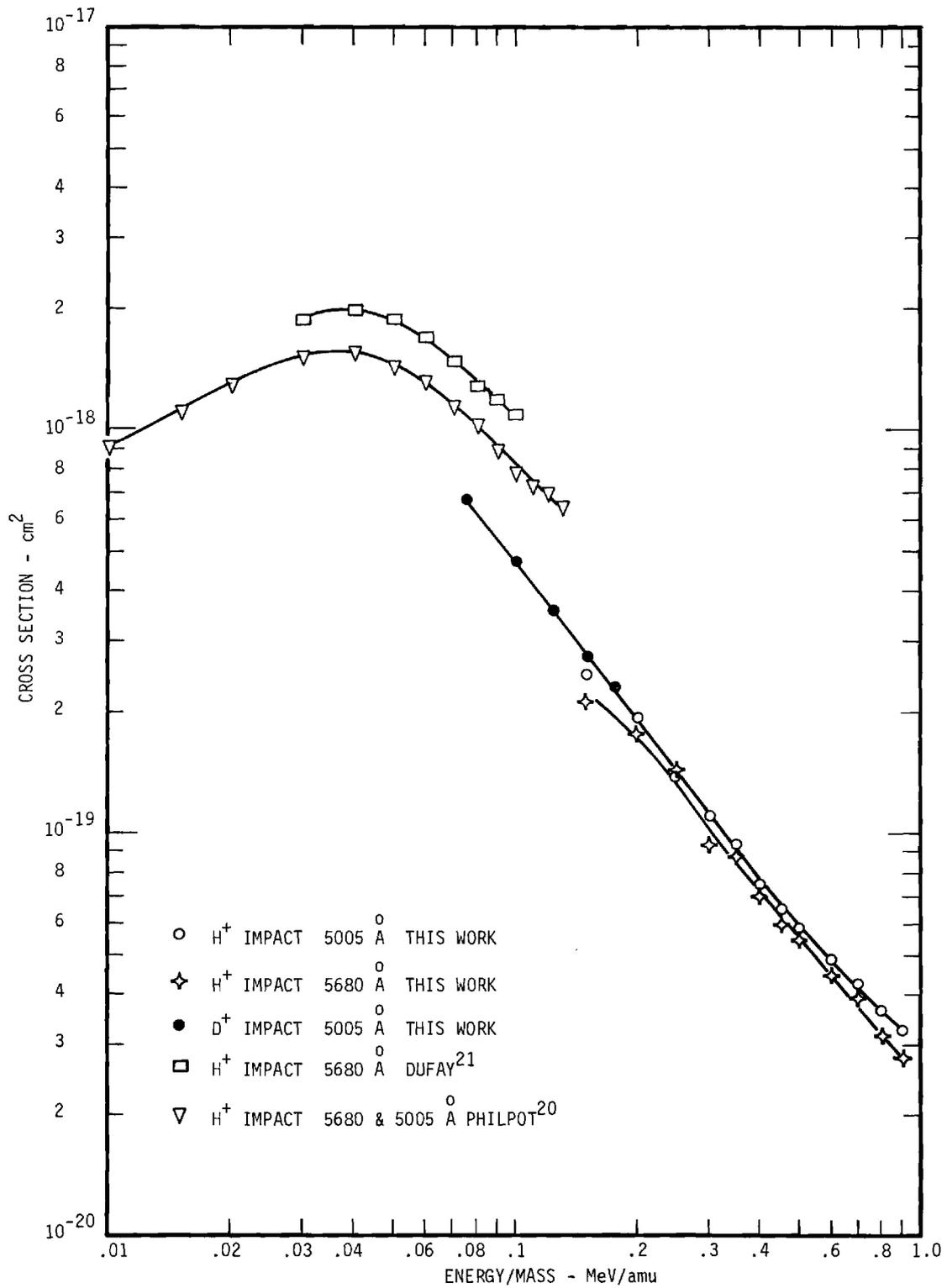
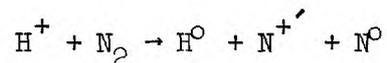
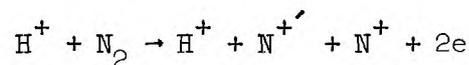
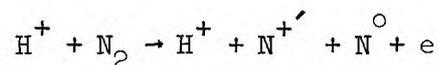


Figure 13. Cross Sections for the Emission of the NII 5005 and 5680 Å Lines Induced by Proton Impact on a Molecular Nitrogen Target.

Progress Report



Of these the third is improbable since the total cross section for all charge transfer processes, irrespective of whether the target is dissociated or excited, is one half the sum of these two emission functions alone, at 1000 Kev. Certainly the total cross section for the formation of all excited N^+ states will be very much higher than the two lines measured. At the present time it is not possible to make a choice between the relative likelihood of the other two mechanisms.

d. Excitation of an Oxygen Target by Proton Impact

The emission from an oxygen target bombarded by protons consists primarily of the O_2^+ first negative system ($b \ ^4\Sigma_g \rightarrow a \ ^4\Pi_u$) with a considerable number of OII and a few OI lines. The Balmer Alpha and Beta lines formed by charge transfer leaving the fast particle in an excited neutral state are also clearly observed. It is our intention to measure the cross sections for the emission of various O_2^+ transitions, and one spectral line of ionized oxygen OII.

The measurement of the molecular transitions in this case turns out to be considerably more complex than for nitrogen, because the rotational structure of the line extends over a considerable range of wavelengths, and strictly speaking the integral of intensity should be determined in order to measure the emission function for a transition between specific vibrational levels. In the case of nitrogen, the range covered by rotational transitions is rather smaller, the majority occurring in a range of about

Progress Report

6 Å, which is well within the bandpass of our spectrometer. In dealing with oxygen we intend to use the same technique as Hughes²⁵ of measuring the integrated intensity over all rotational states at a few impact energies and then determining only the variation of the intensity of the band head with impact energy. We have already confirmed that the distribution among rotational states does not change with energy and therefore this is a valid procedure. A further complication lies in the fact that some of the rotational bands for different vibrational transitions overlap appreciably, and the degree of interference will have to be estimated using a predicted shape function of the rotational structure intensity and a direct measurement of the intensity of the band head. Even when the cross sections for the vibrational transitions are determined, the limited spectral range of our instrument will prevent measurement of more than one transition out of any particular upper vibrational level, and an estimation of level population cross section will only be possible using calculated Franck-Condon factors. It is clear that the analysis of the molecular emission data will be difficult and at the present time it has not been completed. We do however have good measurements on the relative variation of the $v' = 0$ and $v' = 1$, population cross sections and these are shown normalized to the data of Hughes²⁵ in figure 14. We observe a considerable difference between the energy dependence of the data by Hughes and the present work. Since additional work by Dufay²¹ agrees with Hughes, extending the available data to 600 KeV., with substantially the same energy dependence, there is clearly a fundamental problem in reconciling the two sets of data. We intend to pursue this problem further, paying

25. R. H. Hughes and D. K. W. Ng. Phys. Rev. 136, A1222, 1964.

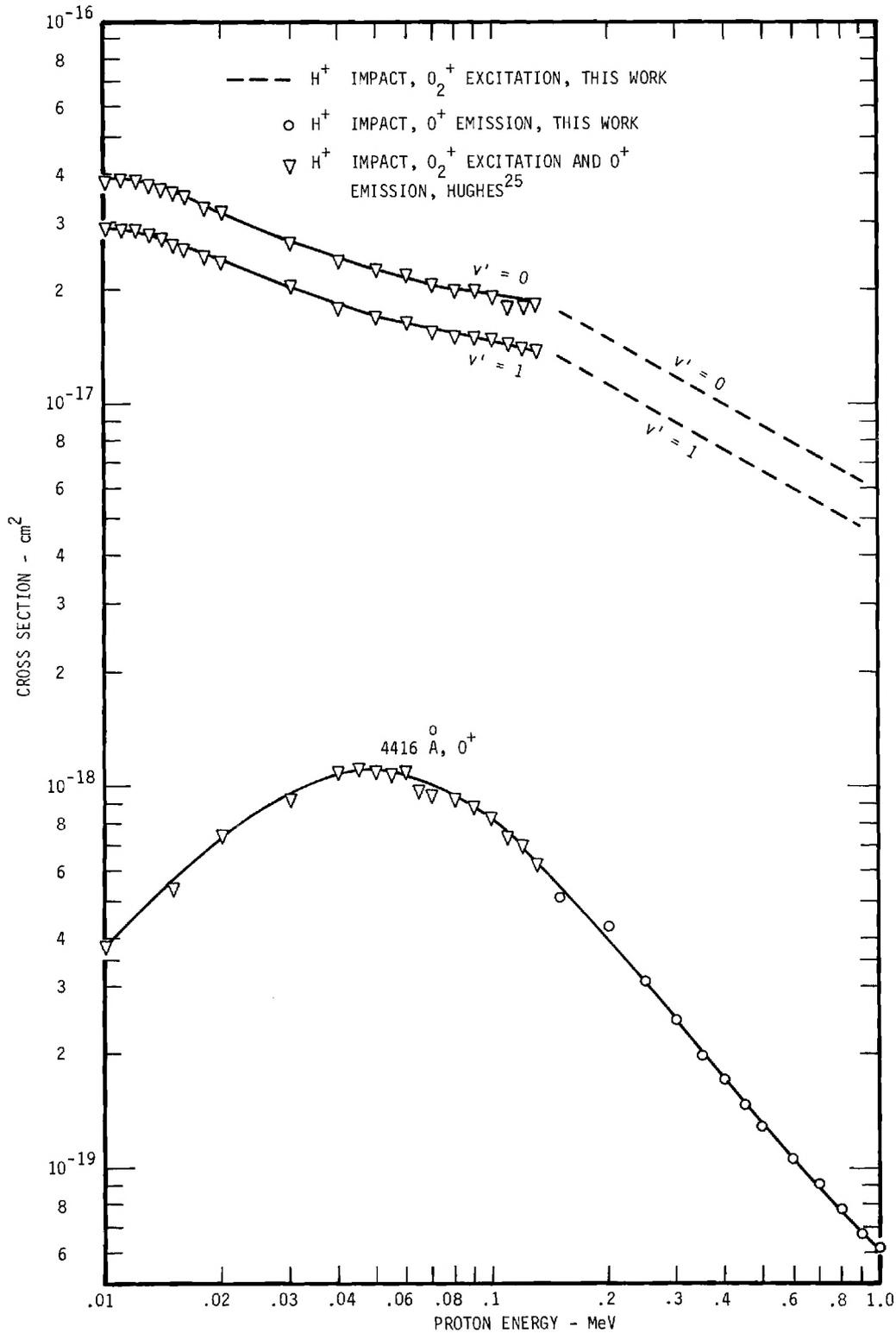


Figure 14. Cross Sections for the Population of the $v'=0$ and $v'=1$ Levels of the $b^4\Sigma_g^+$ State of O_2^+ , and for the Emission of the OII 4415 and 4417 \AA Lines, Induced by Proton Impact of a Molecular Oxygen Target. This Data has been Normalised to the Data of Hughes at 150 keV.

Progress Report

particular attention to the rotational structure and the possible influence of spectrometer slit widths.

Also shown on figure 14 is the emission function for the OII $3s \ ^2P_{1/2, 3/2} \rightarrow 3p \ ^2D_{3/2, 5/2}$ line, normalized to the extrapolated data of Hughes at 150 KeV. Agreement in the cross section curve shapes is excellent and the data only requires to be placed on an absolute basis by the normal calibration techniques.

e. Excitation of an Argon Target by Protons

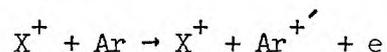
A survey of the spectrum emitted as a result of the impact of protons on argon indicates the formation of excited states of both neutral and singly ionized argon. The very great complexity of the spectra of argon and its ions makes a definite identification of each spectral line very difficult since a resolution of about 16 \AA must be used in order to collect useful light intensities. There are indications of Ar^{++} and Ar^{+++} emission in the ultraviolet spectral regions, but we cannot confirm the identification.

A useful comparison may be made with the general features of the observed spectrum in the present work, and the previous investigations of Thomas.²⁶ Proton impact seems to produce a considerable emission from both the Ar and Ar^+ systems, with the neutral lines, although fewer in number, usually more intense than the ion lines. Previous work,²⁶ using high energy incident beams of heavy ions, He^+ , Ne^+ , Ar^+ , and Kr^+ , indicated emission only from the Ar^+ excited states in the target, with no identification of the emission of neutral lines. Comparison of the emission functions with the known data for production of singly charged ions in the target by ionization and charge transfer, suggested that the latter

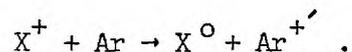
26. E. W. Thomas, Proc. Phys. Soc. (London), 85, 363, 1965.

Progress Report

process preferentially produced excitation. That is to say the excitation was more likely to occur as



than



In the case of heavy incident particles at energies from 100 to 500 KeV, the ionization and charge transfer cross sections are of comparable magnitude. In the present work with incident protons, the charge transfer cross section is less than 5% of the ionization cross section and we observe that the Ar^+ emission is not dominating the overall spectrum. This leads to the fairly general suggestion that charge transfer may involve a preferential formation of excited target states. Only a coincidence experiment of considerable complexity could be expected to elucidate this problem.

The impossibility of taking into account all cascade and branching transitions will dictate that data from such measurements could not be expressed as cross sections for the formation of an excited state but must inevitably be left in the form of emission functions. Although no measurements have yet been carried out, it is our intention to investigate the $Ar^+ \ 4p \ 4D^0_{1\frac{1}{2}} \rightarrow 4s \ 4P_{\frac{1}{2}}$ transition for comparison with the previous data by heavy particle impact, and also the neutral line $ArI \ 5p \ 2 \rightarrow 4s \ 1^0$.

VII. Program for the Remainder of the Contract Year

Our primary objective will be to complete the measurement of cross sections for the formation of excited states in the target. The recently identified problem associated with the absolute calibration of the apparatus at wavelengths below about 4000 Å (see section V e) due to scattered light must be eliminated and certain cross sections for helium and nitrogen targets will require correction to take into account this error in absolute

Progress Report

calibration. The data obtained with an oxygen target must be analyzed fully and absolute cross sections for the formation of particular excited states determined. Proton impact on argon will be investigated, measurements being made on one excited state of Ar and one of Ar^+ . The excitation of a molecular hydrogen target will also be considered, utilizing the Doppler shift of light from the fast particle to separate emission from the target and fast particle systems. It is anticipated that the emission functions of the Balmer Alpha and Beta lines will be measured.

The apparatus designed to measure the formation of excited states in the fast beam will be brought into operation and initial checks and calibrations carried out. It is hoped that some preliminary data on the formation of the $3s$, $3p$, and $3d$ states of hydrogen by charge transfer processes as protons pass through a nitrogen target will be obtained before the end of this period.

VIII. Program for the Future

It is intended that the future effort in this program should be devoted to the study of the formation of fast excited hydrogen atoms, produced by the passage of a beam of fast particles through a gas cell. The equipment required for this type of work is currently being assembled and tested. Two basic techniques are available. Emission from the fast particles may be observed as the beam traverses a long gas cell. An exponential build-up of intensity with penetration through the cell toward some constant value is expected. Alternatively the beam may be passed through a short cell and the emission observed from the fast beam as it traverses a subsequent long, evacuated flight tube. In this case one expects an exponential decay of intensity with increasing distance from the gas cell. In both cases the exponent of the decay or build-up is

Progress Report

characteristic of the velocity of the excited particle and the lifetime of the excited state. This is of considerable advantage when dealing with the formation of the $3s$, $3p$, and $3d$ states of hydrogen, where the radiative decays $3s-2p$, $3p-2s$, and $3d-2p$, emit photons of essentially the same wavelength which cannot be resolved. The three states have considerably different lifetimes and the intensity of emission of the Balmer Alpha line will vary as the sum of three exponentials. Knowledge of the three lifetimes and the careful analysis of the build-up or decay of emission should in principle allow the separate determinations of cross sections for the formation of the $3s$, $3p$, and $3d$ states.

We feel that it is necessary to carry out the experiment using the two modes of experimental operation because of problems that may be foreseen with each. In the case where the emission is observed in the gas space itself, there is always the problem of isolating the spectral line of interest, and there is an indication from our preliminary surveys that conflicting spectral lines lying close to the Balmer Alpha will be encountered with many of the target gasses we plan to use. The obvious answer to this problem is to go to the evacuated flight tube configuration where the problem is entirely eliminated. The use of a short gas cell followed by a flight tube is in its turn subject to errors involved in precise determination of the cell's length and the possibility that the cell apertures will intercept part of the fast particle beam. Also this whole experimental procedure for determining the excitation functions of the $3s$, $3p$, and $3d$ states requires that mixing of these states by magnetic or electric fields should not occur, and the possibility of building up insulating deposits on the gas cell exit aperture must be avoided.

Progress Report

It is our intention to study first the formation of excited H atoms by charge transfer as the proton beam traverses various target gases. Later the work will be extended using the same experimental configuration to study the formation of these same states by the dissociation of H_2^+ and H_3^+ ions incident on the same targets. Apart from the academic interest in these cases, there is considerable interest in the processes which lead to the formation of higher excited states from the point of view of designing injection systems for plasma machines. Data obtained for the formation of the relatively low lying excited states may be used to test the validity of theoretical predictions which are applicable to all excited levels. Also there are well established rules by which measurements on the formation of lower states may be extrapolated directly to give the cross sections for the higher levels.

IX. Publications and Travel

There have been no publications in the open literature during this year. A continuing uncertainty in our absolute calibrations, certainly the most vexing problem associated with the measurement of excitation cross sections by optical techniques, has made us reluctant to present our data for publication. However, the work on helium and nitrogen now appears to be complete, and will probably be submitted before the end of the contract year.

A Technical Report giving a categorized listing of all sources of data on excitation processes resulting from collisions between atomic structures, has been prepared by one of us (E.W.T.)³ and will receive a limited circulation.

Progress Report

Dr. Thomas and Mr. Bent attended the "Fiftieth Anniversary Meeting" of the Optical Society of America, in Washington, D.C., in March, 1966, and a paper was presented.²⁷

The 19th Annual Gaseous Electronics Conference was held in Atlanta in October, 1966, and was attended by Dr. Thomas, Mr. Bent, and Mr. Edwards. Dr. Thomas was on the local committee, and also received an invitation to be a session chairman. A paper was presented by Mr. Bent.²⁸

Dr. Thomas gave an invited seminar in December, 1965 to the physics department of Auburn University, Alabama. Periodic visits have been made by Dr. Thomas, accompanied on occasion by Mr. Bent, to the Oak Ridge National Laboratory, for the purpose of consultation with C. F. Barnett and others, as well as the use of facilities for equipment calibration.

X. Personnel

The work described in this report was carried out as part of A.E.C. Contract No. AT-(40-1)-2591 and has been under the jurisdiction of Dr. Edward Thomas.

Mr. Gary Bent continues as senior graduate student on the project and is expected to write a thesis on this work which should be prepared within about six months.

Mr. Lee Edwards has been working for this project approximately full time since June, 1966. He is at the present time the recipient of a N.A.S.A. scholarship and draws no financial support from the project. Mr. Edwards has a first class academic record, and we expect that this work will provide the basis for his Ph.D. thesis problem in the future.

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27. E. W. Thomas and G. D. Bent, Journal of the Optical Society of America 56, 552, 1966 (Abstract only).
28. E. W. Thomas, G. D. Bent and J. L. Edwards. (Abstract to be published in Bul. Am. Phys. Soc.).

Progress Report

During the course of the year a total of four other graduate students have worked on various facets of this program, two of them at no cost to the project. We feel that by providing facilities for such men to work for relatively short periods, we are not only advancing the progress of the project but also contributing to the education of the people involved. Mr. Ben DeMayo was responsible for constructing the equipment for carrying out the work on vacuum gauge calibration reported in the Appendix, while Mr. Frederik Davis has carried out some circuit development work. Both of these men were working on special problems for academic credit and received no remuneration from the project. Mr. W. Baucum and Mr. J. Takacs have recently been employed to deal with a backlog of data in need of processing and certain problems of ion beam alignment. Both of these men are employed with funds made surplus by Mr. Edwards' scholarship, and their continued employment will be periodically assessed in the light of available funds.

XI. Accelerator Usage

Use of the Van de Graaff accelerator is shared with a project to measure ionization cross sections, also under AEC Contract No. AT-(40-1)-2591. No difficulty is experienced in switching the ion beam from one experiment to the other. A period of one day suffices to make the required mechanical connections and to align the ion beam with the experiment. The accelerator operation time is shared equally by the two experiments, alternating at intervals of one month or longer.

XII. Incident Report

There have been no incidents for which a report is required during the performance of the research under this contract in the present reporting period.

APPENDIX I

The Calibration of a Differential Capacitance
Manometer for Pressure Measurement

by

E. W. Thomas and B. de Mayo

Progress Report

APPENDIX I

The Calibration of a Differential Capacitance

Manometer for Pressure Measurement

An essential requirement in any measurement of an atomic collision cross section by a beam-target experiment, is a knowledge of the target density. When gaseous targets are used, typical operating pressures are in the range 10^{-5} to 10^{-2} mm Hg., a range in which the McLeod gauge is the only absolute pressure measuring device available. It has recently been shown that the cold traps used on such gauges to prevent contamination of the experimental system with mercury may introduce an appreciable error, due to the streaming of mercury onto the cold trap, acting as a diffusion pump.^{1,2,3)} Our own experience⁴⁾ has been that when a "dirty" McLeod gauge is used, the pumping effect, if it exists at all, produces an error of about 5% or less for Nitrogen, about half the value predicted theoretically. By a "dirty" McLeod gauge, we mean one which has been in use for some time without cleaning. We believe that a dirty mercury surface will inhibit the rate of evaporation of mercury, resulting in a reduction of the pumping

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1. Ishii, H. and Nakayama, K. I. Trans. 8th Nat. Vac. Symp. Pergamon Press. London. (1962). p. 519.
 2. Meinke, G. and Reich, G., Vakuum Technik. 12, 79, (1963).
 3. Takaishi, T., Trans. Faraday Soc., 61, 840, (1965).
 4. Thomas, E.W., Bent, G. D. Technical Status Report No. 1. Project B-176-002, November 30. (1965) p. 13.

Progress Report

error. Since all published determinations of this error were carried out in very clean systems, the effect would have been at its full predicted value. Unfortunately a literature search has failed to disclose any information on the manner in which surface contamination effects vapor pressure.

It has been frequently suggested that an alternative pressure gauge may be devised around the deflection suffered by a metal diaphragm when subjected to a pressure differential. A number of such devices are commercially available, where the diaphragm is made the common plate in a differential capacitor. As shown schematically in Fig. 1, one side is connected to a reference pressure of about 10^{-6} mm Hg., the other to the test system. Based on the conclusions of a recent survey⁵⁾, and on our own scrutiny of available types, it was decided to purchase the differential capacitance manometer known commercially as the Baratron⁶⁾, for assessment and subsequent use, in the present program. The major concern with the use of the Baratron is the accuracy of calibration. The manufacturer utilizes a dead weight testing procedure to calibrate with high accuracy between 1.0 and 0.1 mm Hg., and assumes a linear extrapolation to lower pressures. For our purposes the accuracy of calibration must be checked in the 10^{-5} to 10^{-3} mm Hg. region, and a simple test stand was designed and built to carry out this work.

The test system, shown diagrammatically as Figure two, incorporates a reference vacuum system for the Baratron gauge which produced a pressure of around 2×10^{-6} mm Hg. and test vacuum system which had a test chamber whose pressure could be varied from 4×10^{-6} mm Hg. up to atmospheric

5. Romy, P. R. UCRL-11218, TID-4500.

6. M.K.S. Instruments, Inc., Burlington, Mass.

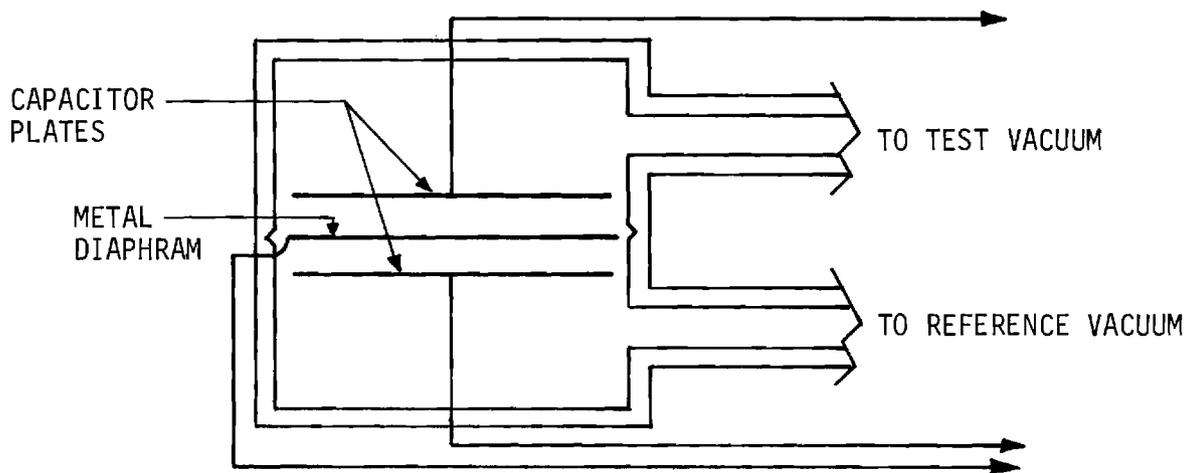


Figure 15. Schematic Diagram of Baratron Sensing Head.

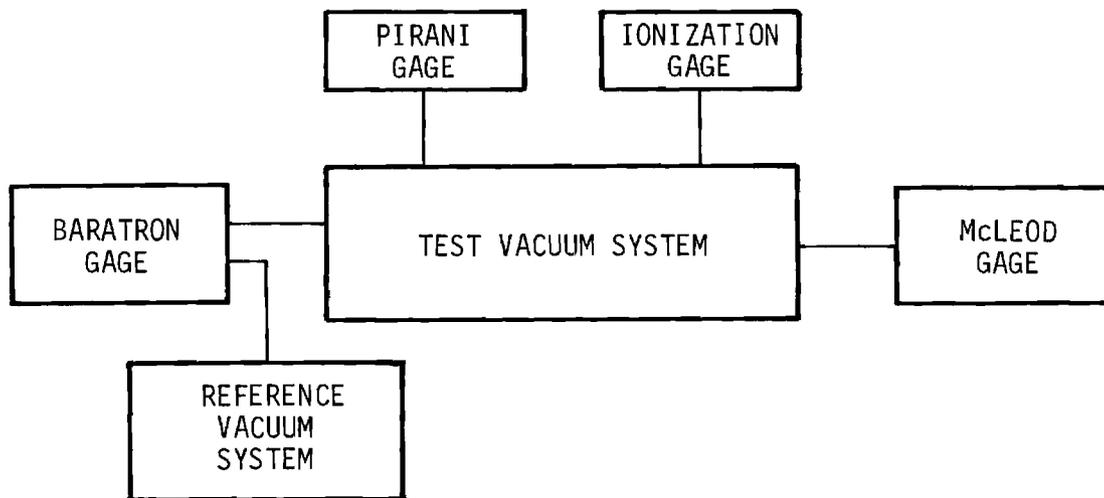


Figure 16. Block Diagram of Vacuum System Test Rig.

Progress Report

pressure by adjusting the gate valve and admitting gas into the test chamber through the needle valve. The gate valve was partially closed in attaining the higher pressures in order to keep the diffusion pump operating properly, since its pumping speed drops drastically if the pressure to which its port is exposed is above 10^{-3} mm Hg. The Baratron gauge and a standard, trapped McLeod gauge made by CVC, as well as an Edwards High Vacuum, Ltd., Pirani gauge and a Veeco ionization gauge, were connected into the test chamber.

As in our previous work on the pumping effect of the cold trap in the McLeod gauge, no precautions were taken to insure that this gauge was "clean", beyond baking the capillary tubes lightly to reduce sticking of the mercury columns.

A comparison was made between the readings of the McLeod and Baratron gauges in the pressure range 10^{-4} to 5×10^{-3} mm Hg. The McLeod gauge was operated in the variable compression mode^{4,7)} to reduce the effects of sticking, and capillary depression, while also providing a check that gas absorption on the capillary wall was insignificant. The Baratron was operated in its "direct reading" mode measuring the off balance conditions in the capacitance bridge, rather than using the electrical nulling of the bridge circuit, which is also provided. The manufacturer recommends this former procedure, as being most accurate for low pressure measurements.

Initially the operation of the Baratron was assessed from the point of view of its stability and drift characteristics. Utilizing the built in thermostatic temperature control, the short term zero drift was generally

7. Carr, P. H. Vacuum, 14, 37, (1963).

Progress Report

less than 10^{-5} mm Hg. Some difficulty was experienced with a long term drift which could not be accommodated by use of the fine zero control provided. As a last resort, use was made of the pre-set course zero control located at the pressure measuring head, a procedure not recommended by the manufacturer. Others⁸⁾ have adopted this course of action and agree with our observation that it does not seem to have an appreciable effect on the calibration accuracy. Some hysteresis effects were found when the device was subjected to a pressure differential of a few mm Hg. Zero continued to drift for an hour after the differential was removed, and eventually stabilized at a value, somewhat different from the original conditions. This effect was only appreciable on the most sensitive range of the device (3×10^{-4} mm F.S.D.).

Comparison of the Baratron with the McLeod gauge was carried out with He, Ar, O₂, N₂, gases providing the test pressure. Logarithmic plots of McLeod against Baratron reading are shown in figures 3, 4, 5, and 6. A perfect agreement of Baratron and McLeod would place the experimental points on a line of 45° slope, which is also indicated. It is noted that in all cases, agreement between the gauges from 5×10^{-3} to 5×10^{-4} mm Hg., is better than the nominal 5% accuracy of the McLeod gauge. Below this pressure, the McLeod tends to read higher than the Baratron. We do not attach great significance to this since the accuracy of both gauges becomes poor. 10% might be considered as a conservative estimate for both gauges at 10^{-4} mm.

8. Barnett, C. F. private communication.

Progress Report

It is significant that all the calibrations agree exactly, independent of gas. If the mercury pumping effect had been appreciable one would have expected the McLeod and Baratron measurements to disagree by about 10% for the heavier gases, N₂, O₂, and Ar, whilst agreeing exactly only for helium, where the effect is expected to be negligible. In accordance with our previous observation⁴⁾ we again must conclude that the mercury pumping effect is negligible, and repeat our contention that contaminated mercury surfaces might be the cause of this.

We conclude that the Baratron is an excellent device for pressure measurement, and the calibration may be relied on down to 5×10^{-4} mm Hg. We again find evidence supporting our previous contention that the mercury pumping error in the McLeod gauges we are using is small, perhaps due to contamination of the mercury surface in the columns. Evidence in support of the error continues to be published, in particular a recent paper by Ulterback⁹⁾ which essentially utilizes the same procedure as in this work. This conflict of conclusions serves to confirm that the McLeod gauge is a difficult device to use properly. It is generally agreed^{5,8,9)} that the Baratron behaves perfectly when used with helium, where the pumping error is expected to be negligible. Since the capacitance monometer should be insensitive to the type of gas being used, we would conclude that it may also be utilized for other heavier gases with equal precision. We have no hesitation in utilizing the device for measurement of pressures above 5×10^{-4} mm Hg., and we intend to utilize it instead of the McLeod gauge for all further work in this program.

9. Ulterback, N. G. and Griffith, T. Review of Scientific Instruments, 37, 866, (1966).

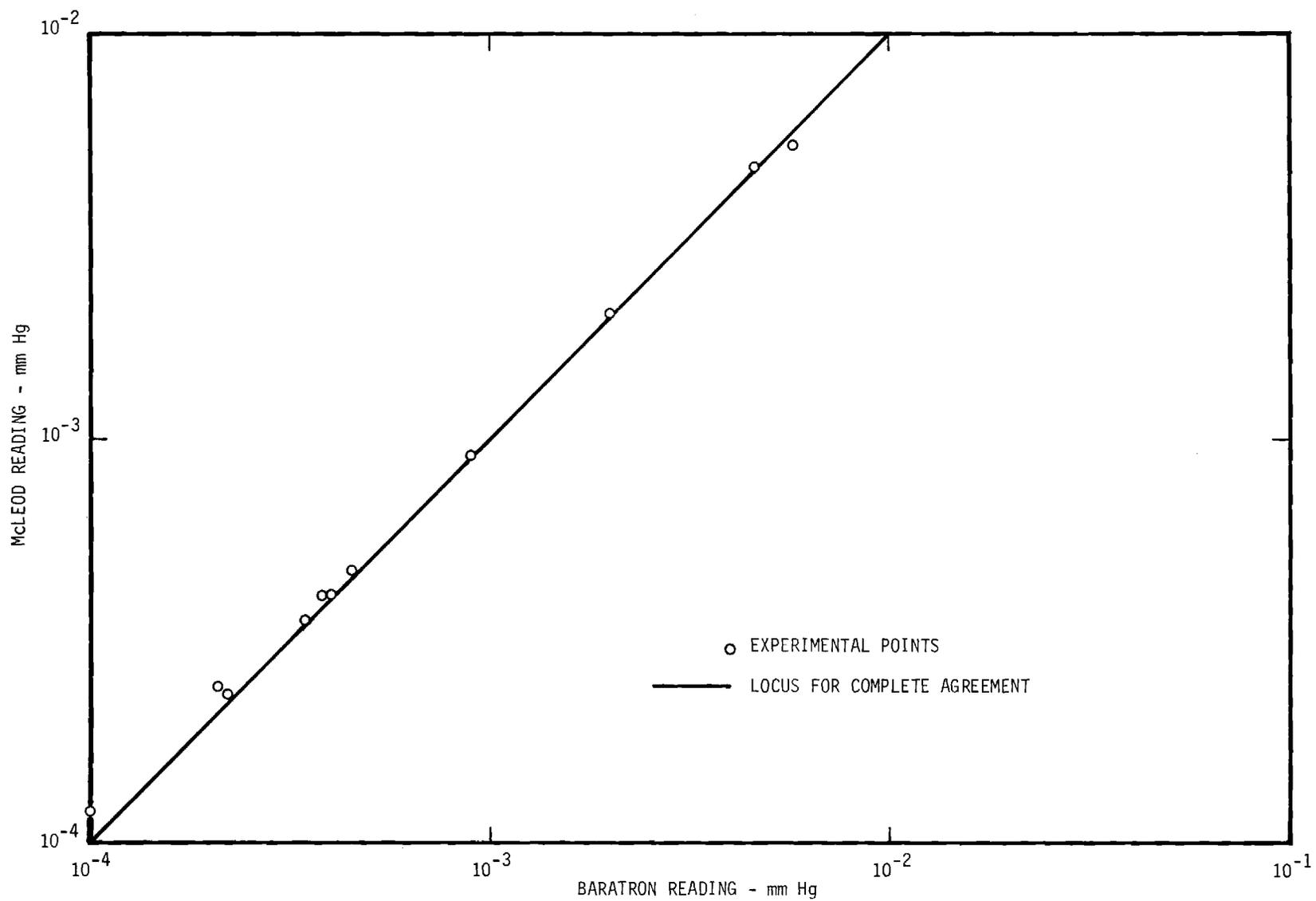


Figure 17. Comparison of McLeod and Baratron Gauge Readings for Helium.

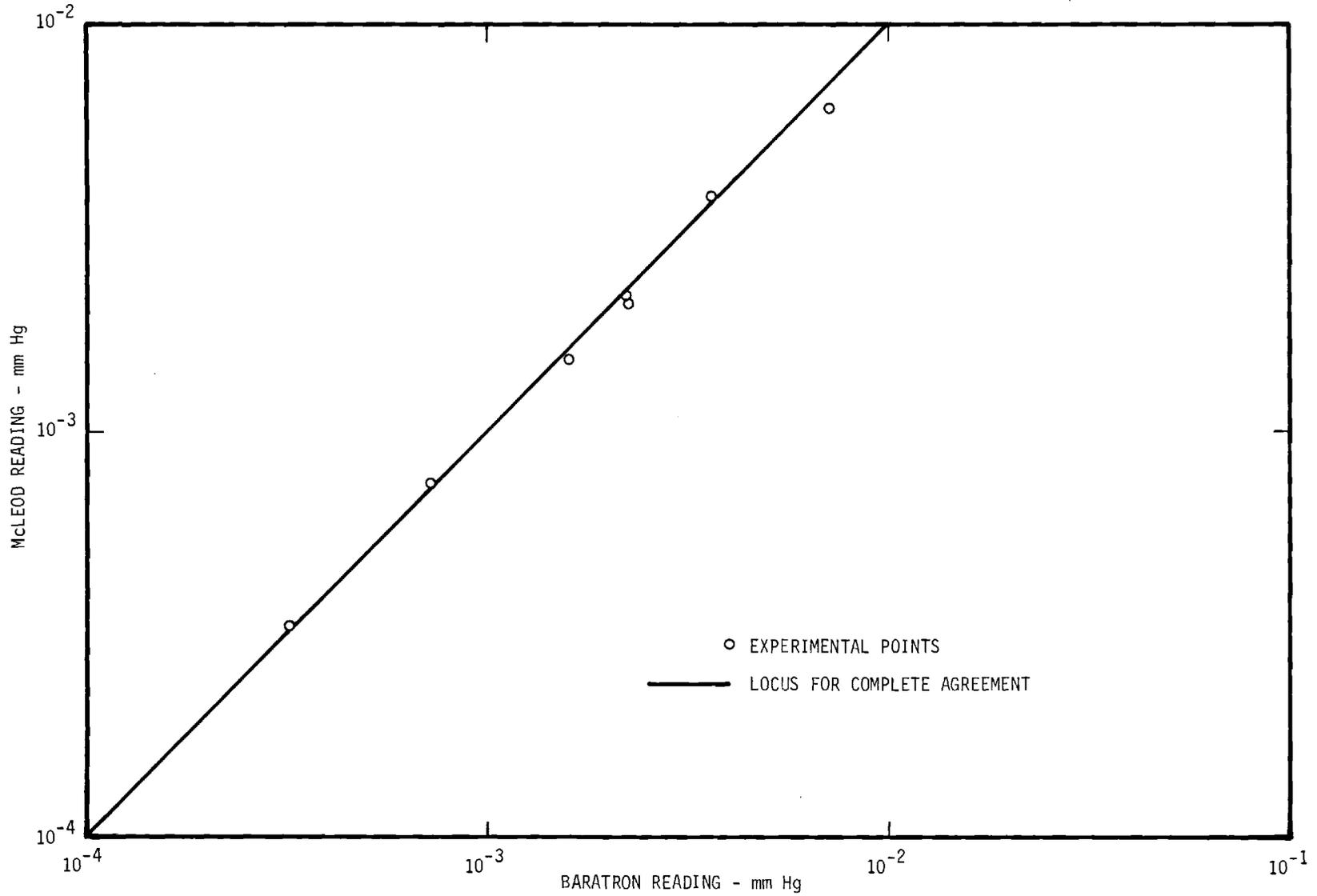


Figure 18. Comparison of McLeod and Baratron Gauge Readings for Argon.

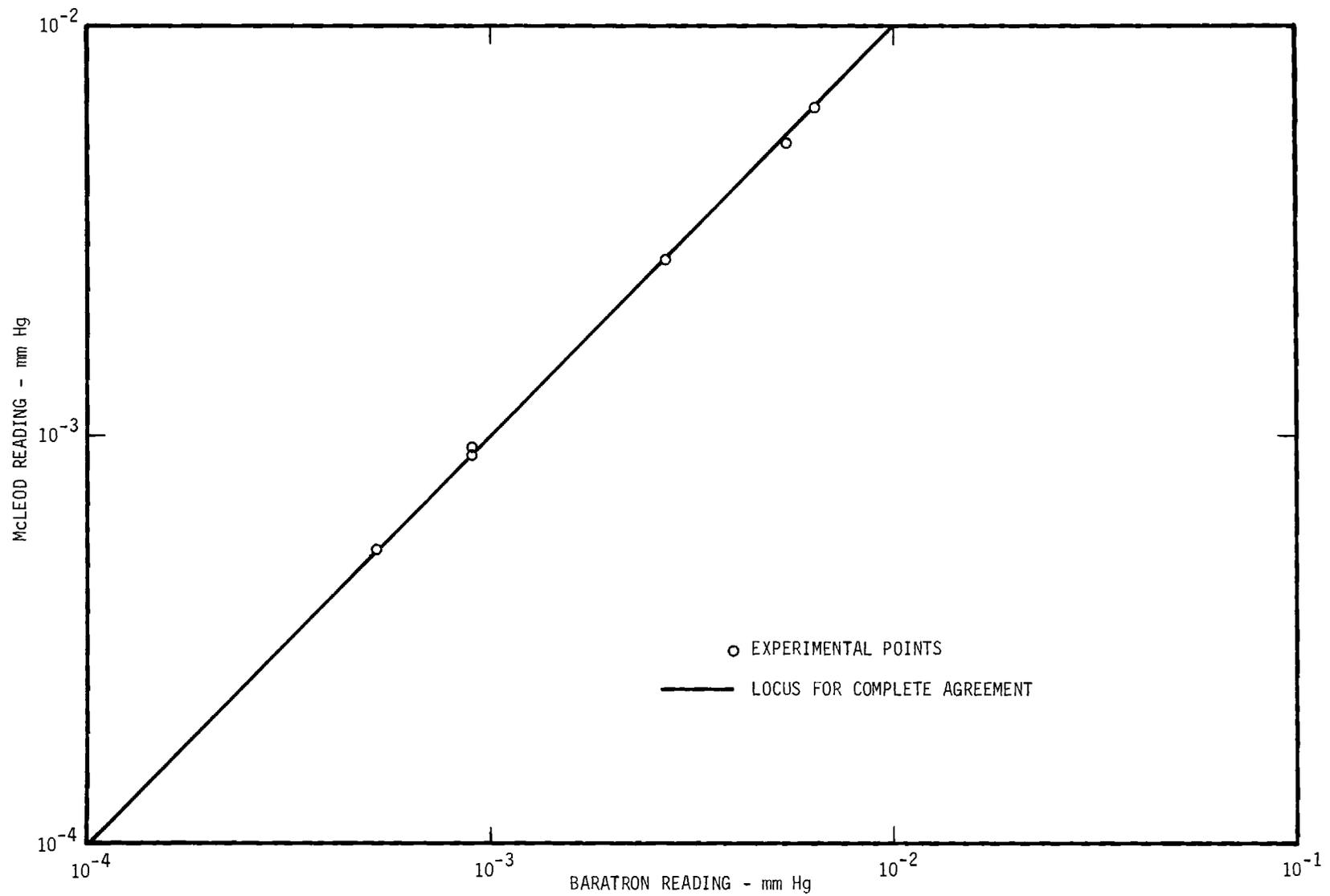


Figure 19. Comparison of McLeod and Baratron Gauge Readings for Nitrogen.

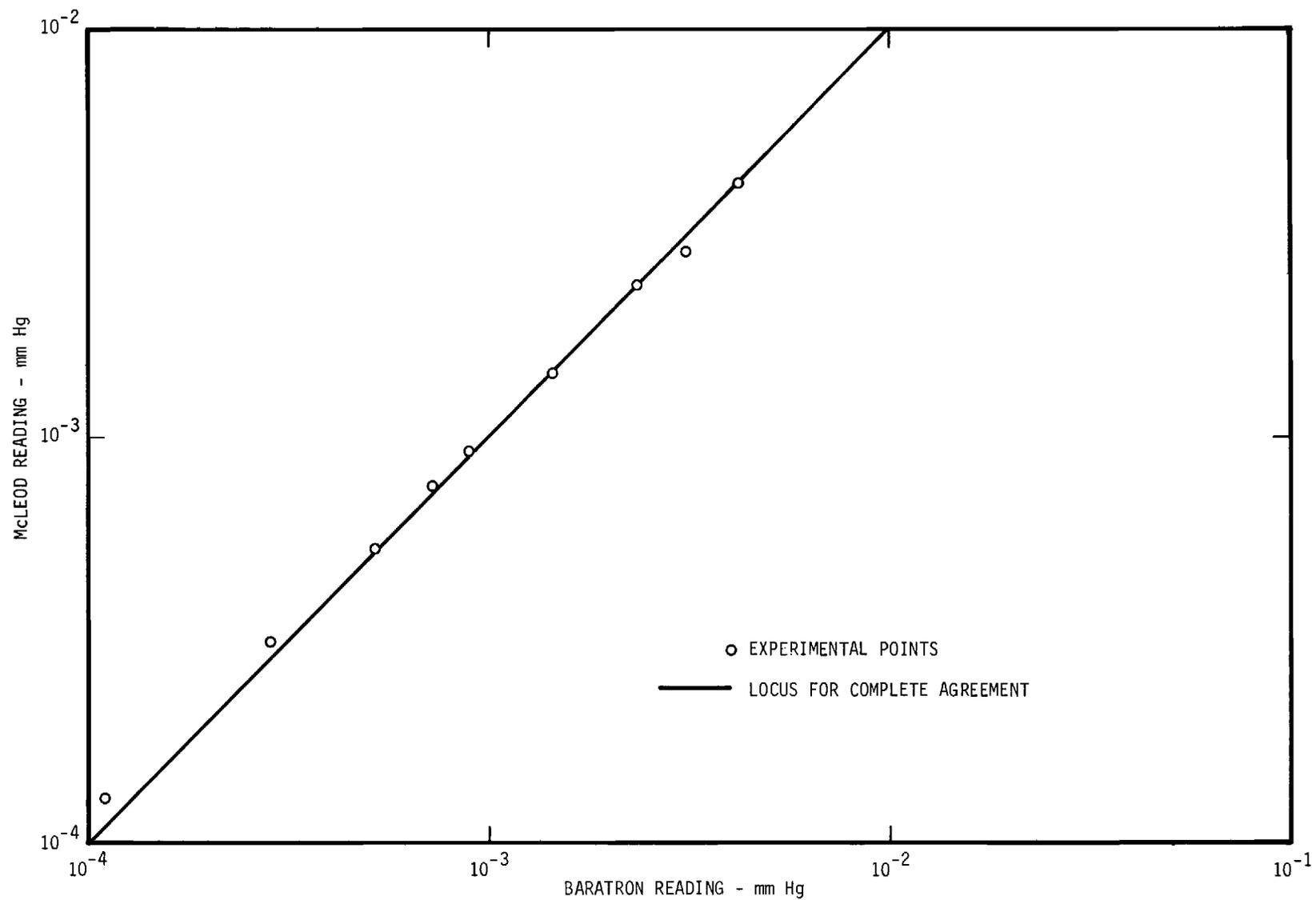


Figure 20. Comparison of McLeod and Baratron Gauge Readings for Oxygen.

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ORO-2591-33

EMISSION AND EXCITATION
CROSS SECTIONS

PROGRESS REPORT NO. 3

Covering the Period

December 1, 1966 to November 30, 1967

By E. W. Thomas

J. L. Edwards

G. D. Bent



Report No. ORO-2591-33

Contract No. AT-(40-1)2591

U. S. ATOMIC ENERGY COMMISSION

OAK RIDGE, TENNESSEE

1 December



School of Physics
GEORGIA INSTITUTE OF TECHNOLOGY
Atlanta, Georgia

ORO-2591-33

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~~30 November 1967~~
1 December 1967

Progress Report

TABLE OF CONTENTS

	<u>Page</u>
<u>List of Figures</u>	iii
I. <u>Title</u>	1
II. <u>Introduction</u>	1
III. <u>Objective and Method</u>	1
IV. <u>Summary of Progress Made During This Reporting Period</u> . . .	6
V. <u>Formation of Excited States in the Target</u>	9
(i) The Excitation of Helium	9
(ii) The Excitation of Nitrogen	11
(iii) The Excitation of Oxygen	13
(iv) The Excitation of Hydrogen	13
(v) The Excitation of Argon	14
(vi) Conclusions	17
VI. <u>Charge Transfer Studies</u>	22
(i) The Experimental System	25
(ii) Experimental Results	25
(iii) Problems of Data Handling and Analysis	29
VII. <u>Program for the Remainder of the Contract Year</u>	31
VIII. <u>Program for the Future</u>	33
IX. <u>Publications and Travel</u>	34
X. <u>Personnel</u>	37
XI. <u>Accelerator Usage</u>	37
XII. <u>Incident Report</u>	38

Progress Report

LIST OF FIGURES

<u>Figure</u>		<u>Page</u>
1.	Emission Cross Sections for the ArI 4200 A and ArII 4431 A Lines Induced by the Impact of Protons on Argon	16
2.	Balmer Alpha Emission Cross Section in Arbitrary Units Shown as a Function of Penetration Through the Target Divided by Projectile Velocity	27

Progress Report No. 3, Project No. B-2021

I. Title

Emission and Excitation Cross Sections

II. Introduction

This report summarizes the work performed on excitation phenomena as part of contract AT-(40-1)-2591 for the U.S. Atomic Energy Commission. The work was initiated on March 1, 1965, as a result of Modification No. 7, increasing the scope and financing of the existing contract. The present report covers the period 1 December 1966 to 30 November 1967 which corresponds to the first 9 months of the 12 month period covered by Modification No. 9 to this contract, plus the final three months of the preceding contract period.

A program for the measurement of ionization cross sections which was the subject of the original contract and subsequent Modifications 1 through 9 has been continued and is reported separately.¹

III. Objective and Method

The objective of this research is the measurement of cross sections for production of particles in excited states as the result of the impact of protons on various gaseous targets under single collision conditions. The energy of the incident particles ranges from 0.15 to 1.0 MeV. A prime objective in the present program is the acquisition of a body of data which can be used (a) for comparison with accurate quantum mechanical calculations where these are available, or (b) in the more complex situations, to indicate systematics in behavior useful in predicting

1. D. W. Martin, L. J. Puckett, and G. O. Taylor. "Ionization and Charge Transfer Cross Sections", Progress Report No. 20. ~~30 November~~ 1967. U.S. Atomic Energy Commission Report Number ORO-2591-34.

1 December

Progress Report

basic characteristics and in assisting with the formulation of a phenomenological basis for semi-empirical and statistical theories. It is of the utmost importance that the approximations made in theoretical predictions should be tested at every available opportunity in order to establish the validity of calculations in situations where experimental verification is at the present time impossible for technical reasons. A second objective is the provision of data which is of direct value for design and diagnostic applications in thermonuclear, upper atmospheric, and other practical situations.

In general, the atomic and molecular processes which occur when fast atoms or ions collide with the molecules of the target gas, involve the rearrangement of the electrons associated with the two systems. We may use the term "charge rearrangement" to refer to cases where electrons are ejected from either of the colliding systems or transferred between them. Charge rearrangement may be investigated by observing the change of the ionic composition of the fast beam and the target region. In addition to the gross removal or transfer of charge, there is a probability that one or more of the electrons remaining bound to the nucleus after the collision may be in an excited state. Quantitative measurement of the photon flux emitted as a result of subsequent decay to the ground state allows the determination of the cross section for the formation of excited states. Clearly such a technique cannot be used for investigating the formation of metastable or other long-lived states, and these are not considered in the present work.

Progress Report

The source of the energetic protons is a 1 MeV Van de Graaff positive ion accelerator, which is equipped with a beam analyzing and stabilizing system. The ion beam is collimated by a series of small apertures, traverses a collision chamber containing the target gas, and is monitored on a Faraday cup. Quartz windows situated in the walls of the chamber allow spectral analysis and detection of emission from the collision region. The chamber dimensions and gas pressure are such that the target is "thin" in the sense that only a small fraction of the incident particles undergo any collisions at all. Also the path traveled by the photons before leaving the gas-filled collision chamber has been arranged to be short to reduce absorption.

In general, separate techniques must be utilized for measuring the formation of excited states in the static target and in the fast beam. In the former case the excited target atom will acquire only a small momentum as a result of the collision and will decay to the ground state with the emission of photons at essentially the same point as it was excited. However in the case of the fast incident particles, the velocities used in the present work are so high that the excited particle will move an appreciable distance during the finite lifetime of the excited state. Consequently, emission from the fast particle will vary with the position of observation in the collision chamber. In either case the measurement technique consists of determining the rate of photon emission per unit length of the ion beam path in the collision chamber, per unit incident beam flux, per unit target density. This apparent cross section for the emission of a photon of a particular wavelength will be dependent on the

Progress Report

position of observation in the case of the excited fast particles, but in the case of target molecules will be invariant with respect to position along the beam.

For the measurement of the formation of excited target particles, the photons emitted from a definite fixed part of the ion beam path in the chamber are focused onto the entrance slit of a grating spectrometer, by a simple lens. A low noise photomultiplier at the exit slit serves to detect the photons in the dispersed spectrum. Enhancement of the signal-to-noise ratio is obtained by chopping the light signal with a vibrating reed before it enters the spectrometer and applying a narrow band, phase sensitive detection system to the output of the photomultiplier. The determination of the detection efficiency of the system is made quite simply by substituting a source of emission of known output in place of the collision experiment. The ratio of the emissions of the experiment and the standard will be equal to the ratio of observed signals. It may be shown that when using a substitution technique where the standard is of the same geometrical shape as the source of experimental emission, then knowledge of the actual length of particle beam path observed by the detection equipment, and of the solid angle from which light is collected, is unimportant. The emission cross section is the cross section for a single depopulating transition. Any one excited state will in general have a number of possible paths by which it may decay radiatively, and, in addition, there will be a series of cascade populations of the level due to transitions from higher excited states. In principle, if the emission functions for all the radiative populating

Progress Report

and depopulating transitions associated with a particular level are known, then the cross section for the formation of the excited state may be determined. In certain cases knowledge of the relevant transition probabilities allows the determination of an excitation cross section with the measurement of only a small number of emission cross sections.

It may be shown that the emission from excited fast particles will build up exponentially toward some asymptotic value as the beam traverses the chamber. If one again defines the cross section for photon emission in the same way as for target excitation this quantity will now vary with penetration x through the chamber. Neglecting for the moment cascade transitions, if Q_j is the cross section for the collisional excitation of the state j , A_{jk} is the transition probability for the $j \rightarrow k$ decay, and $\sum_k A_{jk}$ is the sum of all the radiative decay transition probabilities out of the level j to all levels k , then the emission cross section will vary as

$$Q_{jk} = Q_j \frac{A_{jk}}{\sum_{k < j} A_{jk}} \left(1 - \exp - \frac{x}{v\tau}\right) \quad (1)$$

Here v is the velocity of the projectile and τ the lifetime of the excited state. Measurement of the emission cross section at one or more values of x , with knowledge of the transition probabilities and lifetime, allows the evaluation of Q_j . The problem of assessing cascade contributions may be severe and will have to be treated separately for each specific case.

Progress Report

The detection system designed for this phase of the program is based on a photomultiplier fitted with a suitable interference filter to transmit only the emission of interest. This detector system views the collision region through a long side window and may be moved to allow determination of the emission at various beam penetrations through the target. Pulse counting techniques are used and the effects of the thermal noise of the photomultiplier reduced by cooling the photocathode.

The measurement of emission from the fast beam particle is designed to investigate the process of charge transfer leaving the fast particle in an excited state. The process of most interest here is the formation of excited hydrogen atoms in the projectile beam. The Balmer alpha emission is produced by three separate decays from the 3s, 3p and 3d states into the 2s and 2p levels. Since the states of the same principal quantum number are essentially degenerate in energy it is impossible to separate the contributions from these states by normal spectroscopic means. However, each of the upper states has a distinct lifetime and therefore the variation of emission cross section with penetration through the chamber is given by the sum of three expressions of the type given in equation (1) with the three appropriate lifetimes. In principle it is possible to fit such an equation to the observed variation of emission with penetration and so to determine the three separate cross sections for exciting the 3s, 3p and 3d states.

IV. Summary of Progress Made During This Reporting Period

The objectives of the work proposed for the period covered by this report were twofold. The extended program of measurements on the formation

Progress Report

of excited states in the target particles was to be completed and brought to a conclusion. The experimental system for investigation of charge transfer processes leading to the formation of excited fast particles was to be constructed and brought into operation. These two experiments are separate in many respects, although being based on the use of essentially the same hardware. We will summarize briefly below the progress which has been made on these two efforts and indicate what data we will discuss in this present report.

The work on the investigation of the formation of excited target structures has been completed, analyzed, and submitted for publication. We list below the titles and status of eight reports that have been written on this work during the period of the present contract.

- (a) "The Excitation of Molecular Nitrogen and Oxygen by the Impact of 0.15 to 1.0 MeV Protons", by E. W. Thomas, G. D. Bent, and J. L. Edwards. Published in the Proceedings of the Fifth International Conference on the Physics of Electronic and Atomic Collisions. (Pub. Nauka Publishing House, Leningrad, 1967) page 286.
AEC Report Number ORO-2591-24.
- (b) "Balmer Emissions from a Molecular Hydrogen Target Induced by the Impact of Fast Protons", by J. L. Edwards and E. W. Thomas.
Ibid. p. 288. AEC Report Number ORO-2591-25.
- (c) "A Comparison of the Excitation Produced in a Helium Target by the Impact of H^+ , D^+ , H_2^+ , H_3^+ Ions at High Energy", by E. W. Thomas and G. D. Bent. (Scheduled for publication in J. Opt. Soc. Am. December 1967). AEC Report Number ORO-2591-27.

Progress Report

- (d) "The Formation of Excited States in a Helium Target by the Impact of 0.15 to 1.0 MeV Protons and Deuterons. I Experimental", by E. W. Thomas and G. D. Bent. (Scheduled for publication in the Physical Review 164, Dec. 5, 1967). AEC Report Number ORO-2591-28.
- (e) "The Formation of Excited States in a Helium Target by the Impact of 0.15 to 1.0 MeV Protons and Deuterons. II Comparison with Theory", by E. W. Thomas. (Scheduled for publication in the Physical Review 164, Dec. 5, 1967). AEC Report Number ORO-2591-29.
- (f) "Cross Sections for the Formation of Excited States in a Nitrogen Target by the Impact of 0.15 to 1.0 MeV Protons", by E. W. Thomas, G. D. Bent and J. L. Edwards. (Accepted for publication in the Physical Review). AEC Report Number ORO-2591-30.
- (g) "Emissions from a Molecular Hydrogen Target Induced by the Impact of Fast Protons", by J. L. Edwards and E. W. Thomas. (Submitted to the Physical Review). AEC Report Number ORO-2591-31.
- (h) "The Formation of Excited Ions by the Impact of 0.15 to 1.0 MeV Protons on a Molecular Oxygen Target", by E. W. Thomas and G. D. Bent. (Submitted for publication in Proc. Phys. Soc. London). AEC Report Number ORO-2591-32.

These reports fully detail the experimental procedures that have been employed in this work and also include the vast proportion of the experimental measurements. In Section V of the present report we will briefly discuss certain additional observations that have been made, but which are generally of poor quality and will not be submitted for publication.

Progress Report

We will not discuss the work contained in the reports listed above neither will we repeat the discussion of experimental techniques which has been covered in previous reports in this series.² We now consider this part of the program completed and do not anticipate any further studies of target excitation.

A preliminary study has been carried out on the formation of excited states in the projectile beam by charge transfer as protons traverse various gaseous targets. The capabilities of the equipment have been assessed and areas indicated where improvements might usefully be made. It will be possible to make charge transfer measurements for protons incident on heavy targets over the full energy range available from the accelerator. However in the case of helium target it would appear that low signals and noise problems will limit the upper energy to approximately 600 keV. In Section VI of this report we discuss in detail the present status of the experiment and display certain preliminary measurements.

V. The Formation of Excited States in the Target

(i) The Excitation of Helium

The formation of excited states in a helium target by the impact of protons has been considered at some length and two papers prepared,

2. E. W. Thomas, J. L. Edwards, and G. D. Bent. "Emission and Excitation Cross Sections", Progress Report No. 3, 30 November 1967. U.S. AEC Report Number ORO-2591-23

Progress Report

one dealing with the experimental measurements,³ and the other to compare the predictions of theory with these experimental results.⁴ A brief study has been made on the relative efficiencies of H^+ , D^+ , H_2^+ and H_3^+ ions in exciting the helium atom.⁵

According to the Wigner spin conservation rule the impact of protons on helium in the singlet ground state, should not result in the formation of triplet states. Clearly the impact of electrons or neutral hydrogen can cause a transition from the singlet ground state of helium to a triplet excited state through the mechanism of exchange. There is some interest in attempting to explore quite how important the Wigner spin conservation rule is in governing the transitions which can take place as a result of a collision. An attempt was made to detect the emission of the HeI $3^3P - 2^3S$ (3889 A) line induced by the impact of protons on a helium target. A very weak emission of this line was observed but investigations of the intensity of emission as a function of target pressure indicated that the source was not the direct excitation process. This suggested that the excitation was caused either by a neutral hydrogen atom formed by a previous charge changing collision in the target chamber or by the impact of an electron which had resulted from the ionization of a target atom in a previous collision. Reference

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3. E. W. Thomas, G. D. Bent. Phys. Rev. 164, Dec. 1967.
 4. E. W. Thomas. Phys. Rev. 164, Dec. 1967.
 5. E. W. Thomas and G. D. Bent. J. Opt. Soc. Am. (To be published) December 1967.

Progress Report

to existing charge transfer data and also to the measurements by Van Eck et al.⁶ on the cross sections for the excitation of helium by neutral atom impact, indicated that the impact of neutral hydrogen atoms could not explain the major part of the observed triplet emission. The secondary electron hypothesis may be assessed using the measurements of Rudd et al.⁷ of the cross sections for producing electrons of various kinetic energies by ionization of the target, and also the various direct measurements of electron excitation cross sections. It is found that the observed emission from the triplet state is consistent with the explanation that the excitation is caused by electrons ejected from target atoms in a previous collision. It was not possible to measure the cross section for the direct excitation of the 3^3P state. We estimate that an upper limit to the cross section is 10^{-21} cm². In connection with this attempt to assess the importance of spin conservation in governing the possible transitions a similar investigation was carried out using a nitrogen target and is reported in the next section.

(ii) The Excitation of Nitrogen

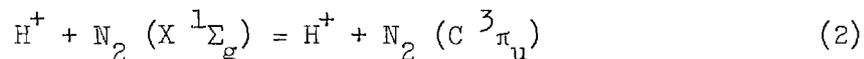
An extended series of measurements were made on the formation of various excited states of the molecular ion N_2^+ and the emission functions of certain lines from the atomic ion N^+ . These were partially reported

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6. J. Van Eck, F. J. De Heer and J. Kistemaker. *Physica* 30, 1171, 1964.
 7. M. E. Rudd, C. A. Sautter and C. L. Bailey. *Phys. Rev.* 151, 20, 1966

Progress Report

in the proceedings of a conference⁸ and have been fully reported for formal publication.⁹

Part of this program included an attempt to investigate the importance of the Wigner spin conservation rule in this particular collision combination. The ground state of the N_2 molecule is singlet. The emissions of the N_2 second positive system arise from an electronic state which has a triplet character. An attempt was made to study the following mechanism which is formally forbidden.



Emission of the 3371 Å ($v' = 0 \rightarrow v'' = 0$) line was quite strong and the dependence on target pressure indicated that both the direct process described by equation (2) and also some secondary processes were present. As in our previous investigation of a forbidden transition in helium which was described above, the secondary process was believed to be excitation by electrons ejected from target atoms by a previous ionizing collision. However in this case it was possible to measure the cross section for the process described by equation (2) and it was found to be some three orders of magnitude less than that due to electrons of the same impact velocity.

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8. E. W. Thomas, G. D. Bent and J. L. Edwards. Proc. Fifth Int. Conf. on the Physics of Electronic and Atomic Collisions. (Pub. Nauka Publishing House, Leningrad 1967) Page 286.
9. E. W. Thomas and G. D. Bent. Phys. Rev. (To be published).

Progress Report

This result was taken as indicating the importance of spin conservation in governing the mechanisms which occur during the collision. Clearly in the case of the complex molecular nitrogen target, violation of the spin conservation rule is more likely than for the helium target.

(iii) The Excitation of Oxygen

As a parallel to the measurements on the nitrogen target some consideration was also given to oxygen where similar processes are expected. The major experimental difficulty with the emission in this case is that the rotational structure of the O_2^+ emissions extends over a considerable spectral range. Resort was made to rather unsatisfactory integrating procedures to arrive at a measurement of absolute cross sections for the excitation of certain levels. This work has been prepared for publication.¹⁰

(iv) The Excitation of Hydrogen

A brief program of investigation was carried out on the formation of various excited states as the result of proton impact on a molecular hydrogen target. This is of considerable interest since the molecule is the simplest possible neutral molecule and there is some possibility of coming to a theoretical understanding of the processes which occur during proton impact. Of particular interest is the excitation of the molecular states. Measurement of these cross sections is difficult due to the low signals and the very considerable complexity of the H_2 emission spectrum. However it was possible to determine emission cross section for two spectral lines. Both transitions were identified as being due to dipole forbidden transitions and were shown to exhibit the behavior predicted for such a

10. E. W. Thomas and G. D. Bent. Submitted to Proc. Phys. Soc. (London).

Progress Report

transition in a simple atomic target.

The work on the hydrogen target included both molecular and atomic emissions. This work has been prepared for publication.¹¹

(v) The Excitation of Argon

The spectrum emitted as a result of the impact of protons on an argon target is exceedingly complex, and due to the low signal levels it proved impossible to utilize sufficient resolution to separate all the various emissions. It was intended to make measurements of the emission cross sections for transitions in the neutral and ionized argon systems. In practice it was not possible to make positive identification of emissions from Ar^{2+} or more highly ionized systems. Such emissions, if they occur at all, are of very low intensity. In the present program we were therefore restricted to measurements on the ArI and ArII spectra.

In figure 1 are shown the emission cross sections for the ArI 4200 Å $5p_2 - 4s_1$ (Paschen Notation) transition, and the ArII 4431 Å $4p^4 P^0_{2\frac{1}{2}} - 3d^4 D_{2\frac{1}{2}}$ transition. Although it is not possible to derive cross sections for the excitation of levels from these data it is expected that cascade is relatively small and that the functions shown here are proportional to the cross sections for exciting the parent level of the transition. In all cases the accuracy of the measurements are within $\pm 25\%$.

There are no theoretical predictions with which these measurements may be compared, and no other published data for proton impact excitation at these high energies. It would seem clear that the formation of the

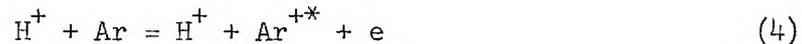
11. J. L. Edwards and E. W. Thomas. Submitted to Phys. Rev.

Progress Report

excited neutral should be by the process:-



The formation of the excited ion might be caused by charge transfer although at these high energies this must be considered unlikely. It seems most probable that the relevant process is the following



It is also possible that the excited Ar^+ ion may be caused by a process of the type shown in equation (3) giving rise to the formation of an auto-ionizing state which subsequently decays with the ejection of an electron. It is impossible to determine whether the autoionizing mechanism is important without some complex coincidence experiment.

In a report¹² of work carried out under the same contract as the excitation work reported here, there are presented some very preliminary measurements on the total cross section for the formation of argon ions of various charge states in the target. It is to be expected that the cross sections for the formation of an excited neutral atom and for the formation of the singly ionized particle in its ground state should be similar since each involves the excitation of one electron, the former

12. L. J. Puckett, D. W. Martin and G. O. Taylor. "Ionization and Charge Transfer Cross Sections for H^0 , He^0 and He^{2+} beams in the energy range 0.15 to 1.0 MeV". Technical Report, December 1967. AEC Report Number ORO-2591-35.

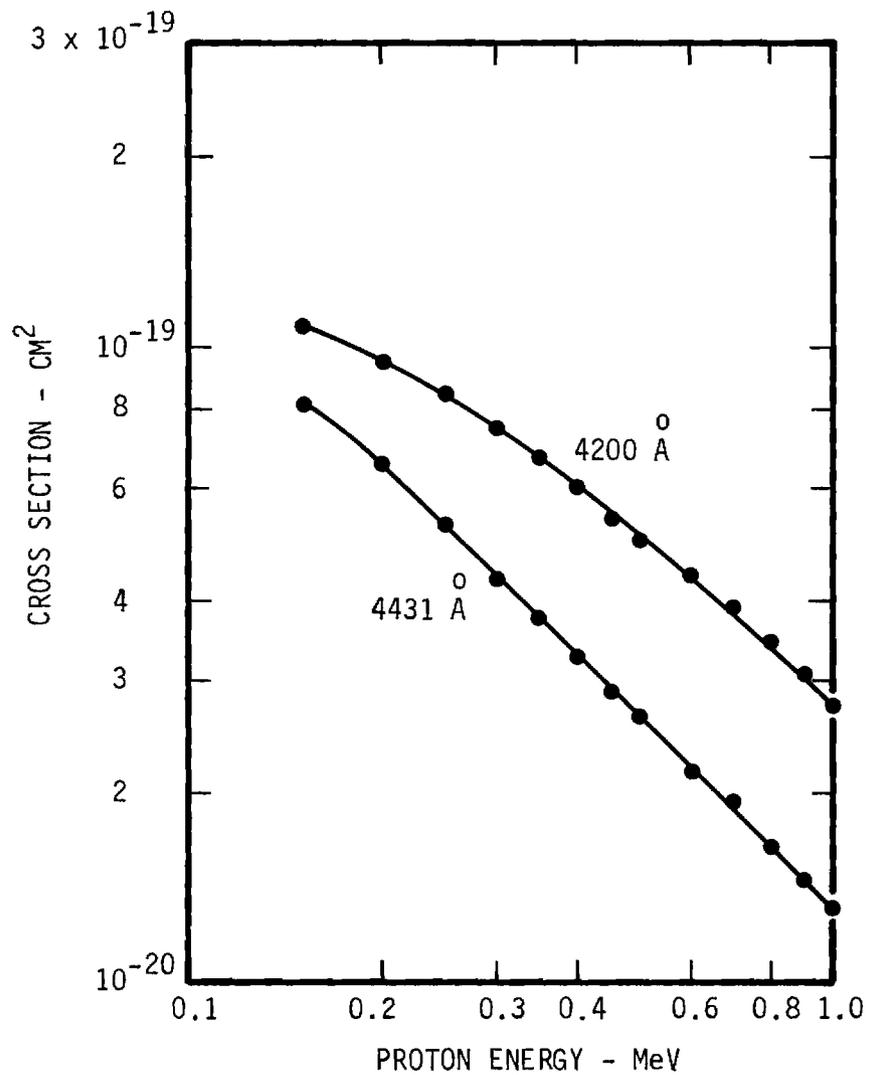


Figure 1. Emission Cross Sections for the ArI 4200 Å and ArII 4431 Å Lines Induced by the Impact of Protons on Argon.

Progress Report

to a bound state the latter to an unbound continuum state. Although the ionization data are very preliminary there does seem to be a similar behavior. By the same argument the formation of the excited Ar^+ ion and the removal of two electrons from the target atom are again expected to be similar and the ionization measurements support this contention. This type of correspondence between excitation and ionization data was first recognized by us in the comparison of data on the ionization of helium with the corresponding data for excitation.⁴

(vi) Conclusions

It is valuable to summarize and consolidate the broad conclusions which may be drawn from the results of this series of measurements. The various parts of the program have been fully reported in separate publications but there has been little opportunity to consider how these measurements effect our overall knowledge of collisional processes.

The major objective and value of the work on helium was that comparisons would be made with theoretical predictions. Our work showed for the first time that the high energy behavior of dipole allowed and dipole forbidden transitions was in accord with the general predictions of the Bethe Born theory. This is of considerable general importance since the Bethe Born formulation predicts the general high energy behavior of any single electron transition induced by proton or electron impact. Although the "dipole allowed" type of transition has been well investigated through the measurement of total ionization cross section, the "quadrupole allowed" transition has never previously been studied under proton impact, and only rarely with electrons. It is generally expected that the

Progress Report

asymptotic high energy behavior for a single electron transition will be the same for both protons and electrons of equal velocity, this in itself being a prediction of the Bethe Born theory. However it is impossible to predict the velocity range where this equality should be observed. The present work showed quite conclusively that in the "quadrupole allowed" transition, such as the excitation of the 1S and 1D states, the agreement of proton and electron impact data did not necessarily occur at the same impact velocity for each multiplet system, and indeed for the 1S system did not occur in the energy range available to the present experiment. In general we observed that for the excitation of the 1P and 1D levels, electron and proton impact excitation measurements on an equal velocity scale would agree at proton energies above 400 keV. However for the 1S levels there is no correspondence at the particle energies available to us (up to 1 MeV protons). It would seem that for the excitation of the 1S state, the Bethe Born formulation agrees with proton impact measurements down to 450 keV, but disagrees substantially with the electron impact data. Naturally it is necessary that those differences should be predicted by any successful theoretical treatment of excitation by proton and electron impact.

Comparison of the measured cross sections for excitation of helium with accurate theoretical predictions was reasonably successful. There was good agreement with distortion approximation calculations for the 1P levels at all energies available to the present experiment. For the 1S and 1D levels the theoretical calculations are very sensitive to the choice of ground state wave function and little agreement was found between

Progress Report

the present experiment and the available theoretical predictions or indeed among the theoretical predictions themselves. It is hoped that the availability of the present data will stimulate a concerted attempt to produce an accurate calculation of the cross sections for "quadrupole allowed" transitions.

Our brief investigation of the excitation of the H_2 molecule was carried out with the objective of indicating whether there was any similarity of behavior between transitions in the H_2 molecule and transitions in the He atom. Each molecular energy level will correspond to a energy level in the "united atom" limit which is obtained as the separation of the atoms is figuratively reduced to zero and the molecule coalesces to become a helium atom. Despite the practical difficulties in dealing with the emission of the radiation from the H_2 molecule we were able to show that as far as the general behavior of the cross sections are concerned there does indeed seem to be strong evidence that H_2 and He behave in a similar manner. This conclusion may be of general value in prompting the application of results known for the relatively simple case of He to the more complex and less well studied case of the H_2 molecule. For example our work indicates that the concept of dipole allowed and quadrupole allowed transitions may be applicable for the H_2 molecule and therefore the nature of the high energy cross section variation may be predicted by the Bethe Born approximation.

Some attention was applied to the consideration of the importance of the Wigner spin conservation rule in governing the transitions which take place as a result of a particular collision. Specifically this rule

Progress Report

would imply that a proton impact could not excite a system from a singlet ground state to a triplet state. In the case of electron impact the rule may still be expected to hold but there is the possibility of exciting the triplet states by an exchange process. In the excitation of He and H₂ by protons the cross sections for the forbidden spin changing collisions were too small to be measured. In the case of the N₂ target the cross sections for the spin changing collision was exceedingly small being approximately three orders of magnitude less than those for electron impact. These results indicate that spin conservation is a very important selection rule governing the transitions which may result from a particular collision.

The measurements on the formation of excited states in a heavy molecular target of nitrogen or oxygen cannot readily be related to theory. We observed in general that proton impact gave rise only to substantial emissions from the excited molecular ion, N₂⁺ and O₂⁺. The formation of excited products of dissociation, or excitation of the neutral molecule was very much less important. It was found that a very substantial proportion of all the N₂⁺ and O₂⁺ ions formed in ionization are produced in the excited state, at least 5% just on the basis of the present limited measurements. It was observed that this proportion was constant over the energy range of the present experiments. Comparison of the cross sections for exciting various levels of the molecular ion indicated that the relative population of excited states was predicted quite accurately by the relative values of the Franck Condon factors, or overlap integrals. Since these are known accurately from

Progress Report

molecular emission spectra, they provide a ready tool for calculating excitation cross sections on a relative basis.

Apart from the general contributions to the understanding of collisional excitation phenomena it is hoped that the present data will have some direct practical applications. The measurement of photon emission from a complex situation such as a plasma or the upper atmosphere provides a ready method of determining what processes and atomic species are present. Our work has very clear applications to the analysis of auroral emissions both from the upper atmosphere of the earth as well as from other planets. Duclos¹³ has suggested a system for analyzing the particle distribution in a plasma by projecting a beam of protons through the region of interest and analyzing the spatial distribution of emission. The present measurements provide information which could be used for measuring the density of neutral atoms in such an experiment. Determination of the ion density by this method is clearly more difficult since there is no direct information on the cross sections for the excitation of ions by protons. In such a case it is possible to calculate the required cross sections utilizing theoretical techniques whose accuracy has been verified by experiments of the type carried out under this program. It would also be possible to determine a proton density by firing a beam of fast neutral atoms across the plasma region and relating the emission to the cross sections determined in the present work. Clearly the cross section is dependent only on relative velocity of impact and not on which of the two particles is technically the "projectile".

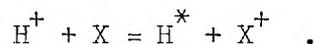
13. D. P. Duclos. Rev. Sci. Inst. 36, 806, 1965.

Progress Report

The investigations of the formation of excited states of the target atoms by proton impact, is now essentially completed. Although other interesting problems of this type exist we consider it more important to proceed with other facets of the program. The equipment used for these studies remains intact as an integral part of the recently developed system for investigating the formation of excited states of the projectile by charge transfer. It may readily be utilized for target excitation cross section measurements if ever a need arises. However there are no immediate plans for further work on target excitation studies.

VI. Charge Transfer Studies

The research program is now entirely devoted to the study of the formation of excited fast particles as the result of the transfer of an electron from the target into an excited state of the projectile. Of most interest is the case of proton projectiles.



The target, here denoted by X may be any of the inert or atmospheric gases.

Since the excited fast particle has a finite lifetime it will proceed a considerable distance through the target before decaying to the ground state with the emission of a photon. One may still define an emission cross section Q_{jk} , in terms of the number of photons of wavelength λ corresponding to the transition $j - k$ emitted in all directions

Progress Report

from a length ℓ of the beam path through the chamber per unit time, the target density N (particles per cc) and the incident beam current n (particles per second)

$$Q_{jk} = \frac{J_{jk}}{N n \ell} \quad (5)$$

Suppose that the excited state j of the projectile has a lifetime τ and the impact velocity is v . Then the emission cross section will be given as a function of the penetration x by the following equation

$$Q_{jk} = Q_j \frac{A_{jk}}{\sum_{k < j} A_{jk}} \left(1 - \exp - \frac{x}{v\tau}\right) \quad (6)$$

In principle it is possible to obtain the cross section Q_j for the formation of the excited state by investigating the emission as a function of penetration.

In the present work we are investigating the formation of excited hydrogen atoms by monitoring the emission of the Balmer alpha radiation. This consists of contributions from three transitions between the $n = 3$ and $n = 2$ states. Since these energy levels are very close it is not possible to separate the three transitions spectroscopically. However the $3s$, $3p$ and $3d$ states which are the parent levels for the three transitions have different lifetimes. Therefore the emission from the

Progress Report

projectile should be represented by the sum of three expressions like that in equation (6) each with one of the appropriate values of lifetime. In principle by fitting this theoretical curve to the observed emission it should be possible to separately elucidate the three cross sections. One may also consider applying this approach to the formation of higher excited states. However as the principal quantum number increases the separation in energy of the various l states gets exceedingly small and stray fields may readily perturb the energy of the various levels causing mixing of the states with consequential loss of information. Indeed simple calculations show that the field due to the space charge of the projectile beam itself becomes a very significant factor in the mixing of levels above $n = 3$. It would seem unlikely that the technique could be applied to other higher states due to this limitation.

This present investigation is of considerable importance since the theory underlying the process of charge transfer is less well understood than that for excitation. It is of considerable interest to determine how accurate the available theory is, particularly in the relatively high energy range available to the present experiment where theory and experiment are expected to agree. Similar work by Hughes et al.¹⁴ at energies up to 130 keV has indicated that agreement between experiment and theory does not occur at energies below 130 keV and that the testing of the formal theory must be carried to higher impact energies. The present experiment is designed to operate to 1 MeV and agreement should be expected in the upper end of the energy range available to the present experiment. Our present program becomes doubly important when one

14. R. H. Hughes, H. R. Dawson, B. M. Doughty, D. B. Kay, and C. A. Stigers. Phys. Rev. 146, 53, 1966.

Progress Report

considers that it may be impossible to determine population cross section for the various angular momentum quantum number states of $n > 3$, since space charge limitations will destroy the separate existence of different l states. For higher states it is not possible to separately test the rather fine theoretical predictions of the population of the different angular momentum states of the projectile. Only the relatively coarse information on total population is likely to be available.

(i) The Experimental System

The equipment used for this study is basically an extension of that which was provided for the investigation of emission from the target. The existing collision chamber was fabricated with a long side window opposite the aperture viewed by the spectrometer in target excitation studies. This chamber has been extended in length to provide a total flight path of 70 cm. A photomultiplier fitted with an interference filter is used to determine the emission from the chamber at various points along the flight path. The detector is cooled to reduce thermal noise and the output is handled by normal pulse counting circuitry. The projectile beam current and the target pressure are monitored with existing equipment in a conventional manner. Both of these parameters may be displayed in a digital form, to facilitate data recording and handling.

(ii) Experimental Results

Our initial approach was to make a survey of the operating conditions of the equipment, and to attempt some measurements of the phenomena which would provide the design parameters for developing

Progress Report

a comprehensive system for data recording and analysis. It was clear from the start that a considerable quantity of numbers would be extracted from the experiment in order to determine each cross section. Some form of rapid data recording system would be required for use during an extended series of measurements. Moreover there was no experimental information on what magnitudes of cross sections were expected at high energies or indeed what other experimental problems might exist. Therefore a fairly general investigation was undertaken to indicate what considerations were important before launching into precise measurement on an extended scale. It is the results of this preliminary investigation that we are able to present here. These measurements are likely to be of poor accuracy and will of course be repeated in more detail.

In figure 2 we show a number of curves of emission as a function of penetration for protons incident on a nitrogen target. The cross section scale is arbitrary. For convenience the abscissa of the graph is the ratio of the penetration through the cell to the velocity of the projectile, for the particular energy of impact under consideration. Thus the abscissa scale is in fact the time of flight from the point of entry to the chamber to the point of observation. Displaying the data in this manner gives a clearer view of the salient features. It is clear that the relative contributions of the 3s, 3p and 3d states are changing as a function of impact energy. At 150 keV the long lived 3s state is contributing the overwhelming component to the emission cross section, but at higher velocities a shoulder begins to appear on the curves close to the position of the beam entry to the collision chamber ($x = 0$). This indicates

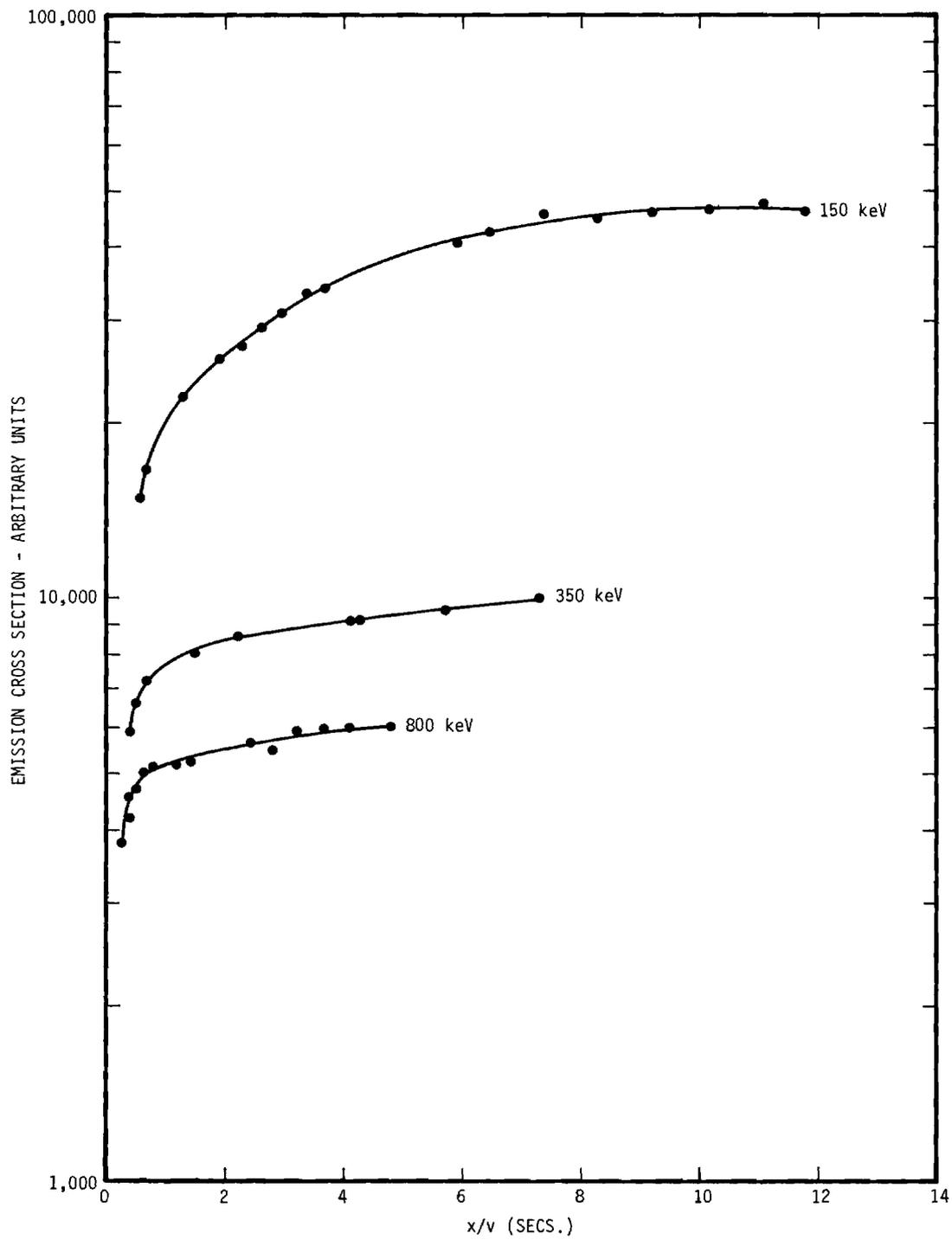


Figure 2. Balmer Alpha Emission Cross Section in Arbitrary Units Shown as a Function of Penetration Through the Target Divided by Projectile Velocity (x/v). The Cases Shown Are for Proton Impact on Nitrogen.

Progress Report

that the shorter lived components from the $3p$ and $3d$ states are becoming more important.

We have attempted to make a manual fit to these curves using a sum of three equations like (6) and assuming the knowledge of the theoretical lifetimes. Such a technique is not very accurate but for the moment suffices to allow separation of the $3s$, $3p$ and $3d$ cross sections. The branching of the $3p$ state decay was taken into account by using theoretical values of the $3p - 2s$ and $3p - 1s$ transition probabilities, and hence relative values of the excitation cross sections deduced. These are shown in Table I. Although the cross section scale is arbitrary, all these numbers are on the same relative basis.

Table I

Target	Energy keV	Cross Section (Arbitrary Units)		
		$3s$	$3p$	$3d$
N_2	150	5.0	10.0	0.15
	350	0.43	1.8	-
	800	0.10	1.6	-
He	150	1.0	-	0.080
	350	0.059	0.041	-

In all cases the population cross section shows the expected rapid decrease with increasing impact energy. It would appear that in both cases the population of the $3s$, and $3p$ states is much more important than the

Progress Report

population of the 3d. The 3s state populations for the N_2 target and He target are in approximately the same ratio as the total charge transfer cross sections.¹⁵ The 3s cross section for the case of helium decreases as the energy changes from 150 to 350 keV by the amount predicted by Mapleton's theory.¹⁶ In all respects the numbers we present here are as expected.

(iii) Problems of Data Handling and Analysis

Although we have taken the measured emission cross section and obtained values for the cross section for populating the various levels the manual curve fitting analysis that was applied must be considered very inadequate. A basic experimental test of the operation of the experiment is to show that the emission cross section curve can be represented uniquely by the sum of three exponential build ups of the type shown in equation (6). We might anticipate that a significant amount of cascade will occur from higher levels, a factor that will add yet another exponential term to our equation. There is always the danger that spurious electric fields are causing a mixing of the adjacent excited states, so introducing a "mixed" state whose lifetime is essentially unknown. These various possibilities must be taken into account. The most suitable technique is to show that the best fit to the experimental data is by a sum of three equations of the form of (6) with the known

15. C. F. Barnett and H. R. Reynolds. Phys. Rev. 109, 355, 1958.

16. R. A. Mapleton. Phys. Rev. 122, 528, 1961.

Progress Report

lifetimes of the excited states. The fit for three components must be substantially better than for four equations and preferably better than for two equations. There is the possibility, which our preliminary data already indicate, that the contribution from one of the levels may be very small, and the corresponding term may be dropped from the composite equation with no decrease in the goodness of fit. Clearly this fitting procedure will be best carried out by a computerized "least squares fit" of the theoretical curve to the experimental results.

Our preliminary investigations indicate clearly the importance of good statistics in the determination of the emission cross section as a function of penetration. The lifetime of the 3s states is sufficiently great that it is distinct from the 3p and 3d states and the emission cross section may be readily determined. However the 3p and 3d states lifetimes only differ by a factor of three and a unique determination of the separate populations of the 3p and 3d levels will require high statistical accuracy in the measurements. It has previously been reported by Hughes et al.¹⁴ in a similar experiment that although the 3s cross section could be determined, the ratios of the 3p, to 3d state population varied from one day to the next. This was ascribed to stray fields which varied from one day to another with a consequent variation in the mixing of the 3p and 3d states. We consider this to be an incorrect interpretation. It seems more likely that, due to poor statistical accuracy in the measurements, it was impossible to make an unambiguous fit of the theoretical curve to the measured data.

Coupled with the two problems discussed in the preceding paragraphs is the general immensity of the data recording problem. A determination

Progress Report

of a single cross section at a single energy by this experiment requires a very considerable body of numerical data. It is our intention to record as much as possible of our data automatically and to do it in a manner which makes it immediately suitable for computerized analysis. At the same time, the provision of semi-automated systems will probably allow an increased amount of time to be spent on the actual accumulation of data numbers and therefore lead to an improvement in statistics. It has been proved that the experiment will operate satisfactorily, and there is a better feel for the problems involved. Some time will now be expended on the improvement of the data logging and analysis systems.

VII. Program for the Remainder of the Contract Year

The majority of the remaining part of the contract year will be devoted to improving the capabilities of our data handling system, particularly in the recording of data. It is of vital importance to the success of the experiment that the statistical accuracy and method of analysis be greatly improved. At the present time we are already in a position to make an extended series of measurements of the $3s$ state population cross section. However the unambiguous separation of the $3p$ and $3d$ state contributions has never previously been achieved in a charge transfer experiment, and we are anxious to improve the analysis procedures so that this is accomplished. Since the same data has to be taken, irrespective of whether it is accurate enough to allow the extraction of all three cross sections it is clearly most efficient to solve all the problems before embarking on an extended measurement program.

Progress Report

Associated with our initial testing and proving of the experimental system a brief investigation will be made of charge transfer into excited states of He due to the impact of He^+ on a gaseous target. In this case there is no complication due to degeneracy of the energy levels of the emitting system, and the build up curve should be fitted quite unambiguously by a single equation of the form given by equation (6). However it is not intended that any appreciable program of measurements should be made on cases involving He^+ projectiles.

It is confidently anticipated that the required improvements and tests will be made before the end of the present contract period and that a start will be made on the program of cross section measurement.

A second possible technique for measuring cross sections for populating particular states of excitation of the projectile is to pass the projectile beam through a gas cell and analyze the emission as it traverses a following evacuated flight tube. This second approach, measuring decay as a function of distance in an evacuated region, is expected to be of particular value whenever the target system produced radiation of approximately the same wavelength as the Balmer alpha line. Since the cross sections for the formation of excited target states are generally much greater than those for charge transfer into the excited state of the projectile, interference from target emission could completely obscure emission from the fast particle. This second technique will be necessary for dealing with charge transfer for protons in hydrogen gas and may also be necessary for other cases. A further advantage of the gas cell approach is that by selecting the cell length it is possible

Progress Report

to selectively enhance the population of the shorter lived excited states in the projectile beam. As the beam traverses the target, processes of collisional excitation and spontaneous radiative decay compete in the population of the excited state. The shorter lived states reach an equilibrium between the population and depopulation processes at a very short penetration through the gas. This equilibrium situation provides the greatest population density of that state in the beam. By using a short gas cell the emergent beam may be arranged to contain a maximum equilibrium population of the short lived excited states, but with the population of the longer lived states considerably below the maximum. In this way the cross sections for populating the short lived states may more readily be elucidated from the data. We have pointed out in the previous section of this report that it is the separation of the contributions of the $3p$ and $3d$ states that causes most trouble in the measurement of these charge transfer cross sections. We consider that the gas cell approach is a necessary complement to the procedure that we are already operating. The system is currently being designed and will be fabricated before the end of the present contract period.

VIII. Program for the Future

The future program will be based on the exploitation of the two complementary techniques which are designed to measure cross sections for the formation of excited fast particles. As we have pointed out we are now in a position to carry out certain of these measurements immediately but that we are concentrating our efforts for the remainder of the present contract period on improving the system and realizing its full potentialities.

Progress Report

Our future program will involve the measurement of the separate cross sections for the formation of the 3s, 3p and 3d states of hydrogen by charge transfer as a proton beam traverses a target gas. We intend to use targets of molecular nitrogen and hydrogen, as well as helium and argon. This will provide a broad spectrum of experience with light and heavy systems, both atomic and molecular. The helium case is of importance for the testing of theoretical predictions, the nitrogen and hydrogen have possible practical applications.

It is also our intention to utilize this system for investigating the formation of these same states of hydrogen resulting from the collisional dissociation of H_2^+ and H_3^+ beams on various target gases. In this case some attention will have to be paid to the possible importance of excited states in the projectile beam. It is our intention to survey the importance of the excited state population by carrying out measurements with a wide variety of source conditions, and looking for any systematic variation. At the high energies utilized in this experiment it would seem possible that the pre-collision excited state of the projectile would only have a rather small influence on the cross section. However we cannot make any firm predictions on this point before experimental surveys have been carried out.

IX. Publications and Travel

A total of eight reports have been prepared in the present contract year, all of which are intended for publication in the open literature. Of these, two have been published in conference proceedings, four have been accepted for publication in refereed journals and two are still

Progress Report

pending, having been submitted for publication. We list below the titles and status of these eight reports.

- (a) "The Excitation of Molecular Nitrogen and Oxygen by the Impact of 0.15 to 1.0 MeV Protons", by E. W. Thomas, G. D. Bent and J. L. Edwards. Published in the Proceedings of the Fifth International Conference on the Physics of Electronic and Atomic Collisions. (Pub. Nauka Publishing House, Leningrad, 1967) Page 286. AEC Report Number ORO-2591-24.
- (b) "Balmer Emissions from a Molecular Hydrogen Target Induced by the Impact of Fast Protons", by J. L. Edwards and E. W. Thomas. Ibid. p. 288. AEC Report Number ORO-2591-25.
- (c) "A Comparison of the Excitation Produced in a Helium Target by the Impact of H^+ , D^+ , H_2^+ , H_3^+ Ions at High Energy", by E. W. Thomas and G. D. Bent. (Scheduled for publication in J. Opt. Soc. Am., December 1967). AEC Report Number ORO-2591-27.
- (d) "The Formation of Excited States in a Helium Target by the Impact of 0.15 to 1.0 MeV Protons and Deuterons. I Experimental", by E. W. Thomas and G. D. Bent. (Scheduled for publication in Phys. Rev. 164, Dec. 5, 1967). AEC Report Number ORO-2591-28.
- (e) "The Formation of Excited States in a Helium Target by the Impact of 0.15 to 1.0 MeV Protons and Deuterons II. Comparison with Theory", by E. W. Thomas. (Scheduled for publication in Phys. Rev. 164, Dec. 5, 1967). AEC Report Number ORO-2591-29.

Progress Report

- (f) "Cross Sections for the Formation of Excited States in a Nitrogen Target by the Impact of 0.15 to 1.0 MeV Protons", by E. W. Thomas, G. D. Bent and J. L. Edwards. (Accepted for publication in the Physical Review). AEC Report Number ORO-2591-30.
- (g) "Emissions from a Molecular Hydrogen Target Induced by the Impact of Fast Protons", J. L. Edwards and E. W. Thomas. (Submitted for publication to The Physical Review). AEC Report Number ORO-2591-31.
- (h) "The Formation of Excited Ions by the Impact of 0.15 to 1.0 MeV Protons on a Molecular Oxygen Target", by E. W. Thomas and G. D. Bent. (Submitted for publication to Proc. Phys. Soc. London). AEC Report Number ORO-2591-32.

Dr. Thomas and Mr. Edwards attended the Fifth International Conference on the Physics of Electronic and Atomic Collisions, in Leningrad, Russia during the period July 17 to 23. Two papers were presented both of which were published in the conference proceedings and are listed above. Associated with the conference both men visited research laboratories in Leningrad, Amsterdam, and London. Dr. Thomas also visited Culham (England) and Lyons (France). The cost of this travel for Dr. Thomas was borne partly by this AEC contract. The travel by Mr. Edwards was at no cost to the contract.

Dr. Thomas gave an invited seminar to the Ballistics Research Laboratory, Aberdeen, Maryland in February 1967. Dr. Thomas also attended a small invited conference on "Diatomic Collisions and Electronic Structure" at the Stanford Research Institute in May 1967. Periodic visits have been

Progress Report

made by Dr. Thomas to the Oak Ridge National Laboratory for the purposes of consultation with C. F. Barnett and others. Mr. Edwards and Mr. Ford have also paid visits to the Oak Ridge Laboratories during the present contract year.

X. Personnel

The work described in this report was carried out as part of AEC Contract No. AT-(40-1)-2591 and has been under the jurisdiction of Dr. Thomas.

Mr. Lee Edwards continues to work full time on this project although he is at present the recipient of a N.A.S.A. scholarship and has not drawn financial support from the project. It is expected that this program will provide the basis for his Ph.D. thesis problem in the future.

Mr. John Ford has recently joined the project and will work approximately half time. It is hoped that he will eventually pick a thesis problem from the work carried out under this contract.

Mr. Gary Bent worked for half of the present year on this program and has now terminated his association with the work. During the course of the year a number of other graduate students have been employed for short periods of time to handle the reduction of data and to carry out small parts of our overall program.

XI. Accelerator Usage

Use of the Van de Graaff accelerator is shared with a project to measure ionization cross sections, also under AEC Contract No. AT-(40-1)-2591.

Progress Report

No difficulty is experienced in switching the ion beam from one experiment to the other. A period of one day suffices to make the required mechanical connections and to align the ion beam with the experiment. The accelerator operation time is shared equally by the two experiments, alternating at intervals of one month or longer.

XII. Incident Report

There have been no incidents for which a report is required during the performance of the research under this contract in the present reporting period.

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ORO-2591-37

EMISSION AND EXCITATION

CROSS SECTIONS

PROGRESS REPORT NO. 4

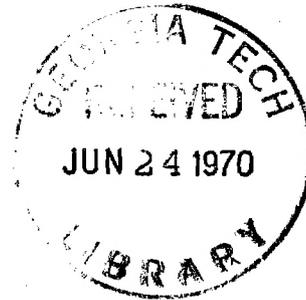
Covering the Period

December 1, 1967 to November 30, 1968

By E. W. Thomas

J. L. Edwards

J. C. Ford



Report No. ORO-2591-37

Contract No. AT-(40-1)-2591

U. S. ATOMIC ENERGY COMMISSION

OAK RIDGE, TENNESSEE

30 November



School of Physics
GEORGIA INSTITUTE OF TECHNOLOGY
Atlanta, Georgia

EMISSION AND EXCITATION CROSS SECTIONS

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30 November 1968

TABLE OF CONTENTS

	Page
I. Title	1
II. Introduction.	1
III. Abstract.	1
IV. Objective	2
V. Experimental Method	4
VI. Summary of Progress Made During the Present Reporting Period.	9
(a) Experimental systems and data handling	10
(b) The effect of Doppler shift on detection sensitivity	12
(c) Weak field Stark effect.	13
(d) Cross section measurements	18
VII. Program for the Remainder of the Contract Year.	20
VIII. Program for the Future.	20
IX. Publications and Travel	21
X. Personnel	22
XI. Accelerator Usage	23
XII. Incident Report	23

I. Title

Emission and Excitation Cross Sections

II. Introduction

This report summarizes the work performed on excitation phenomena as part of contract AT-(40-1)-2591 for the U. S. Atomic Energy Commission. The work was initiated on March 1, 1965, as a result of Modification No. 7, increasing the scope and financing of the existing contract. The present report covers the period 1 December 1967 to 30 November 1968 which corresponds to the first 9 months of the 12 month period covered by Modification No. 10 to this contract, plus the final three months of the preceding contract period.

A program for the measurement of ionization cross sections which was the subject of the original contract and subsequent Modifications 1 through 10 has been continued and is reported separately.¹

III. Abstract

The formation of fast excited H atoms by impact of H^+ , H_2^+ , or H_3^+ ions on various gaseous targets is studied at projectile energies ranging from 0.15 to 1 MeV. Attention is restricted to the formation of the 3s, 3p, and 3d excited states of hydrogen. An interim report on the progress of this work is presented. Particular attention is given to the errors which might be introduced by weak field Stark effects and to the problems which arise due to the Doppler shift of the Balmer alpha emission. Results are presented for the relative values of the cross sections for charge transfer into the 3s,

1. D. W. Martin and G. O. Taylor. "Ionization and Charge Transfer Cross Sections," Progress Report No. 21. 30 November 1968. U. S. Atomic Energy Commission Report Number ORO-2591-38.

3p, and 3d states of hydrogen. A survey is made of the phenomena observed as a result of the dissociation of H_2^+ and H_3^+ by impact on various targets.

IV. Objective

The objective of this research is the measurement of cross sections for production of fast neutral hydrogen atoms in excited states induced by the impact of H^+ , H_2^+ , and H_3^+ on various gaseous targets. The energy of the incident particles ranges from 0.15 to 1.0 MeV.

The processes of interest in the present studies may be described by the following reaction equations:



The target "X" will be helium, argon, hydrogen, or nitrogen. The experiments are designed to measure the cross sections for the formation of neutral hydrogen in the 3s, 3p, and 3d states. The square brackets in equations (1) through (3) are used to indicate that the experiments will not give information on the states of ionization, excitation, or molecular association of the post-collision products. However, in most situations it will be possible to make recourse to other published information on these collision processes and to eliminate from further consideration certain unlikely post-collision states. At the present time, the main thrust of the program is concentrated on the study of the charge transfer process described by equation (1). The two dissociation mechanisms are being surveyed with a view to commencing a detailed study of their behavior.

A prime objective in the present program is the acquisition of a body

of data which can be used for comparison with theoretical predictions. There are few accurate quantum mechanical calculations of the cross sections for the processes which are of interest in the present program. Some semiclassical theories are available which are of doubtful accuracy. It is of the utmost importance that the approximations made in theoretical predictions should be tested at every available opportunity in order to establish the validity of the calculations for situations where experimental verification is impossible.

An important aspect of the present work is that it studies cross sections for the formation of states of different angular momentum quantum numbers. Such measurements will provide a far better understanding of the collision processes than will a measurement of the total population of a particular "n" state or a population integrated over all excited states. It is believed that only for the $n=2$ and $n=3$ states will it be possible to measure the separate cross sections for the formation states with different angular momentum. Stray fields of the magnitudes that are commonly encountered in experimental systems will be sufficient to cause Stark mixing of the sublevels of states where $n \geq 4$. Therefore, such states do not maintain a separate identity under practical circumstances and cannot provide any assistance with the testing of theoretical predictions. It is concluded that detailed test of theory will only be possible for the $n=2$ and $n=3$ levels.

A second objective in the present work is the provision of data which are of direct value for design and diagnostic purposes. A particular application is found in the thermonuclear research program. In some large scale experimental systems, the plasma density is built up by injecting neutral beams of hydrogen atoms into the containment device. Energetic neutral atoms may be trapped by collisional ionization resulting from impact on some form of dense target plasma, or by Lorentz ionization. This latter process is the

spontaneous ionization of an excited neutral particle when it experiences a sufficiently large electric or magnetic field. At high injection energies Lorentz ionization can provide an appreciable part of the trapping capability. However, the fraction of neutral particles which can be trapped in this manner is limited by the small proportion of the neutral beam which can be produced in the necessary highly excited state. The present program is designed to investigate from a fundamental viewpoint processes which give rise to the formation of fast neutral H atoms in excited states. Although the $n=3$ states are not of themselves useful for injection purposes, their study should provide an insight into the general behavior of these processes. Furthermore, it will be possible to estimate cross sections for higher states by using the well known rule that cross sections decrease as n^{-3} at high impact energies.

There are a number of other practical situations in which knowledge of these excitation cross sections is important. In particular, they are relevant to the understanding of certain phenomena observed in natural aurora and in the complex situations which exist some milliseconds after an atmospheric nuclear explosion.

V. Experimental Method

The source of the energetic projectiles for the present experiment is a 1 MeV Van de Graaff positive ion accelerator, which is equipped with a beam analyzing and stabilizing system. The ion beam is collimated by a series of small apertures before entering the experimental region. The formation of excited H atoms in the 3s, 3p, and 3d states is detected by the quantitative measurement of the Balmer alpha photons emitted as the excited atoms decay to the $n=2$ level.

Two distinct experimental arrangements are available. In the first, the photon emission from the H atoms is observed as the beam traverses the

target gas. In the second, the beam traverses a gas cell of definite length where the collisional excitation occurs and the emissions are observed from the H atoms as they emerge from the cell and proceed through an evacuated flight tube. After traversing the collision and detection regions, the ion beam is monitored on a conventional Faraday cup.

At the high impact energies utilized in this experiment, the product of the projectile's velocity and the lifetime of the excited state is comparable with the dimensions of the apparatus. Therefore, in general, a projectile will move an appreciable distance from the point where it was excited before emitting a photon and decaying to a lower state. As a result, the intensity of emission from the projectiles is a function of the position along the flight path where the observation is made. Measurement of the spatial variation of this intensity allows the evaluation of the population of the emitting state. We now consider separately the mathematical formulation which predicts the intensity variation in the two experimental configurations identified above.

First consider the configuration where measurements are made of intensity from the path of the beam through the target. Let us suppose that the apparatus detects the photons emitted as result of the decay of the state j to a lower state k . The state j has a lifetime τ_j , and the cross section for its formation by one of the mechanisms listed as equations (1) through (3) is Q_j . The apparatus measures the flux of photons J_{jk} emitted per second in the transition $j \rightarrow k$ from a length l centimeters of the beam path. A flux of N projectile molecules per second are incident with a velocity v (cm/sec) on a target which has a number density n (molecules/cc). In general, the state j may decay by many paths to lower states i , only one of which ($j \rightarrow k$) is

detected experimentally. Therefore, the detected photons correspond only to the fraction $A_{jk} / \sum_{i<j} A_{ji}$ of the total number of decays. Here A_{jk} (sec^{-1}) is the probability of spontaneous decay of the state j to the state k and this ratio is known as the branching ratio. The observation region l is made shorter than $v\tau_j$, and the target density may be kept sufficiently low that single collision conditions are maintained. It may be shown² that at a point x cm from the entrance to the target region, the intensity is given by

$$J_{jk} = N n l Q_j \frac{A_{jk}}{\sum_{i<j} A_{ji}} \left[1 - \exp\left(-\frac{x}{v\tau_j}\right) \right] \quad (4)$$

In arriving at this result, we have made the assumption that population of the state j by cascade from higher levels is sufficiently small that it may be neglected. This assumption must be subjected to experimental test. In the present experiment, the Balmer alpha emission is due to three transitions; $3s \rightarrow 2p$, $3p \rightarrow 2s$, and $3d \rightarrow 2p$. These all emit photons of essentially the same wavelength and are therefore detected simultaneously. The observed intensity is a sum of three terms of the type given in equation (4).

$$J_{\alpha}(x) = I_1 \left[1 - \exp\left(-\frac{x}{v\tau_1}\right) \right] + I_2 \left[1 - \exp\left(-\frac{x}{v\tau_2}\right) \right] + I_3 \left[1 - \exp\left(-\frac{x}{v\tau_3}\right) \right] \quad (5)$$

where

$$I_j = N n Q_j \frac{A_{jk}}{\sum_{i<j} A_{ji}}$$

The subscripts 1, 2, and 3 are used to indicate, respectively, the 3s, 3p, and 3d states. In fact, the 3s and 3d states can decay only by the Balmer

2. See, for example, C. E. Head and R. H. Hughes, Phys. Rev. 139, A1392 (1965).

alpha transition and therefore the branching ratio $A_{jk} / \sum_{i<j} A_{ji}$ for these states is unity. For the 3p-2s transition, this ratio is 0.118, indicating that only 11.8% of the atoms in the 3p state decay by the emission of a Balmer alpha line, the rest by the Lyman beta. Equation (5) represents a sum of three terms which increase exponentially with x towards some constant value. The lifetimes of the three states are quite different from each other. It is possible to compare the measured function $J_{\alpha}(x)$ with equation (5) and to evaluate the coefficients I_j . In this manner, the cross section for the formation of the 3s, 3p, and 3d states may be measured using the different lifetimes to identify the three levels.

The second experimental configuration involves observation of the decay of the excited states in the beam emerging from a gas cell into an evacuated flight tube. In this case the intensity of emission from each state will simply decay exponentially with distance along the flight tube with a decay length characterized by the lifetime of the excited state. The population of the excited states in the emergent beam will be a function of the cell length L . The intensity of emission in the transition $j \rightarrow k$ as a function of the distance x beyond the exit from the gas cell may be shown³ to be given by the following equation.

$$J_{jk} = N n \ell Q_j \frac{A_{jk}}{\sum_{i<j} A_{ji}} \left[1 - \exp\left(-\frac{L}{v\tau_j}\right) \right] \exp\left(-\frac{x}{v\tau_j}\right) \quad (6)$$

Again the Balmer alpha intensity is in fact the sum of three terms and can be represented by the equation

3. R. H. Hughes, et al., Phys. Rev. 146, 53 (1966).

$$\begin{aligned}
J_Q(x) = I_1 \left[1 - \exp \left(- \frac{L}{v\tau_1} \right) \right] \exp \left(- \frac{x}{v\tau_1} \right) + I_2 \left[1 - \exp \left(- \frac{L}{v\tau_2} \right) \right] \exp \left(- \frac{x}{v\tau_2} \right) \\
+ I_3 \left[1 - \exp \left(- \frac{L}{v\tau_3} \right) \right] \exp \left(- \frac{x}{v\tau_3} \right)
\end{aligned} \tag{7}$$

I_j has the same significance as before. This equation may be fitted to the observed emission and the cross sections evaluated.

The collision and observation regions consist of a gas cell followed by a long flight tube. Plate glass windows in the flight tube allow the observation of the whole flight path. Detection is carried out with a photomultiplier fitted with an interference filter. The detector can be moved along a machined track to measure intensity at various values of the distance x . For the first mode of operation the target gas is introduced into the flight tube and the gas cell is evacuated. For the second mode the target gas is introduced into the cell and the flight tube is evacuated.

The two experimental configurations are complementary, each having different advantages and drawbacks. Observations made in the target region may be subject to interference from collisionally induced target gas emissions. These will be invariant with beam penetration through the gas and will require the inclusion of an additional constant term to equation (5). If this constant is comparable with the other terms, then it is impossible to evaluate the separate 3s, 3p, and 3d excitation cross sections with any accuracy. In particular, the interesting case of an H_2 target becomes quite impossible due to target emission. The approach of using a cell and an evacuated flight tube requires care to ensure that the exit aperture from the cell does not intercept an appreciable fraction of the scattered projectiles. Furthermore, there is an uncertainty as to the "thickness" of a gas cell due to pressure gradi-

ents at the two apertures. In addition, certain experimental problems associated with the weak field Stark effect (discussed fully in section VI (c)) are likely to be different in the two configurations. It is our judgment that both configurations should be employed and that agreement between measurements made using the two different approaches will suggest that the experiments are operating satisfactorily.

VI. Summary of Progress Made During the Present Reporting Period

The objectives of the work in the present reporting period were three-fold. Firstly, it was deemed necessary to make considerable improvements in the data recording systems in order to improve accuracy and stability, as well as to reduce operator fatigue. A typical measurement of light intensity as a function of penetration through the target region requires approximately four hours of work during which some one to two hundred data points are taken. It was pointed out in our previous report⁴ that excellent reproducibility was required in order that the small contributions expected from the short-lived 3p and 3d states could be accurately measured in the presence of the very much larger 3s contribution. The second objective was the detailed investigation of charge transfer processes described by equation (1) using various target gases. The third objective was a survey of the dissociation processes described by equations (2) and (3) with the intention that they should form the basis of an extensive program of measurements. The first of these objectives has been successfully achieved and is complete. The measurement of charge transfer cross sections has moved rather slowly but will be largely completed by the end of the present contract year. The survey of dissociation

4. E. W. Thomas, J. L. Edwards, and G. D. Bent. "Emission and Excitation Cross Sections." Progress Report Number 3. 1 December 1967. AEC Report Number ORO-2591-33.

mechanisms has been carried out and some preliminary measurements are presented in this report. No extensive program of work on dissociation has been commenced as yet.

In addition to the objectives detailed in the proposal, it has been necessary to give some consideration to two additional problems which may effect the accuracy of the measurements. The first is the variation of optical detection sensitivity of the optical system with the energy of the projectile which results from the Doppler shift and broadening of the emitted radiation. The second is the influence of stray electric and magnetic fields on the excited projectiles by the Stark effect. Our studies indicate that the problems introduced by these effects can be overcome.

We will now consider in some detail the progress made in this program. In keeping with the above remarks, the discussion will be divided under the headings: (a) Experimental systems and data handling; (b) The effect of Doppler shift on detection sensitivity; (c) Weak field Stark effect; and (d) Cross section measurements.

(a) Experimental systems and data handling

A major problem with the original equipment was that a high background light emission was observed from the excitation of the background gas in the chamber. It was impossible to identify the source of the emission, but it may have been due to Balmer alpha lines induced by collisional dissociation of hydrocarbons or water vapor. In order to reduce this problem, the system was completely dismantled, cleaned, and reassembled using Viton O rings. Some modifications were made to reduce pumping impedances and to remove certain unnecessary tubulations. As a result, the background pressure dropped by about two orders of magnitude and the background light emission was corres-

pondingly reduced. This no longer constitutes a problem.

The optical system forms an image of the ion beam on the photomultiplier cathode which occupies an area of $3/8$ by $1/4$ inches. It is very important that the detection efficiency be the same over the whole of this area. Detailed tests were made of the constancy of the photocathode sensitivity over the surface of the photomultiplier tube (EMI type 9558 B). It was found that deviations of up to 30% could occur over a distance of 1 cm. Most of the variations could be related to gaps in a metallic coating on the inside of the tube. This coating is used as a focussing electrode and the gaps presumably will produce local distortions in the focussing field. By monitoring the sensitivity along various diameters of the photocathode, it was possible to choose one region where the sensitivity was constant to within 2%. The tube is now utilized with the remainder of the sensitive area baffled so that light from the collision region can fall only on the area of uniform sensitivity.

The various components of the electronics have been completely overhauled and the stability of operation vastly improved. The overall reproducibility of the experimental measurements over a period of some four hours is now better than 1%.

In order to expedite data handling, a completely automated programming system has been built to operate the experiment. The programmer moves the detection system through a series of predetermined steps along the flight tube. At each step the light intensity, beam current, target pressure and position are monitored and recorded on punched tape. Operator activity is confined primarily to overseeing the general quality of the output data, periodically checking electronics stability and maintaining satisfactory accelerator operation.

The intensity measured by the apparatus should have a variation with position x given by either equation (5) or (7). A computer program has been written which carries out a least squares fit of the equations to the data and evaluates the three parameters I_1 , I_2 , and I_3 . For completeness, we have also added a constant K to the right hand side of equations (5) and (7) to represent any emission which might originate from target excitation and which would be independent of position x . The punched tape carrying the experimental data is processed by the Georgia Tech Burroughs B-5500 computer and the output provides the fitted values of I_1 , I_2 , I_3 , and K .

(b) The effect of Doppler shift on detection sensitivity

The observations in this experiment are made at an angle of 90° to the direction of motion of the emitting atom. Due to the high velocities used in this experiment (0.01 to 0.05 of the velocity of light), the transverse relativistic Doppler shift is quite appreciable. Moreover, due to the finite acceptance angle of the detection system, photons emitted in a range of directions from 79° to 101° to the beam axis are detected. Therefore, there is a range of wavelengths being accepted. In the present experiment, the Doppler shift of the transverse emission ranges from 0.53 to 7 Å, and the Doppler width of the line, due to the finite acceptance geometry, ranges from 32 to 116 Å.

The Doppler effect may be regarded as complicating the assessment of the detection sensitivity of the apparatus. The detector utilizes a narrow band filter. As the energy of the projectile is varied, the Doppler width of the line changes and so does the overall sensitivity of the detection system. It must be emphasized that this effect does not in any way influence our procedures for finding the ratio of the 3s, 3p, and 3d cross sections at a given

impact energy. Moreover, measurements of cross sections using different target gases but the same projectile velocity can also be compared directly. However, it becomes difficult to relate the measurements made at different impact energies and to determine the variation of cross section with impact energy.

Published data on the excitation of the 3s, 3p, and 3d states are restricted to the work of Head, Hughes, et al.^{2,3} They used the same technique as is employed here but took no account of the Doppler broadening of the emission. Although it is not possible to accurately assess the Doppler width in their apparatus, it was probably about the same order as in the present work. The highest energies used were 120 keV and the Doppler width under those circumstances might be as high as 30 Å. The neglect of this problem has probably introduced an energy dependent error into the measurements.

A method has been devised for overcoming the Doppler shift problem. Basically it involves determining the transmission of the filter as a function of wavelength and the transmission of the optics as a function of the angle of incidence of the photons on the first lens of the optical system. Absolute calibration of the overall detection efficiency is related to a standard lamp of known emissive power. The complete mathematical justification for the procedures is too long to be displayed here. However, it is believed that they can be carried out quite accurately.

(c) Weak field Stark effect

The experiment is designed to determine the cross sections for the formation of the 3s, 3p, and 3d states using the lifetimes of the states to identify them separately. However, if an electric field is applied to the excited atom, the energy levels will be perturbed and this may result in a

change of the lifetimes of the excited states. Fields of the order one volt per cm may produce an appreciable effect on lifetimes of excited hydrogen atoms. Fields of this magnitude may easily arise in the apparatus from build-up of charge on insulating surfaces and from the Lorentz field produced by the component of the earth's magnetic field transverse to the beam axis. In our previous report,⁴ the probable importance of the weak field Stark effect was anticipated but no detailed attention was paid to its effect on the observations. It appears that the weak field Stark effect in hydrogen is a very complex phenomenon that has only recently been subjected to experimental investigation.

Let us consider a beam of projectiles which are collisionally excited to one specific state at a point on their path. In practice, this might be accomplished by passing the beam through a thin metal foil. In a field free situation, the intensity of emission will decrease exponentially with distance from the point of excitation. The intensity will decrease by a factor of $\exp^{(-1)}$ in a distance equal to the product of velocity v and lifetime τ . However, in the presence of a weak field, the decrease will be given by the sum of a number of exponential terms. Under some circumstances there is also an oscillatory component superimposed on the decay. This remarkable behavior may be understood from basic quantum mechanical considerations. A brief review of the relevant theory is presented below.

In the present example, we consider only the mixing between such hydrogenic states as $3S_{\frac{1}{2}} - 3P_{\frac{1}{2}}$ or $3P_{\frac{3}{2}} - 3D_{\frac{3}{2}}$, though the mixing between states of different j may also be important under some circumstances. An atom initially formed in an excited state, $|n, l = j - \frac{1}{2}, j, m\rangle$, at $x=0$ is then Stark mixed into a state $|n, j, m\rangle$ which is a superposition of the states, $|n, l = j - \frac{1}{2}, j, m\rangle$

and $|n, \ell = j + \frac{1}{2}, j, m\rangle$.

$$|n, j, m\rangle = b^- |n, \ell = j - \frac{1}{2}, j, m\rangle + b^+ |n, \ell = j + \frac{1}{2}, j, m\rangle \quad (8)$$

The mixing is governed by the Stark matrix element

$$\langle \ell = j - \frac{1}{2} | H_{\text{Stark}} | \ell = j + \frac{1}{2} \rangle = \hbar V = -\frac{a}{4} e \frac{a |\vec{\epsilon}| n |m| \sqrt{n^2 - (j + \frac{1}{2})^2}}{j(j+1)} \quad (9)$$

where " $\vec{\epsilon}$ " is the electric field, "a" is the first Bohr radius, and "e" is the electronic charge. Bethe and Salpeter⁵ show that the state, $|n, j, m\rangle$, has two energy eigenvalues each of which depends linearly upon m and $|\vec{\epsilon}|$. For Stark energies large compared to the Lamb shift separation between the two unperturbed states, the ratio, b^-/b^+ , is ± 1 ; otherwise, b^-/b^+ depends upon the field strength, Lamb shift, and quantum numbers n and m.

The state, $|n, j, m\rangle$, then gives rise to the intensity function

$$I(x) = A^- \langle \ell = j - \frac{1}{2} | b_m^{-*} b_m^- | \ell = j - \frac{1}{2} \rangle_{\text{avg.}} e^{\frac{-A^- X}{\text{vel.}}} + A^+ \langle \ell = j + \frac{1}{2} | b_m^{+*} b_m^+ | \ell = j + \frac{1}{2} \rangle_{\text{avg.}} e^{\frac{-A^+ X}{\text{vel.}}} \quad (10)$$

where A^- and A^+ are the spontaneous transition probabilities per unit time for the unperturbed states characterized by $\ell = j - \frac{1}{2}$ and $\ell = j + \frac{1}{2}$, respectively. Vel. is the beam velocity determined by the acceleration potential. When b^-/b^+ depends on m (for very small fields), $I(x)$ should be summed over all possible values of m. This, however, requires a knowledge of the relative m level populations.

When the Stark effect splitting is comparable to the radiation widths

5. H. A. Bethe and E. E. Salpeter, Quantum Mechanics of One and Two-Electron Atoms (Springer-Verlag, Berlin, 1957), p. 239.

of the two states $|n, l = j - \frac{1}{2}, j, m\rangle$ and $|n, l = j + \frac{1}{2}, j, m\rangle$ the situation becomes more complex. Now the decay must be treated by time dependent theory.^{6,7} Here b^- and b^+ are time dependent and must satisfy the following coupled differential equations.

$$\begin{aligned} \frac{d b^-(t)}{dt} &= -i V b^+(t) e^{-i\delta t} - \frac{1}{2} A^- b^-(t) \\ \frac{d b^+(t)}{dt} &= -i V b^-(t) e^{+i\delta t} - \frac{1}{2} A^+ b^+(t) \end{aligned} \quad (11)$$

δ represents the Lamb shift separation between the two unperturbed states in radians per second. Again, $\hbar V$ is the Stark matrix element between the two given unperturbed states. Wangsness⁸ gives the solutions $b^-(t)$ and $b^+(t)$ in terms of their initial values, $b^-(t=0)$ and $b^+(t=0)$. In our case $b^+(t=0)$ is zero. In general, $b^-(t)$ and $b^+(t)$ are complicated functions which lead to a rather complicated expression for the intensity $I(x)$. Qualitatively, $I(x)$ is a combination of sinusoidally modulated terms which decay exponentially with various time constants.

Recent foil-excitation experiments^{9,10,11} have shown the quasi periodic

6. E. Wigner and V. Weisskopf, Z. Physik 63, 54 (1930).

7. H. A. Bethe and E. E. Salpeter, Quantum Mechanics of One and Two-Electron Atoms (Springer-Verlag, Berlin, 1957), p. 288.

8. R. K. Wangsnes, Phys. Rev. 149, 60 (1966).

9. W. S. Bickel, J. of the Optical Soc. of Am. 58, 213 (1968).

10. Bashkin, Bickel, Fink, and Wangsness, Phys. Rev. Let. 15, 285 (1965).

11. Bickel and Bashkin, Phys. Rev. 162, 12 (1967).

nature of $I(x)$. In the present work, however, it should be borne in mind that excitation takes place over a fairly broad interval in x rather than a sharply defined region as in the case of a foil. Thus, for excitation regions large compared to the quasi period of $I(x)$, the average $I(x)$ may not exhibit a periodic nature. Due to the various exponential terms, $I(x)$ may still differ considerably from the unperturbed intensity.

At the present time, the influence of this problem is being assessed in the following manner. The theory by Wangsness⁸ is being used to predict $I(x)$ for various very small fields of the order one volt per cm. These predictions will then be folded into our practical experimental situation which involves a collision region which is somewhat larger than the quasi period of the spatial oscillations. This will, in principle, provide intensity predictions which will reduce to equations (5) and (7) for a field free case. Our observed intensities will be examined to see whether the Stark mixed predictions fit the observations better than the field free formulae. Finally, it is intended to apply small fields to the observation region to see whether the weak field Stark predictions can be verified.

Clearly, the weak field Stark effect may seriously distort the operation of the experiment. It would be impossible to correct for the effect since any fields present in the system would be stray fields possibly varying in space and time. In principle, it would be possible to design a field free experiment, but this would involve a considerable increase in complexity. However, it must be emphatically stated that there is no evidence from the present observations that the Stark effect is appreciably affecting the experiment. All existing measurements appear to be consistent with a three component contribution to the intensity, with the lifetimes of the states

being equal to those predicted theoretically for a field free situation. It would be expected that, if Stark mixing were appreciable, then the three component equation might not fit the data and the lifetimes would certainly not be correct.

(d) Cross section measurements

At the time of writing, some results are available for the charge transfer process and a survey has been completed on the dissociation mechanism. The cross section information is only in the form of relative values for the formation of the three states at one impact energy. No comparison between cross sections at different impact energies has been made. All measurements have been carried out using the experimental configuration where the emission from the target region is detected and equation (5) described the operation of the experiment.

(1) Charge transfer. The available results are as follows, at 150 keV:

	Q(3s)	Q(3p)	Q(3d)
$H^+ + He$	1	--	0.055
$H^+ + N_2$	3.66	--	0.494

350 keV:

$H^+ + He$	1	0.17	--
$H^+ + N_2$	1.24	3.61	--

The two groups of data have each been normalized to the Q(3s) cross section for the $H^+ + He$ reaction. Gaps in the table indicate that the cross sections were too small to be measured. These data have been confirmed by a number of experimental runs and are reproducible to better than 10%. Each experimental run involves taking some 100 to 200 data points all of which are used in the

least squares fitting procedure.

Some comparison may be made with data from other sources. Hughes, et al.³ present the $Q(3s)$ cross section for energies up to 100 keV for a variety of target gases. They find that at 100 keV the cross section for impact on N_2 is five times greater than that for a helium target, compared with a factor of 3.7 at 150 keV in the present work. Mapleton¹² predicts theoretically that at 125 keV for impact on helium:

$$Q(3s) : Q(3p) : Q(3d) = 1 : 0.57 : 0.03$$

and at 395 keV:

$$Q(3s) : Q(3p) : Q(3d) = 1 : 0.27 : 0.0074$$

These predictions do not bear much of a resemblance to the present work at 150 and 350 keV.

A survey has also been carried out using an argon and hydrogen target. It is found that the high emission from the hydrogen target renders measurements on this case using the present technique impossible. However, the argon case seems quite feasible.

(ii) Dissociation. Preliminary surveys have been carried out for H_2^+ and H_3^+ impact on He and N_2 targets. As expected, the cross sections for forming excited H by dissociation are many orders of magnitude greater than those for the charge transfer process. An interesting feature of the dissociation phenomena is that the cross sections for forming the short-lived 3p and 3d states are generally much greater than for the long-lived 3s state. As an example, for impact of 300 keV H_2^+ on helium

$$Q(3s) : Q(3p) : Q(3d) = 1 : 2.55 : 1.79$$

12. R. A. Mapleton, Phys. Rev. 122, 528 (1961).

and for impact of 150 keV H_2^+ on N_2

$$Q(3s) : Q(3p) : Q(3d) = 1 : 4.1 : 3.7$$

Analysis of additional data on these cases is proceeding at the present time.

VII. Program for the Remainder of the Contract Year

The operation of the experiment will be examined to ascertain whether any Stark mixing of states is being induced by stray weak electric fields. In particular, tests are to be made to actually determine the apparent lifetimes of the 3s, 3p, and 3d states using this apparatus. This may be achieved by taking equation (5), which described the intensity as a function of penetration, and treating the lifetimes as unknown quantities. As a result, a six parameter fit must be made to the data. If such a fit gives the theoretically predicted field free lifetimes, then it may be concluded that stray field mixing is not a problem. Initial tests suggest that this is in fact the case. Direct tests of the possible influence of stray fields will be made by deliberately applying small fields in the observation region and determining the field strengths at which the experimentally determined cross sections become appreciably affected.

Measurements of the relative values of $Q(3s)$, $Q(3p)$, and $Q(3d)$ cross sections in charge transfer collisions will be continued. A greater range of impact energies will be covered and also argon will be included as a target in addition to He and N_2 .

Two months of accelerator operation time have been scheduled in order that these objectives will be achieved during the remainder of the contract year.

VIII. Program for the Future

The relative cross section values measured during the present contract

year will be placed on an absolute basis by the calibration of the detection sensitivity of the optical system. Due allowance will be made during these procedures for the energy dependent effects of Doppler shift and broadening.

The cross sections for forming excited fact atomic hydrogen by impact of protons on a molecular hydrogen target will be measured using a gas cell followed by an evacuated observation region. This configuration of the apparatus has not yet been utilized and some detailed check of its operation will be necessary.

Finally, the program will devote some detailed study to the collisional dissociation mechanisms described by equations (2) and (3). In principle, these can be studied in precisely the same way as the present work on charge transfer. This work may be complicated somewhat by the angular divergence and energy spread of the excited H atoms formed by dissociation. Moreover, it will be necessary to assess the influence of vibrationally excited states of the incident projectiles.

IX. Publications and Travel

A total of six reports have been published in the open literature during the present reporting period. These were, in fact, all prepared and submitted at the time of our last report but have been published during the present period. For completeness they are again listed here.

1. "A Comparison of the Excitation Produced in a Helium Target by the Impact of H⁺, D⁺, H₂⁺, H₃⁺ Ions at High Energy," by E. W. Thomas and G. D. Bent. J. Opt. Soc. Am. 58, 138 (1968). AEC Report No. ORO-2591-27.

2. "The Formation of Excited States in a Helium Target by the Impact of 0.15 to 1.0 MeV Protons and Deuterons. I. Experimental," by E. W. Thomas and G. D. Bent. Phys. Rev. 164, 143 (1967). AEC Report No. ORO-2591-28.

3. "The Formation of Excited States in a Helium Target by the Impact of 0.15 to 1.0 MeV Protons and Deuterons. II. Comparison with Theory," by E. W. Thomas. Phys. Rev. 164, 151 (1967). AEC Report No. ORO-2591-29.

4. "Cross Sections for the Formation of Excited States in a Nitrogen Target by the Impact of 0.15 to 1.0 MeV Protons," by E. W. Thomas, G. D. Bent, and J. L. Edwards. Phys. Rev. 165, 32 (1968). AEC Report No. ORO-2591-30.

5. "Emissions from a Molecular Hydrogen Target Induced by the Impact of Fast Protons," by J. L. Edwards and E. W. Thomas. Phys. Rev. 165, 161 (1968). AEC Report No. ORO-2591-31.

6. "The Formation of Excited Ions by the Impact of 0.15 to 1.0 MeV Protons on a Molecular Oxygen Target," by E. W. Thomas and G. D. Bent. Proc. Phys. Soc. J. of Phys. B. 1, 233 (1968). AEC Report No. ORO-2591-32.

With the support of this contract, a complete file is maintained of published data in the field of collisional excitation. A categorized listing of these data sources has been prepared and is being issued as a "Technical Report" at the same time as this Progress Report. This will supersede a similar report issued two years ago.¹³ The details are as follows: "A Listing of Publications Concerning the Formation and Destruction of Excited States by Collisions between Atomic Systems," by E. W. Thomas. Technical Report No. 2, 31 August 1968. AEC Report No. ORO-2591-38.

During the present year, Dr. Thomas gave invited papers to the School of Physics at Auburn University (January 1968) and to the meeting of the Southeastern Section of the American Physical Society in Athens, Georgia (October 1968). Dr. Thomas attended the "International Conference on Atomic Physics" in New York (June 1968). Dr. Thomas and Mr. Ford attended the "Gaseous Electronics Conference" in Boulder, Colorado (October 1968). Visits have also been made to the Oak Ridge National Laboratory for the purpose of consulting with research workers in the Thermonuclear Division.

X. Personnel

The work described in this report was carried out as part of AEC Con-

13. "A Listing of Available Experimental Data on the Formation and Destruction of Excited States by Collisions Between Atomic Systems," by E. W. Thomas. Technical Report No. 1, 30 October 1966. AEC Report No. ORO-2591-22.

tract No. AT-(40-1)-2591 and has been under the jurisdiction of Dr. Thomas. He has devoted one-third time during the academic year to this project and four-fifths time during the summer.

Mr. Lee Edwards continues to work full time on this project and draws half time financial support. The work on charge transfer has been his responsibility and will form the basis of his thesis problem. It is hoped that this work will be completed during the coming contract year and that a thesis will be written at that time.

Mr. John Ford continues to work on this project and to draw half time support. He has recently passed the qualifying exams and has now commenced full time work on the project. It is expected that the work carried out under this program will provide the basis for his Ph.D. thesis problem in the future.

During the course of the present year, assistance has also been received from Mr. R. Landers (Graduate Student), Mr. Floyd Richie (Undergraduate), and Mr. David Jacobi (Undergraduate).

XI. Accelerator Usage

Use of the Van de Graaff accelerator is shared with a project to measure ionization cross sections, also under AEC contract No. AT-(40-1)-2591. The accelerator is designed so that it may be rapidly switched from one experiment to another. A period of one day suffices to make the required mechanical connections and to align the ion beam with the experiment.

XII. Incident Report

There have been no incidents for which a report is required during the performance of the research under this contract in the present reporting period.

E-2013

EMISSION AND EXCITATION
CROSS SECTIONS

ORO-2591-44

PROGRESS REPORT NO. 5

Covering the Period

December 1, 1968 to November 30, 1969

By E. W. Thomas

J. L. Edwards

J. C. Ford



Report No. ORO-2591-44

Contract No. AT-(40-1)-2591

U. S. ATOMIC ENERGY COMMISSION

OAK RIDGE, TENNESSEE



30 November

Engineering Experiment Station
GEORGIA INSTITUTE OF TECHNOLOGY
Atlanta, Georgia

EMISSION AND EXCITATION CROSS SECTIONS

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30 November 1969

TABLE OF CONTENTS

	Page
I. Title	1
II. Introduction	1
III. Abstract	1
IV. Objective	2
V. Experimental Method	4
VI. Summary of Progress Made During the Present Reporting Period	11
(1) Development of the Experimental Technique.	12
(2) The Influence of Cascade, Polarization and Stray Electric Fields on the Validity of the Data . . .	20
(3) Detailed Measurements for Targets of Helium and Nitrogen	23
(4) General Comparison between Experimental Data	28
VII. Program for the Remainder of the Contract Year.	33
VIII. Program for the Future.	34
IX. Publications and Travel	35
X. Personnel	36
XI. Accelerator Usage	37
XII. Incident Report	37

I. Title

Emission and Excitation Cross Sections

II. Introduction

This report summarizes the work performed on excitation phenomena as part of contract AT-(40-1)-2591 for the U. S. Atomic Energy Commission. The work was initiated on March 1, 1965, as a result of Modification No. 7, increasing the scope and financing of the existing contract. The present report covers the period 1 December 1968 to 30 November 1969 which corresponds to the first 9 months of the 12 month period covered by Modification No. 11 to this contract, plus the final three months of the preceding contract period.

A program for the measurement of ionization cross sections which was the subject of the original contract and subsequent Modifications 1 through 11 has been continued and is reported separately.¹

III. Abstract

A study has been made of the charge transfer processes whereby fast neutral atoms are formed in the 3s, 3p and 3d excited states as a result of impact of protons on targets of helium, hydrogen, argon and nitrogen. The technique involves quantitative measurement of Balmer alpha radiation emitted in the spontaneous decay of the excited atom. A time of flight technique allows separation of the 3s, 3p and 3d state contributions to the emission.

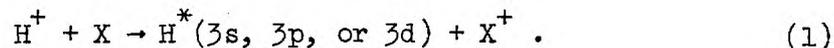
1. D. W. Martin and G. O. Taylor. "Ionization and Charge Transfer Cross Sections," Progress Report No. 22. 30 November 1969. U. S. Atomic Energy Commission Report Number ORO-2591-43.

It is necessary to assess the influence of processes whereby the excited atoms are collisionally destroyed before they undergo spontaneous radiative decay. Detailed measurements of the collisional formation and destruction processes are presented for targets of He and N₂ and impact energies in the range 75 to 400 keV. Preliminary data for the case of H₂ and Ar targets are also shown. Comparison is made with theoretical predictions. The influence of stray electric fields on the validity of the measurements is discussed.

IV. Objective

The objective of this research is the measurement of cross sections for the formation of fast neutral hydrogen atoms in excited states through the impact of fast protons on gaseous targets. Impact energies range from 75 to 400 keV.

The process of primary interest in the present investigation is the direct formation of excited hydrogen atoms in the 3s, 3p and 3d states by transfer of an electron from a target atom.



Targets "X" of helium, hydrogen, argon and nitrogen have been used. It was also intended to study the formation of these same excited states as a result of the dissociation of H₂⁺ and H₃⁺ ions on the same targets. However, due to a slight change in our program it proved impossible to devote any appreciable amount of time to these dissociation processes during the present

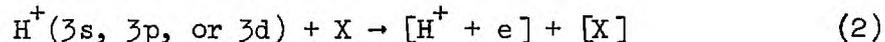
reporting period.

Reasons for choosing these particular reactions as the subject of experimental investigation are discussed in the previous Progress Report.² They may be briefly summarized as follows. The approximations made in theoretical predictions should be tested at every available opportunity in order to establish the validity of calculations in situations where experimental verification is impossible. By measuring separately the cross sections for formation of states having particular principal and angular momentum quantum numbers, a far better test of theory is obtained than if measurements are made only of the sum of cross sections for formation of several states. Stray fields of the magnitudes commonly encountered in experimental systems are sufficient to cause Stark mixing of the sublevels of states having a principal quantum number n of 4 or more, thereby destroying their separate identity. A detailed test of theory is therefore possible only for the $n=2$ and $n=3$ levels. If the validity of theoretical predictions can be established for the $n=3$ states by comparison with experiment, it should be possible to estimate cross sections for the production of states of large n . A knowledge of these cross sections is of value in designing a system for controlled fusion of hydrogen nuclei in which the plasma density is increased by injection of beams of highly excited hydrogen atoms into the containment device. Knowledge of these cross sections is also of practical importance to the understanding of certain phenomena

2. E. W. Thomas, et al., AEC Report No. ORO-2591-37, 30 November 1968.

observed in natural auroras and in the complex situations which exist some milliseconds after an atmospheric nuclear explosion.

A process of secondary interest is the collisional destruction of excited atoms prior to their spontaneous radiative decay.



The brackets are used to indicate that these experiments will give no information on the states of the post-collision products. However, published theoretical predictions³ indicate that nearly all such collisions at the impact velocities of this experiment result in ionization of the hydrogen atom.

V. Experimental Method

The source of the energetic projectiles for the present experiment is a 1 MeV Van de Graaff positive ion accelerator, which is equipped with a beam analyzing and stabilizing system. The ion beam is collimated by a series of small apertures before entering the experimental region. The formation of excited H atoms in the 3s, 3p, and 3d states is detected by the quantitative measurement of the Balmer alpha photons emitted as the excited atoms decay to the n=2 level.

Two distinct experimental arrangements are available. In the first, the photon emission from the H atoms is observed as the beam traverses the target gas (Figure 1). In the second, the beam traverses a gas cell of definite length where the collisional excitation occurs and the emissions are observed from the H atoms as they emerge from the cell and proceed through an evacuated flight tube (Figure 2). After traversing the collision

³. D. R. Bates and J. C. G. Walker, Planetary and Space Science, 14, 1367 (1966).

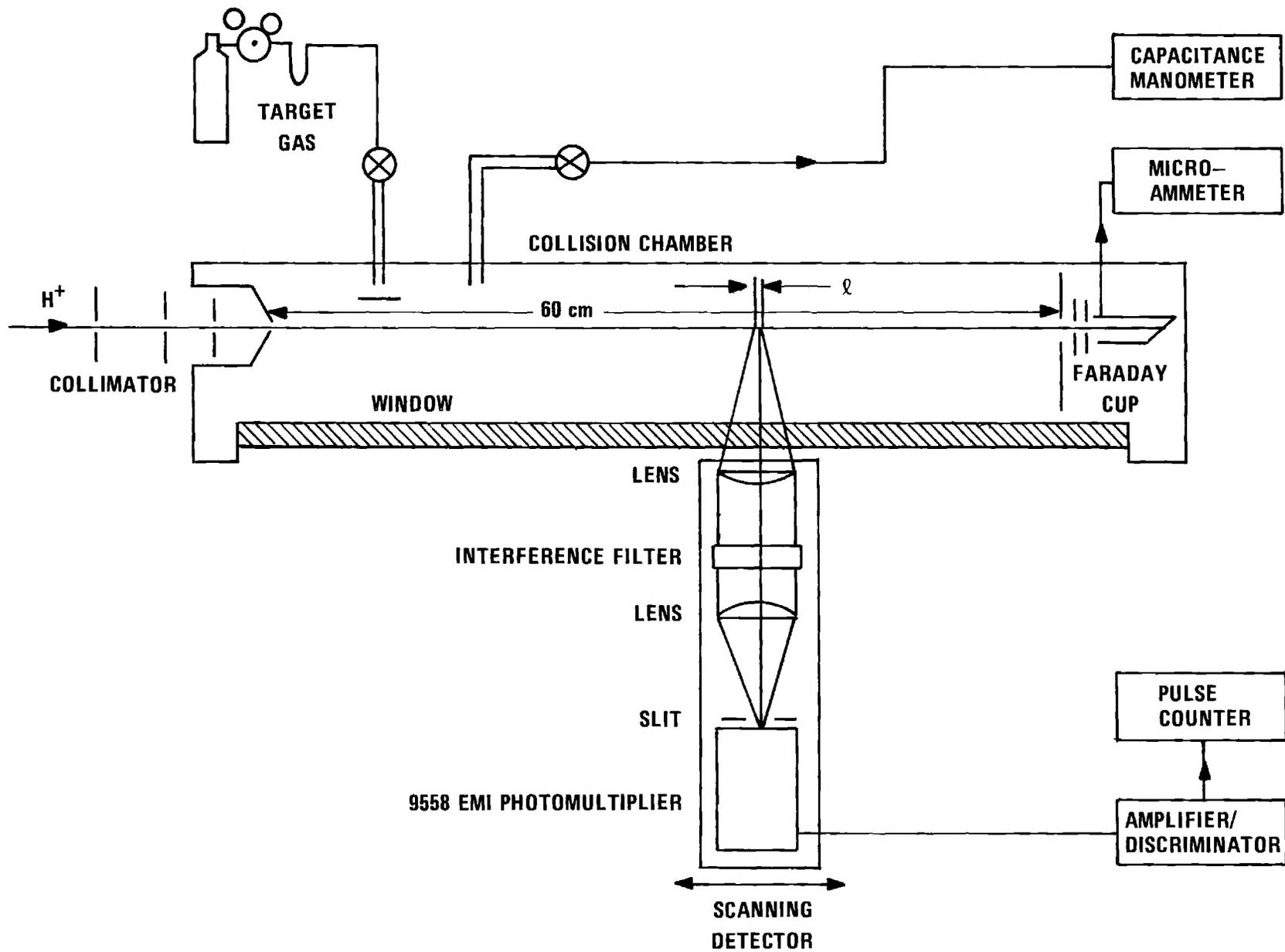


Figure 1. Schematic Diagram of the Apparatus for Measuring Emission in the Target Region.

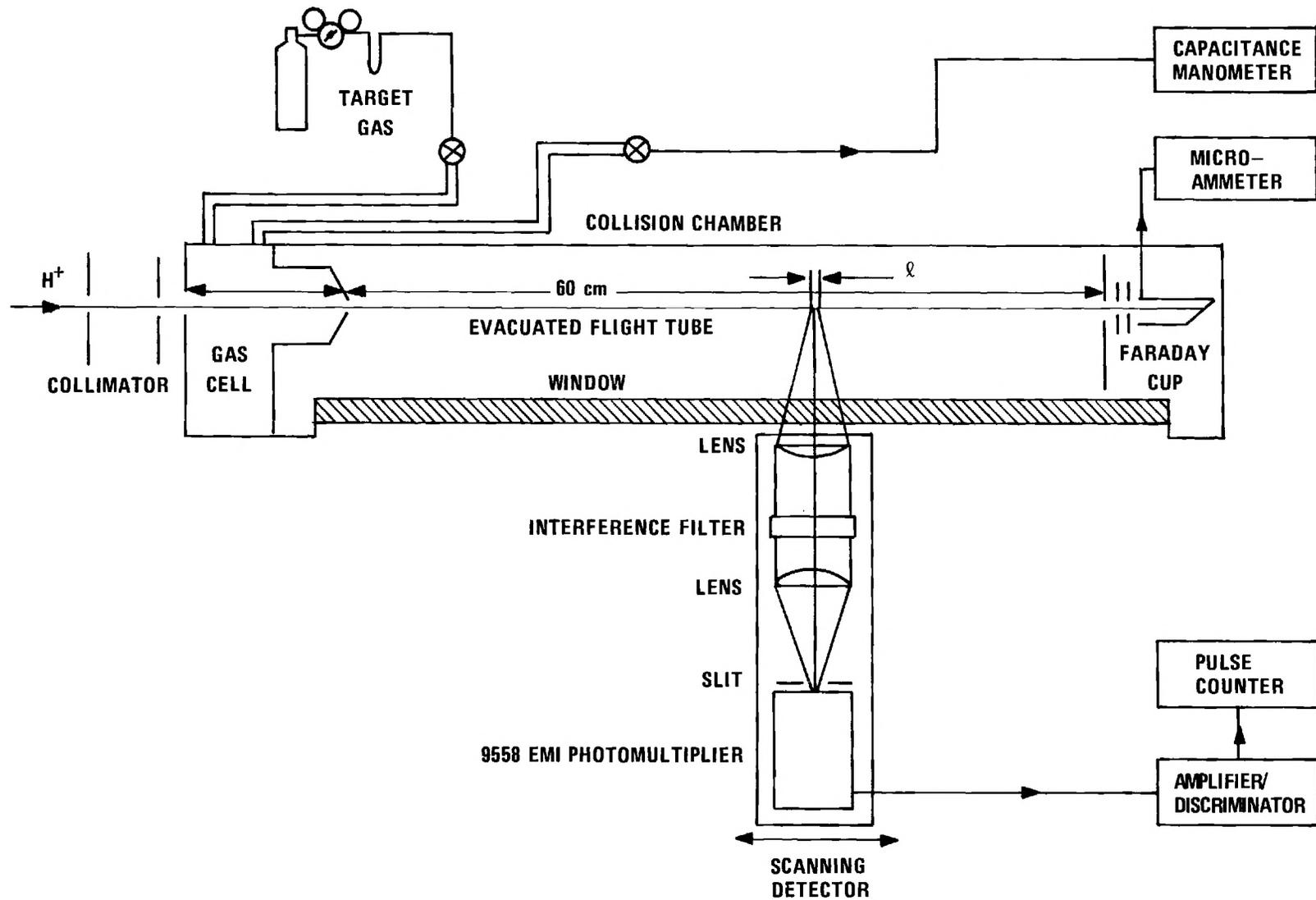


Figure 2. Schematic Diagram of the Apparatus for Measuring Emission from the Beam which has Passed Through a Short Gas Cell.

and detection regions, the ion beam is monitored on a conventional Faraday cup.

At the high impact energies utilized in this experiment, the product of the projectile's velocity and the lifetime of the excited state is a length comparable with the dimensions of the apparatus. Therefore, in general, a projectile will move an appreciable distance from the point where it was excited before emitting a photon and decaying to a lower state. As a result, the intensity of emission from the projectiles is a function of the position along the flight path where the observation is made. Measurement of the spatial variation of this intensity allows the evaluation of the population of the emitting state.

The present experimental apparatus consists of a gas cell followed by a long flight tube. Plate glass windows in the flight tube allow observation of the entire flight path beyond the exit aperture of the gas cell. Detection of the Balmer alpha radiation is carried out with a photomultiplier tube fitted with an interference filter. The detector can be moved along a machined track to measure emission intensity at any position along the flight path. For the first mode of operation the target gas is introduced into the flight tube and the gas cell is evacuated. For the second mode, the target gas is introduced into the cell and the flight tube is evacuated. Target gas pressure is measured by a differential capacitance manometer which has been calibrated against a McLeod gauge.

We now consider separately the mathematical formulation which predicts the intensity variation in the two experimental configurations identified above.

First consider the configuration where measurements are made of intensity from the path of the beam through the target (Figure 1). Let us suppose that the apparatus detects the photons emitted as result of the decay of the state j to a lower state k . The state j has a lifetime τ_j , and the cross section for its formation by the mechanism of equation (1) is Q_j . The apparatus measures the flux of photons J_{jk} emitted per second in the transition $j \rightarrow k$ from a length l centimeters of the beam path. A flux of F projectile ions per second are incident with a velocity v (cm/sec) on a target which has a number density n (molecules/cc). In general, the state j may decay by many paths to lower states i , only one of which ($j \rightarrow k$) is detected experimentally. Therefore, the detected photons correspond to the fraction $A_{jk} / \sum_{i < j} A_{ji}$ of the total number of decays. Here A_{jk} (sec^{-1}) is the probability of spontaneous decay of the state j to the state k and this fraction is known as the branching ratio. If the observation length l is made much shorter than $v\tau_j$, and the target density is kept sufficiently low that single collision conditions are maintained, then it may be shown⁴ that at a point x cm from the entrance to the target region the photon flux is given by

$$J_{jk}(x) = F n l Q_j \frac{A_{jk}}{\sum_{i < j} A_{ji}} \left[1 - \exp \left(- \frac{x}{v\tau_j} \right) \right]. \quad (3)$$

In arriving at this result, we have made the assumptions that the proton beam has not been depleted significantly in passing through the target region and that population of the state j by cascade from higher levels is sufficiently small that it may be neglected. These assumptions must be subjected

⁴. See, for example, C. E. Head and R. H. Hughes, Phys. Rev. 139, A1392 (1965).

to further scrutiny. In the present experiment, the Balmer alpha emission is due to three transitions; $3s \rightarrow 2p$, $3p \rightarrow 2s$, and $3d \rightarrow 2p$. These all emit photons of essentially the same wavelength and are therefore detected simultaneously. The observed flux is a sum of three terms of the type given in equation (3).

$$J_{\alpha}(x) = I_0 \left[1 - \exp \left(-\frac{x}{v\tau_0} \right) \right] + I_1 \left[1 - \exp \left(-\frac{x}{v\tau_1} \right) \right] + I_2 \left[1 - \exp \left(-\frac{x}{v\tau_2} \right) \right] \quad (4)$$

where

$$I_j = F n \ell Q_j \frac{A_{jk}}{\sum_{i<j} A_{ji}}$$

The subscripts 0, 1, and 2 are used to indicate, respectively, the $3s$, $3p$ and $3d$ states. In fact, the $3s$ and $3d$ states can decay only by the Balmer alpha transition and therefore the branching ratio $A_{jk} / \sum_{i<j} A_{ji}$ for these states is unity. For the $3p \rightarrow 2s$ transition, this ratio is 0.118, indicating that only 11.8% of the atoms in the $3p$ state decay by the emission of a Balmer alpha line, the rest by the Lyman beta. Equation (4) represents a sum of three terms which increase exponentially with x towards some constant value. Because the three lifetimes of the states are quite different, it is possible to compare the measured function $J_{\alpha}(x)$ with equation (4) and to evaluate the coefficients I_j . In this manner, the cross section for the formation of the $3s$, $3p$ and $3d$ states may be measured using the different lifetimes to identify the three sublevels.

The second experimental configuration involves observation of the decay of the excited states in the beam emerging from a gas cell into an evacuated flight tube (Figure 2). In this case the intensity of emission from each

state will simply decay exponentially with distance along the flight tube with a decay length characterized by the lifetime of the excited state. The population of the excited states in the emergent beam will be a function of the cell length L . The photon flux emitted in the transition $j \rightarrow k$ as a function of the distance x beyond the exit from the gas cell may be shown⁵ to be given by the following equation.

$$J_{jk} = F n \ell Q_j \frac{A_{jk}}{\sum_{i < j} A_{ji}} \left[1 - \exp\left(-\frac{L}{v\tau_j}\right) \right] \exp\left(-\frac{x}{v\tau_j}\right) \quad (5)$$

Again the Balmer alpha photon flux is in fact the sum of three terms and can be represented by the equation

$$J_{\alpha}(x) = I_0 \left[1 - \exp\left(-\frac{L}{v\tau_0}\right) \right] \exp\left(-\frac{x}{v\tau_0}\right) + I_1 \left[1 - \exp\left(-\frac{L}{v\tau_1}\right) \right] \exp\left(-\frac{x}{v\tau_1}\right) + I_2 \left[1 - \exp\left(-\frac{L}{v\tau_2}\right) \right] \exp\left(-\frac{x}{v\tau_2}\right) \quad (6)$$

I_j has the same significance as before. This equation may be fitted to the observed emission and the cross sections evaluated.

The two experimental configurations are complementary, each having different advantages and drawbacks. Observations made in the target region may be subject to interference from collisionally induced target gas emissions. These will be invariant with beam penetration through the gas and will require the inclusion of an additional constant term to equation (4). Unless this constant is small in comparison with the other terms, it is impossible to evaluate the separate $3s$, $3p$ and $3d$ excitation cross

5. R. H. Hughes, et al., Phys. Rev. 146, 53 (1966).

sections with any accuracy. In particular, the interesting case of an H_2 target becomes quite impossible due to target emission. The approach of using a cell and an evacuated flight tube not only allows the use of an H_2 target but also enhances the populations of the short-lived $3p$ and $3d$ states relative to the $3s$ population, which tends to dominate in the other configuration. This enhancement is possible because the $3p$ and $3d$ populations will approach their equilibrium (maximum) values within a short distance (10 to 20 cm) of the entrance to the target region, whereas the long-lived $3s$ state may only achieve 20% of its maximum population in the length of the cell. The gas cell approach requires care to ensure that the exit aperture from the cell does not intercept an appreciable fraction of the scattered projectiles. Furthermore, there is an uncertainty as to the "thickness" of a gas cell due to pressure gradients at the two apertures. An assessment of such end effects can be made by varying the length of the gas cell. It is our judgement that both configurations should be employed and that agreement between measurements made using the two approaches will suggest that the experiments are operating satisfactorily.

VI. Summary of Progress Made During the Present Reporting Period

During this reporting period considerable progress has been made on the development of the equipment and the academic study of the collision problems of interest. The major development of equipment has been the successful introduction of the gas cell mode of operation of the experiment. The study of charge transfer into the excited state has concentrated on the use of targets of helium and nitrogen. Early in this reporting period it was discovered that the cross section for collisional destruction

of the excited atom (Equation (2)) is very high. This resulted in an appreciable fraction of the excited atoms in the $3s$ state being collisionally destroyed before spontaneous radiative decay. In principle this problem should be removed by operating the experiment at a sufficiently low target density that collisional destruction is negligible. In practice it is not possible to achieve this condition while maintaining sufficient signal strength to provide adequate statistical accuracy. This problem has been overcome by measuring the cross section for the destruction process and thereby assessing its influence on the measurement of the population of the $3s$ state.

(1) Development of the Experimental Technique

During the present reporting period the final developments of the experimental apparatus and techniques envisaged in our proposals have been completed. The gas cell mode of operation of the experiment has been brought into use. The detection efficiency of the optical system has been determined with due allowance for Doppler shift and broadening of the emission. These developments are reviewed below. For other experimental details the reader is referred to our previous report on this apparatus.²

(a) Introduction of the "Gas Cell" Mode of Operation

In this reporting period the gas cell approach to the pursuit of this problem has been brought into operation. At the same time the opportunity has been taken to improve the vacuum pumping systems on the apparatus and the collimation of the incident ion beam. Tests of the new system are completely satisfactory. It is expected that the gas cell configuration will have considerable advantages over the method of observing

emission within the collision chamber itself that has been utilized in most of the studies reported herein. The new arrangement essentially separates the analysis of the excited state population from the formation of the excited states. This makes the analysis technique independent of any emission that may be produced from the target gas and independent of any processes of collisional destruction of the excited atom. Moreover it has the advantage of enhancing the population of the short-lived $3p$ and $3d$ states relative to the long-lived $3s$ state in the region of observation. This should improve the accuracy with which the $3p$ and $3d$ state cross sections can be measured. It may also tend to reduce the accuracy with which the $3s$ state is determined.

As examples of the output from this device figures 3 and 4 display the variation of emission with distance from the exit of the gas cell for the four target gases under study. The variation of emission should be represented by a sum of three exponential decays like equation (6). It is quite clear that, at large x the emission is represented by a single exponential decay characteristic of the $3s$ state lifetime. Subtracting the $3s$ state contribution from the total intensity should give the contributions from the $3p$ and $3d$ states. A fit of two exponential decays of the required lifetimes to the short-lived component allows the deconvolution of the $3p$ and $3d$ state contributions. In figures 3 and 4 are shown a synthesized curve of emission against x obtained using the three excited state populations derived from the analysis. The excellent agreement gives considerable confidence that the experiment is operating in a manner consistent with the theoretical prediction of equation (6). It appears that there is no

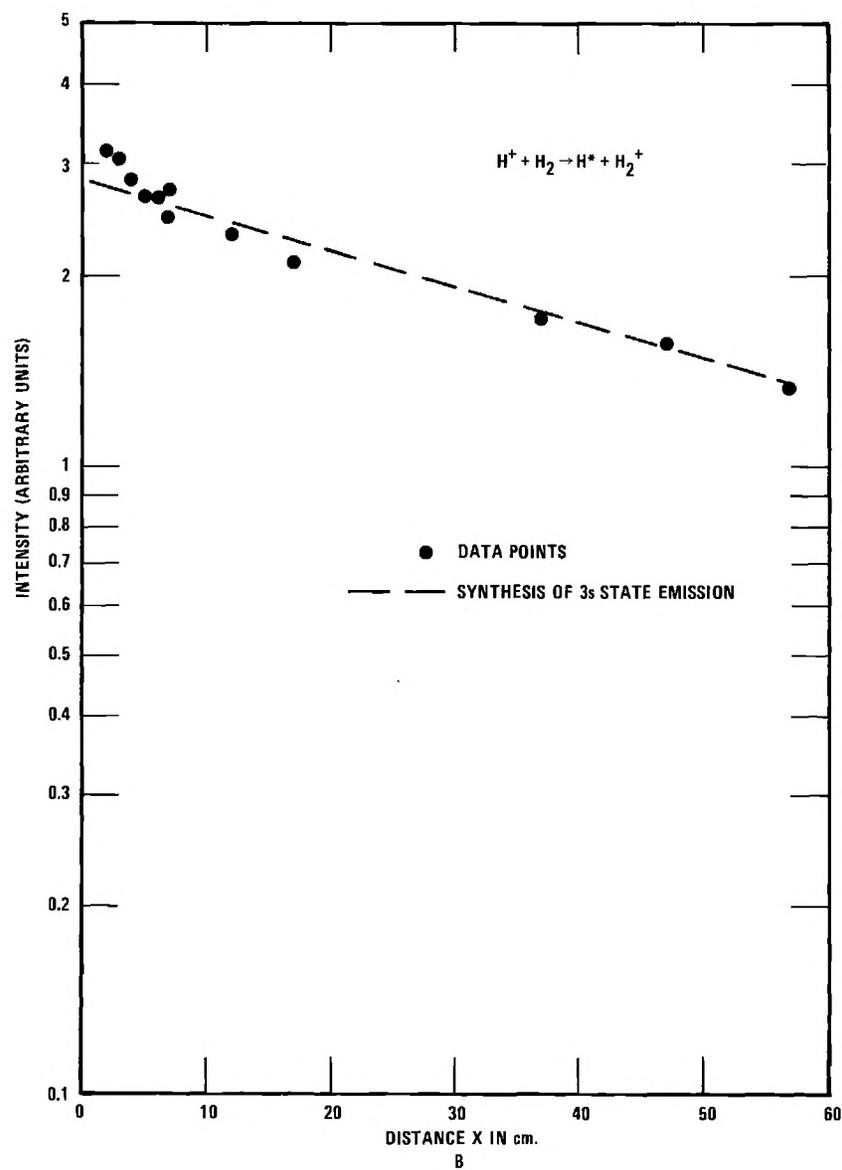
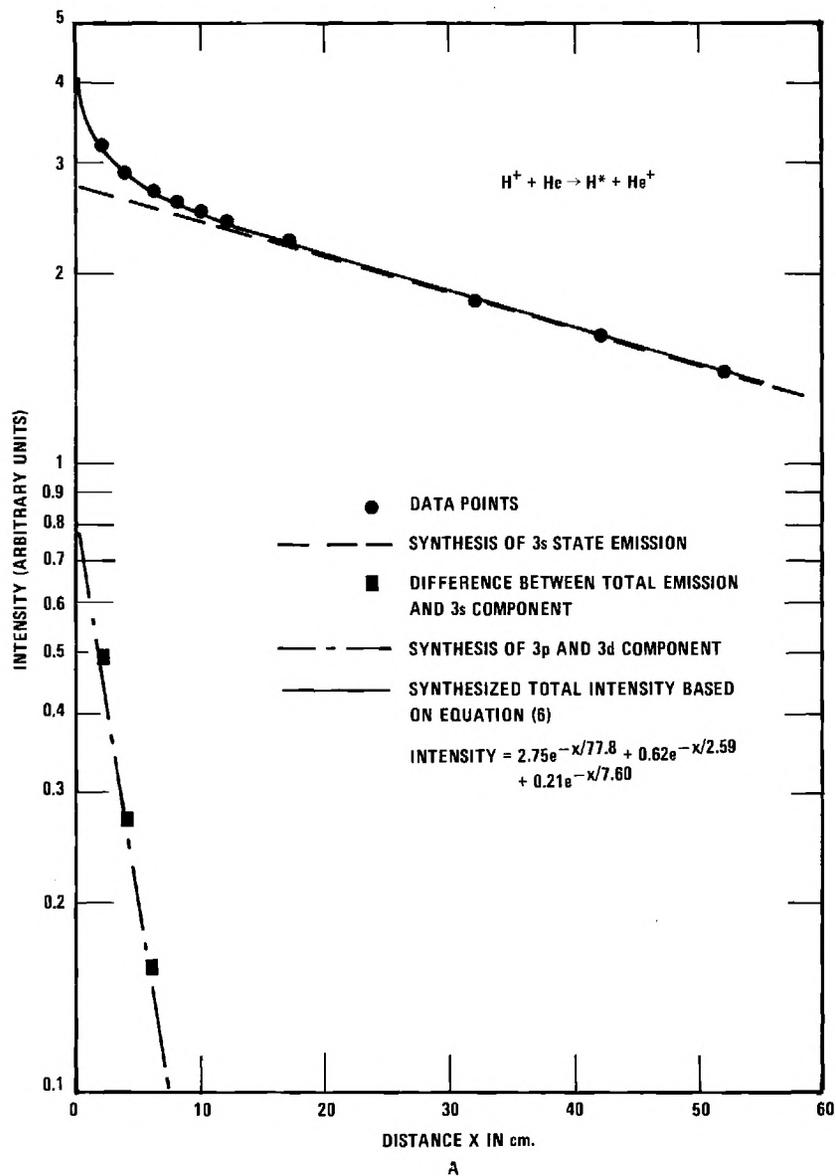


Figure 3. Intensity of Balmer Alpha Emission as a Function of Distance from the Exit of the Gas Cell for Targets of Helium and Hydrogen. Energy 125 keV, Target Pressure 10^{-3} Torr. Analysis in Terms of Equation (6) gives the Cross Sections for Forming the 3s, 3p, and 3d States with the Helium Target as 1 : 0.266 ; 0.013. The Scatter in the Data for the Hydrogen Case is too Great to Allow a meaningful Deconvolution of the 3p and 3d State Contributions.

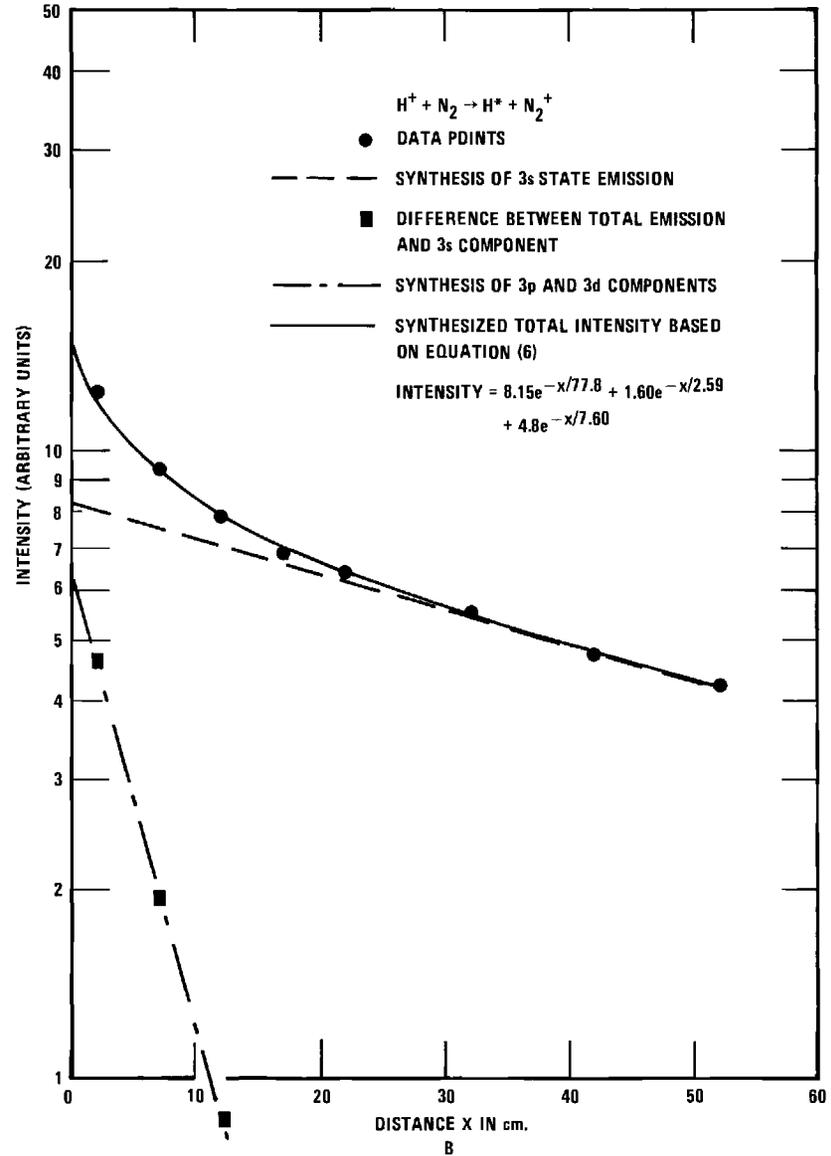
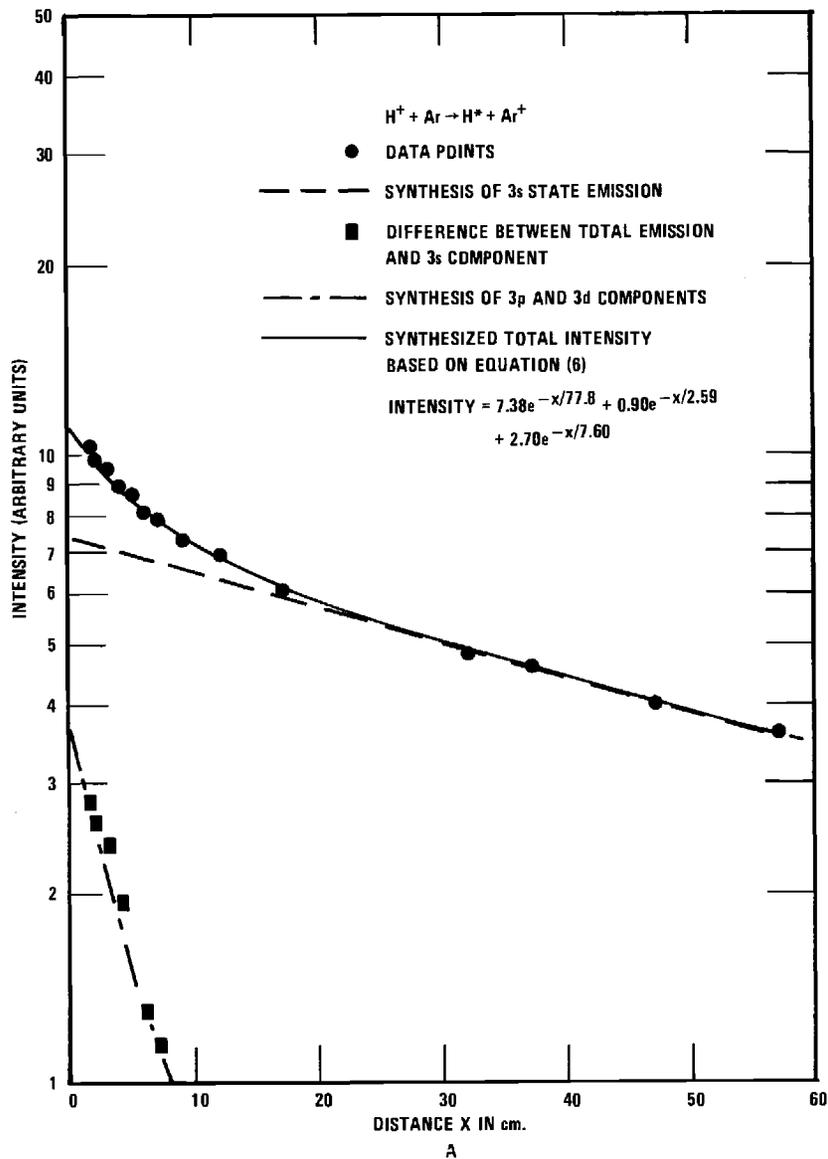


Figure 4. Intensity of Balmer Alpha Emission as a Function of Distance from the Exit of the Gas Cell for Targets of Argon and Nitrogen. Energy 125K, Target Pressure 10^{-3} Torr. Analysis in Terms of Equation (6) gives the Cross Sections for Forming the 3s, 3p and 3d States in the Ratios 1; 0.144; 0.064 for Argon and 1 : 0.232 : 0.104, for Nitrogen.

significant contribution from longer lived states that populate the $3s$, $3p$ and $3d$ states by cascade.

(b) The Assessment of the Effect of Doppler Shift on the Sensitivity of the Optical System

The arrangement of the detector of Balmer alpha (H_{α}) photons is indicated in Figure 1. Light emitted within a 12° cone at 90° to the beam axis is focused at infinity by a lens, filtered by a narrow band H_{α} interference filter, and focused by a second lens to form an image of the beam on the cathode of an EMI 9558 photomultiplier tube. A mask restricts the photomultiplier's view to a 6mm length of beam. This length is sufficiently short to insure that a negligible error (less than 0.2%) is introduced by assuming that the emission intensity per unit length of beam at the point of intersection of the beam axis and the optical axis of the detector assembly is the observed flux divided by the length of beam within view.

The high velocity of the radiating hydrogen atoms (0.012 c to 0.03 c for 75 to 400 keV energies) causes significant Doppler shifts in the wavelength of the observed radiation. Although the optical axis of the detector is at 90° to the beam axis, the finite aperture of the optical system admits radiation emitted at angles from 78° to 102° to the beam axis. The wavelength of H_{α} radiation observed at exactly 90° to the beam axis is increased by relativistic time dilation, 0.5 Å at 75 keV to 2.1 Å at 400 keV; and the Doppler spread of emissions accepted by the finite aperture ranges from 34 Å at 75 keV to 80 Å at 400 keV.

Because the Doppler shifts vary with the velocity of the emitting particle, the effective sensitivity of the detector varies with the impact

energy of the incident protons. It should be emphasized that this dependence has no effect on measurements of the relative magnitudes of Q_{3s} , Q_{3p} , and Q_{3d} at a given energy, but it will affect the apparent dependence on energy of these cross sections.

The determination of detection sensitivity in the presence of the Doppler shift and broadening is complicated and will not be discussed in detail here. However, we can summarize the approach as follows. The optical system may be divided into a series of segments by drawing imaginary chords across the circular aperture perpendicular to the beam direction. (See figures 5 and 6). Light which traverses the optical system through a particular section of the aperture must be emitted at a definite angle θ to the beam path and therefore exhibits a wavelength λ calculated from the well known equation for Doppler shift. In practice, of course, each segment transmits light from a small range of θ and having a small range of wavelengths λ . However, by making the widths of the apertures sufficiently small the error involved in the assumption of unique values of θ and λ can be made negligibly small. One may determine the signal due to light falling on a segment of the aperture in terms of the detection sensitivity of the photomultiplier $D(\lambda)$, the transmittance of the aperture $T(\theta)$, and the transmittance of the filter to the appropriate wavelength $t(\lambda)$. The total signal is given by the sum of all the contributions from the separate segments. The efficiency of the optical system, defined as the ratio of signal to the emitted light intensity, is given by

$$\eta = \sum_k T(\theta_k) t(\lambda) D(\lambda) \quad (7)$$

Here k refers to the segment number k of the optical aperture.

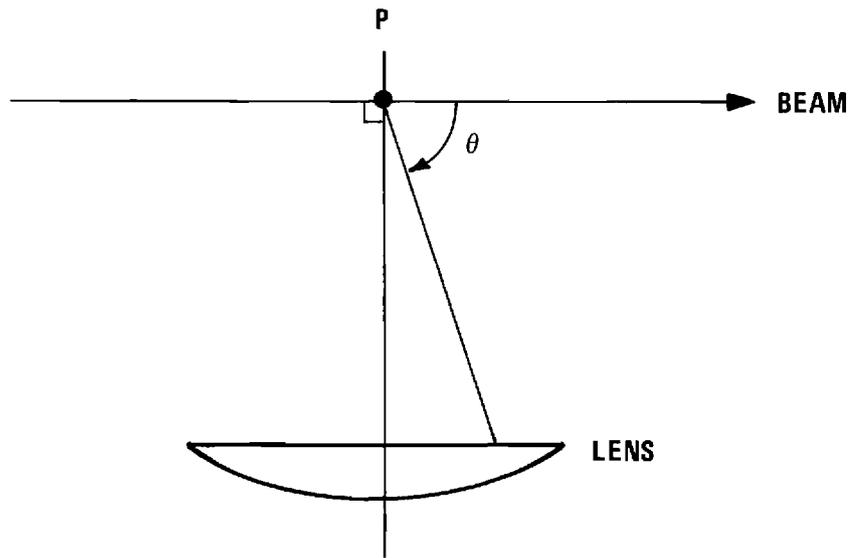


Figure 5. Geometry of the Optical Aperture.

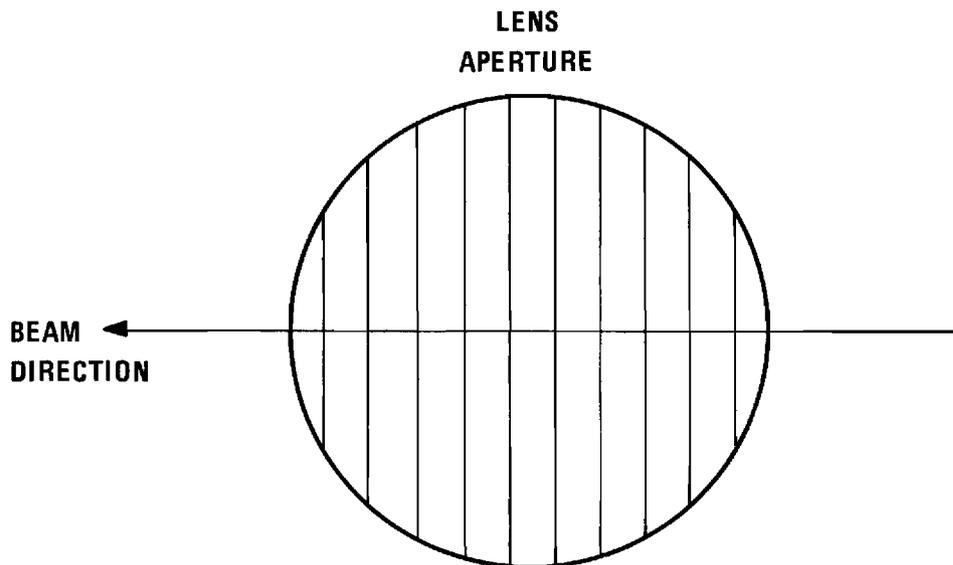


Figure 6. Division of the Optical Aperture into Segments for Measurement of $T(\theta)$. (See Text).

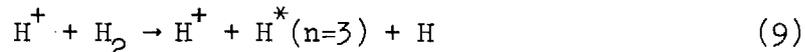
If one writes $t(\lambda)$ as $t_0\alpha(\lambda)$ and $D(\lambda)$ as $D_0\beta(\lambda)$ then equation (7) becomes

$$\eta = t_0 D_0 \sum_k T(\theta_k) \alpha(\lambda_k) \beta(\lambda_k) \quad (8)$$

Here t_0 can be taken as the value of $t(\lambda)$ at the wavelength of the Balmer alpha line and D_0 as the value of $D(\lambda)$ at that same wavelength. The relative variation of transmittance of the filter with wavelength, $\alpha(\lambda)$, is obtained by direct measurement. The relative variation of photo-multiplier detection sensitivity with wavelength, $\beta(\lambda)$, is taken from the manufacturer's published data. The transmittance of each segment of the optics, $T(\theta)$ is obtained by direct measurement. As will be shown in the next section it proves unnecessary to determine absolute values of t_0 and D_0 .

(c) Calibration of Detection Sensitivity

Absolute calibration of the measured cross sections has been accomplished by comparison of H_α emission intensities obtained from the charge exchange process with those obtained from the dissociative excitation of molecular hydrogen:



The cross section for emission of the H_α line in this reaction was measured previously⁶ in this laboratory with an estimated uncertainty of $\pm 25\%$. The emitting atoms in this process have low velocities, and since Doppler effects are now negligible, the expression for the efficiency of the detector reduces

6. J. L. Edwards and E. W. Thomas, Phys. Rev. 165, 16 (1968).

to

$$\eta(0) = t_0 D_0 \sum_k T(\theta_k) \quad (10)$$

In determining absolute charge exchange cross sections for particles traveling with velocity v , ratios of detector efficiency are required:

$$\frac{\eta(v)}{\eta(0)} = \frac{\sum_k T(\theta_k) \alpha(\lambda_k) \beta(\lambda_k)}{\sum_k T(\theta_k)} \quad (11)$$

t_0 and D_0 drop out of the ratio, and only the variations of t and D with wavelength enter into the calculation.

(2) The Influence of Cascade, Polarization and Stray Electric Fields on the Validity of the Data

In order to assess the reliability of the measurements achieved in the present reporting period it is necessary to discuss the possible influence of cascade, polarization and the Stark effect mixing of states. It is our assessment that these problems do not influence the measurements by an amount which is significantly greater than the statistical accuracy of our data.

(a) Cascade

In addition to direct collisional excitation, the $3s$, $3p$ and $3d$ states may also be populated by cascade from higher levels. This has two important consequences. First, the measured cross section will then not represent only the formation of the state by collision but will include a component due to cascade. Secondly, and perhaps more important, the dependence of emission intensity on distance will be different for atoms populated by cascade than

for atoms populated by direct excitation. The cascade process will be dependent on both the lifetime of the parent level of the cascade transition and also the lifetime of the $n=3$ state that is populated. This second problem might invalidate the analysis of the separate cross sections which uses a deconvolution technique based on the $3s$, $3p$ and $3d$ state lifetimes.

One may estimate the population of higher n states by taking the present measurements of the $3s$, $3p$ and $3d$ state cross sections and extrapolating them to higher n states assuming that cross sections for a given angular momentum substate decrease as n^{-3} . This general rule is well established⁷ by theory and serves well for an approximate assessment of the problem. Taking due account of the branching ratios for decay of higher states the population of the $n=3$ level by cascade from higher np and nd states is very small and will influence the data by an amount that is smaller than the statistical reproducibility of our measurements. There is no method by which one can reasonably estimate the population of higher nf states. However, all theoretical predictions suggest that it is far less than for the corresponding nd state. Therefore, it too can be neglected. The only cascade contribution of any significance is from the ns states into the $3p$ level. At low energies, which constitutes the worst case for this problem, the contribution from higher ns states might amount to as much as 2.3% of the total emission. Generally, it will be less. It is intended to carry out a computerized synthesis of the influence of this cascade to determine whether it introduces any significant distortion of

7. J. R. Oppenheimer, Phys. Rev. 31, 349, 1928.

the deconvolution procedures. The result of this computer synthesis will provide an estimate of the error that must be assigned to our data through this cause. It is expected that this error will be rather small compared with the other known errors.

(b) Polarization

Emission from the 3p and 3d states may exhibit polarization. The polarization fraction is related to the population of the different magnetic quantum number sub-levels, and is zero if these levels are all equally populated. It may be shown that the collisionally induced emission will be anisotropic if polarization is present. A measurement of emission at one angle does not allow the determination of a cross section unless correction is made for this anisotropy.

Much of the research discussed in this report has been directed at the 3s-2p emission which is unpolarized and therefore emitted isotropically. No attempt has been made to measure polarization for the 3p-2s and 3d-2p emissions. Because of the small signal intensity the statistical accuracy would be so poor as to render the measurement meaningless. Consequently, it is not known whether the emissions are isotropic. In the highly unlikely event that the emission is 100% polarized then the apparent cross section for emission at 90° to the beam path will differ from the true cross section by 50%. There is good reason to believe that in fact the populations of the different magnetic quantum number states will be equal, resulting in zero polarization and absence of anisotropy.

(c) Stark Effect Mixing

Small fields will, in principle, cause Stark mixing of the 3s, 3p and 3d states. There is a danger that stray fields in the apparatus might be

sufficient to cause this effect. Tests have shown that the 3s state is not affected by any fields that might conceivably exist in our apparatus. It has not proved possible to carry out tests on the 3p and 3d states with any meaningful statistical accuracy. At the present time there is no evidence that these states are mixed by the fields that exist in the apparatus.

(3) Detailed Measurements for Targets of Helium and Nitrogen

A detailed study has been made of the formation of the excited H atoms by charge transfer in targets of helium and nitrogen. The influence of the collisional destruction process has introduced considerable complexities into the analysis but these problems have been overcome. Comparisons are made between theory and experiment. Data have been obtained by measurement of emission from the collision region.

(a) The Influence of Collisional Destruction

The collisional destruction of excited atoms (Equation (2)) has the effect of reducing the effective lifetime of the excited state. As a result it is necessary to alter equation (4) by the introduction of a term $(1/v\tau + nQ_1)$ to replace the term $(1/v\tau)$. Here n is the density in the target region and Q_1 is the cross section for the destruction process. In addition there are some corresponding changes to the factors I_0 , I_1 and I_2 of equation (4). If nQ_1 is much smaller than $1/v\tau$ then it may be neglected and the analysis of the experiment is as described in section V of this report. In principle this can be achieved by making the target density n sufficiently small. In practice the cross section Q_1 is very large and it is not possible to reduce the target density sufficiently to remove the influence of the destruction process without causing unacceptable reductions

in signal intensity. High statistical accuracy is a necessary requirement for the deconvolution of the emission variation into three parts with the characteristic lifetimes of the three relevant excited states. This cannot be achieved with low signal levels.

The process of collisional destruction is a mechanism of some considerable intrinsic interest. In fact our previous proposals had listed such mechanisms as a possible subject for future study. It was decided that the best method of handling its influence on the present experiments was to measure it directly along with the cross sections for creation of the excited states, that had been our original aim. Measurement of Q_1 can be obtained from a fit of the modified form of equation (4) to the experimental data. To ensure the validity of the fit, data were taken for two or three different pressures and the derived value of Q_1 shown to be independent of pressure.

(b) Measured Values of Charge Transfer Cross Sections

Measurement of the cross section for the formation of the excited states of atomic hydrogen are shown in figures 7 through 10 for targets of He and N_2 . Uncertainty in the absolute values of cross sections is estimated to be $\pm 50\%$. Half of this comes from uncertainty in the emission cross section data to which the present work was normalized. Uncertainty in the relative variations of cross sections with energy are indicated with error bars.

Figure 7 shows the cross section for the formation of the $3s$ state for protons incident on helium. For comparison the predictions by Mapleton and the previous measurements by Hughes et al.⁵ and by Andreev et al.⁸ are

8. E. P. Andreev et al., Fifth International Conf. on the Physics of Electronic and Atomic Collisions. (Nauka Publishing House, Leningrad, 1967) p. 307.

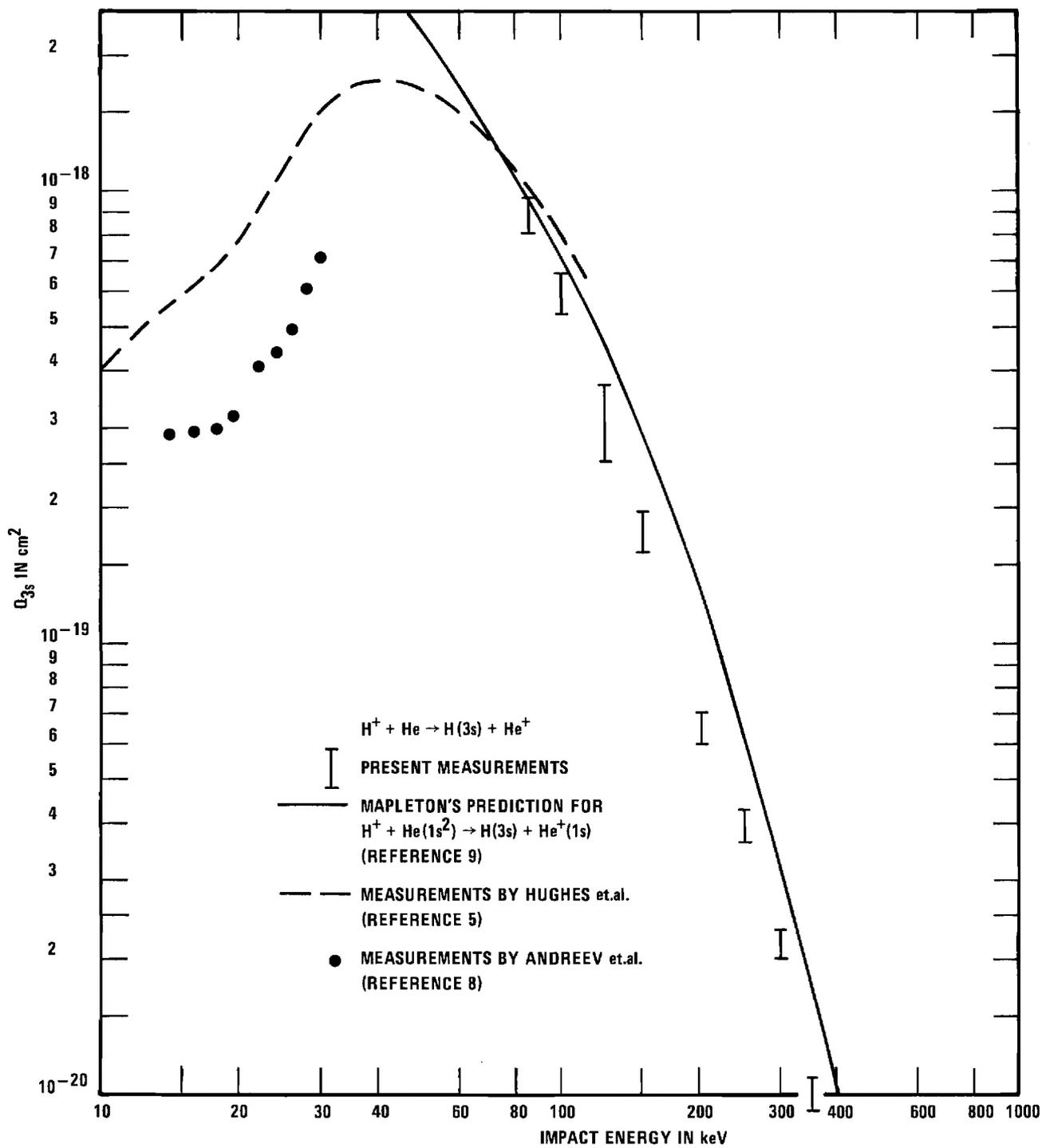


Figure 7. Cross Section Q_{3s} for Charge Transfer into the 3s State of H from a Target of Helium.

also shown. The general form of our measurements is in agreement with Mapleton's predictions.⁹ The systematic discrepancy between theory and experiment might be due to an erroneous calibration of detection sensitivity. It appears that the present measurements confirm the general validity of Mapleton's theory down to impact energies of 75 keV.

Figure 8 presents measurements of the cross section for the formation of the 3d state, again compared with predictions of Mapleton.⁹ This cross section is about two orders of magnitude smaller than the cross section for the formation of the 3s state, which means that only a few percent of the measured light intensity is due to transitions from the 3d state. Statistical fluctuations in the data are typically 1 or 2%, sometimes larger. As a result it has been necessary to assign very large random error bars to the data for this state. Nevertheless the agreement between theory and experiment is surprisingly good.

The measurements of the cross sections for the formation of the 3p state are so poor that it would be misleading to present them here. They do allow us to establish an upper bound to the cross section for the formation of the state. This bound lies below the theoretical predictions of Mapleton⁹ by a factor of between five and ten at all energies from 75 to 400 keV. This very large discrepancy is most surprising. There seems no obvious reason why theoretical predictions should be good for the 3s and 3d states while being very poor for the 3p level. More recently we have determined the cross section for the 3p state utilizing the gas cell configuration of the

9. R. A. Mapleton. Phys. Rev. 122, 528 (1961).

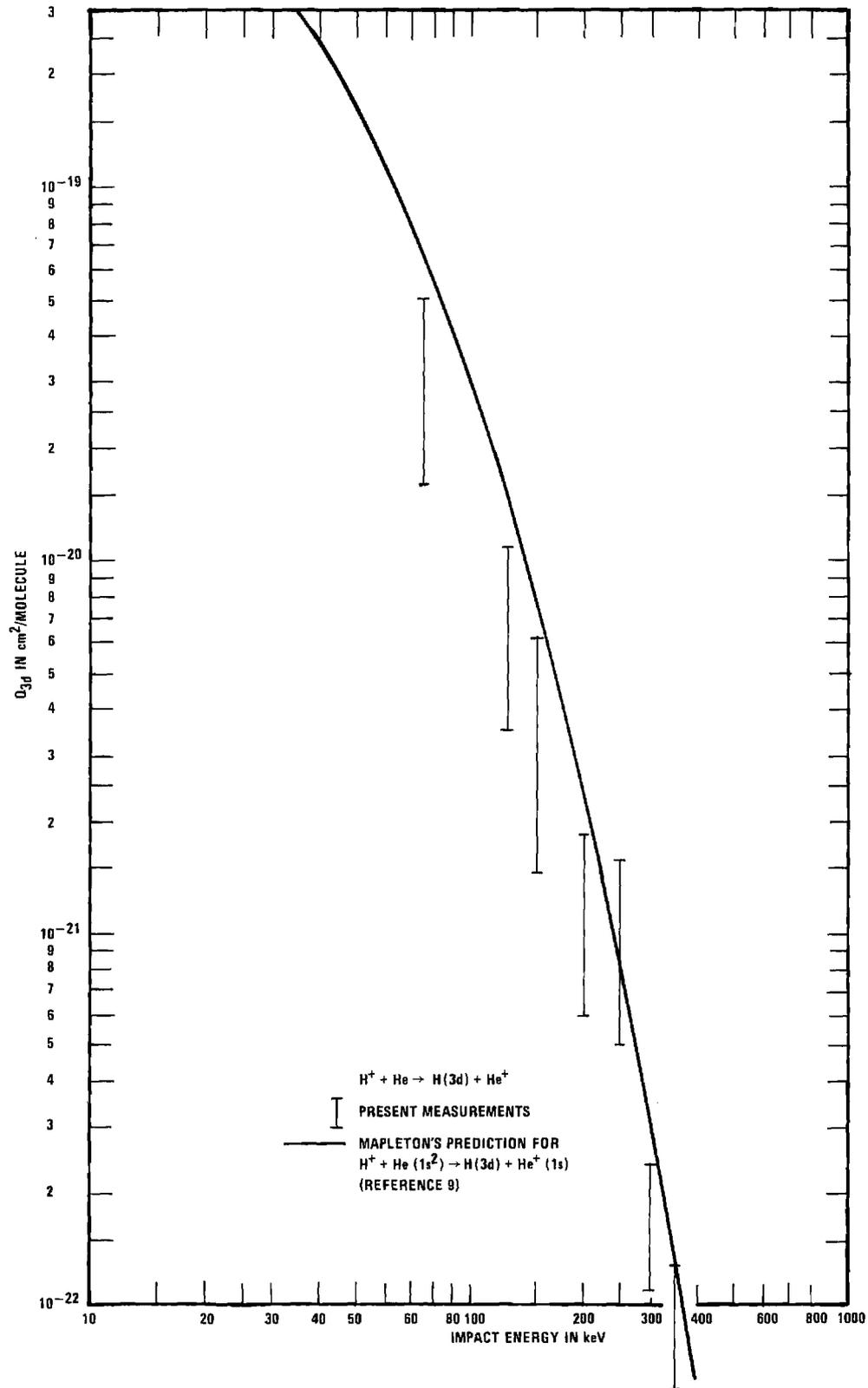


Figure 8. Cross Section Q_{3d} for Charge Transfer into the 3d State of H from a Target of Helium.

experiment. This new procedure provides a much better measurement on the short-lived states than was obtainable with the previous scheme. The discrepancy exists in that data also; the $3p$ state cross section lies far lower in magnitude than theory predicts. There are also general theories for the prediction of relative populations of excited states from the work of Hiskes,¹⁰ and of Butler and May.¹¹ These theories are in general agreement with the work by Mapleton⁹ and therefore in disagreement with experiment. It must be concluded that theory is in error.

Figures 9 and 10 show the cross sections for formation of the $3s$ and $3d$ states of H by impact of H^+ on a target of N_2 . There are no detailed theoretical predictions with which these may be compared.

Figures 11 and 12 show the cross sections for the destruction of the excited state by impact on He and N_2 targets. Comparison is made with the theoretical predictions of Bates and Walker.³ General agreement is found between theory and experiment.

(4) General Comparisons between Experimental Data

It is always interesting to examine the general behavior of collision cross sections to see whether any general behaviors emerge that can be useful for order of magnitude predictions for other cases. In the following table we show the relative cross sections for populating the $3s$, $3p$ and $3d$ states, expressed as a percentage of the total cross section for formation of the $n=3$ level. The data were obtained from the new "gas cell" configuration of the experiment which should produce fairly accurate estimates of the $3p$

10. J. Hiskes, Phys. Rev. 180, 146, 1969.

11. Butler and May, Phys. Rev. 137, A10, 1965.

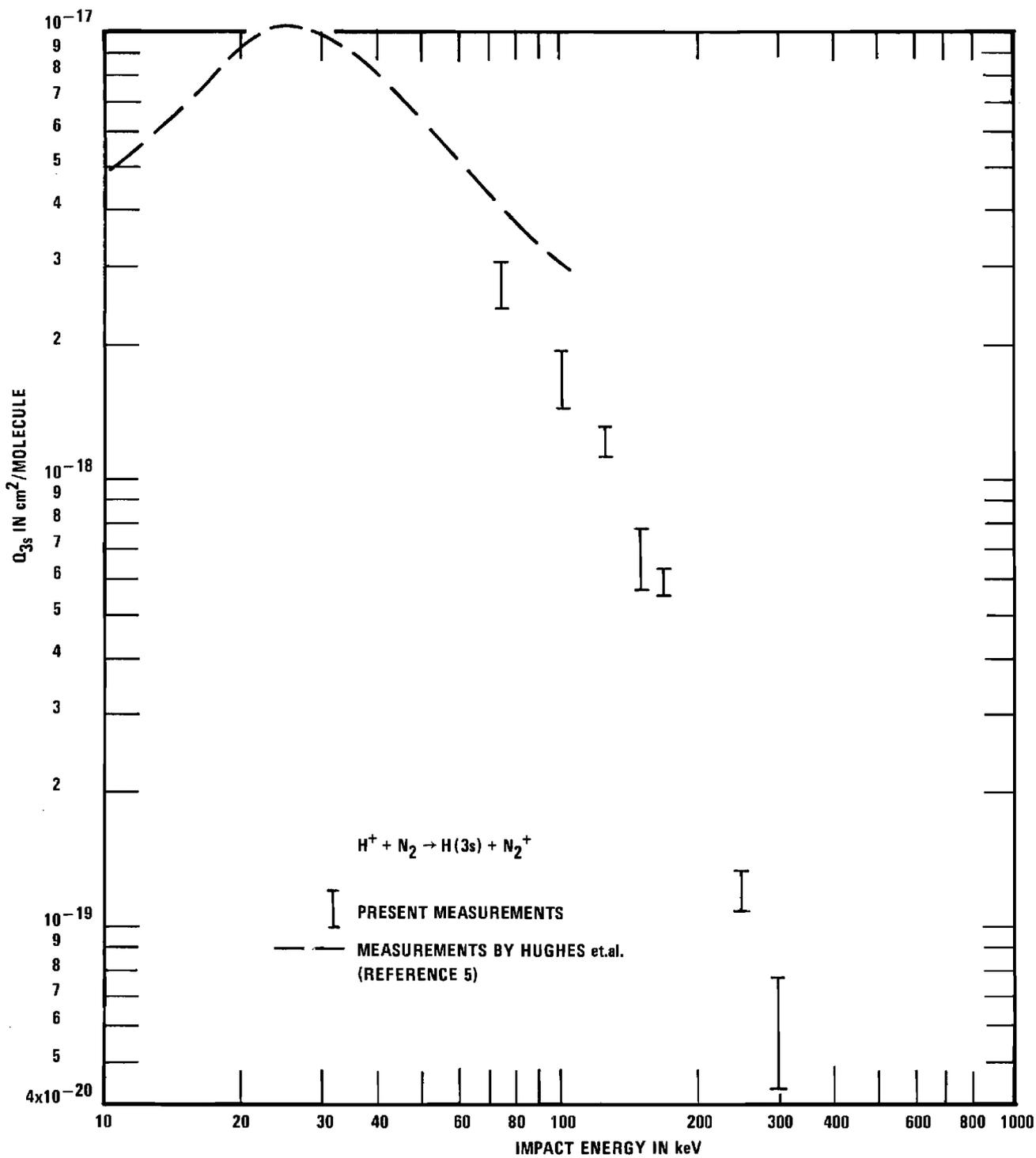


Figure 9. Cross Section Q_{3s} for Charge Transfer into the 3s State of H from a Target of Nitrogen.

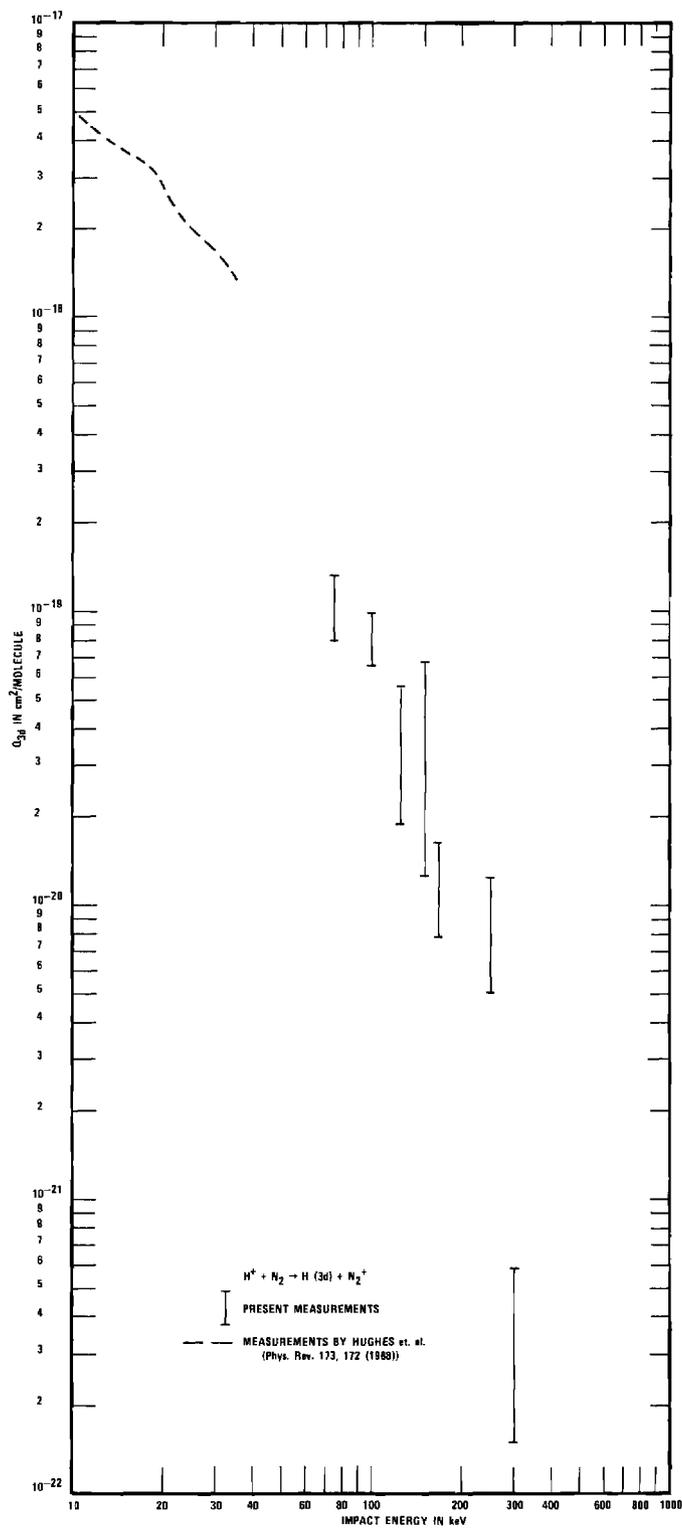


Figure 10. Cross Section Q_{3d} for Charge Transfer into the 3d State of H from a Target of Nitrogen.

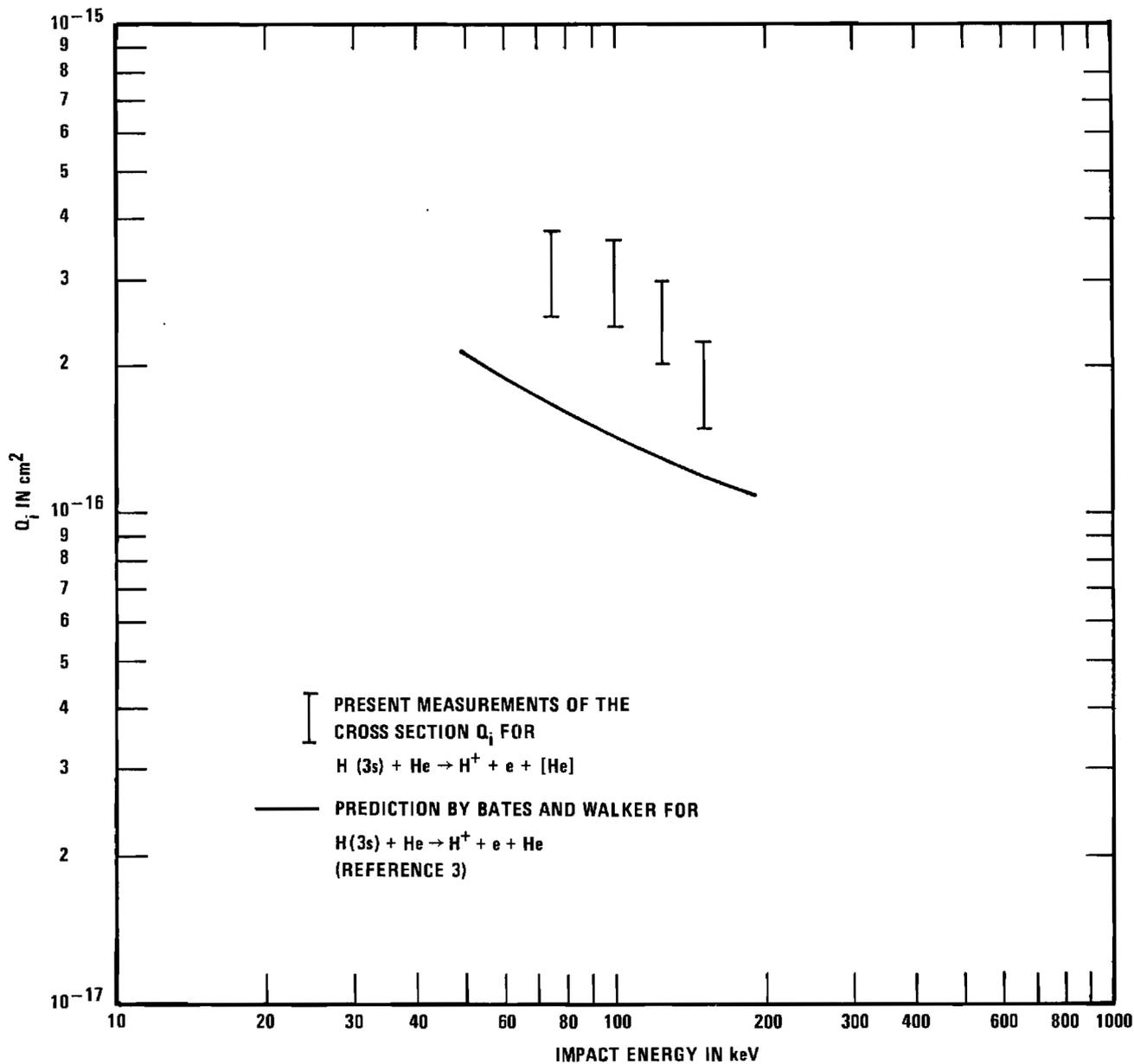


Figure 11. Cross Section Q_1 for Collisional Destruction of H Atoms in the 3s State by Impact on a Target of Helium.

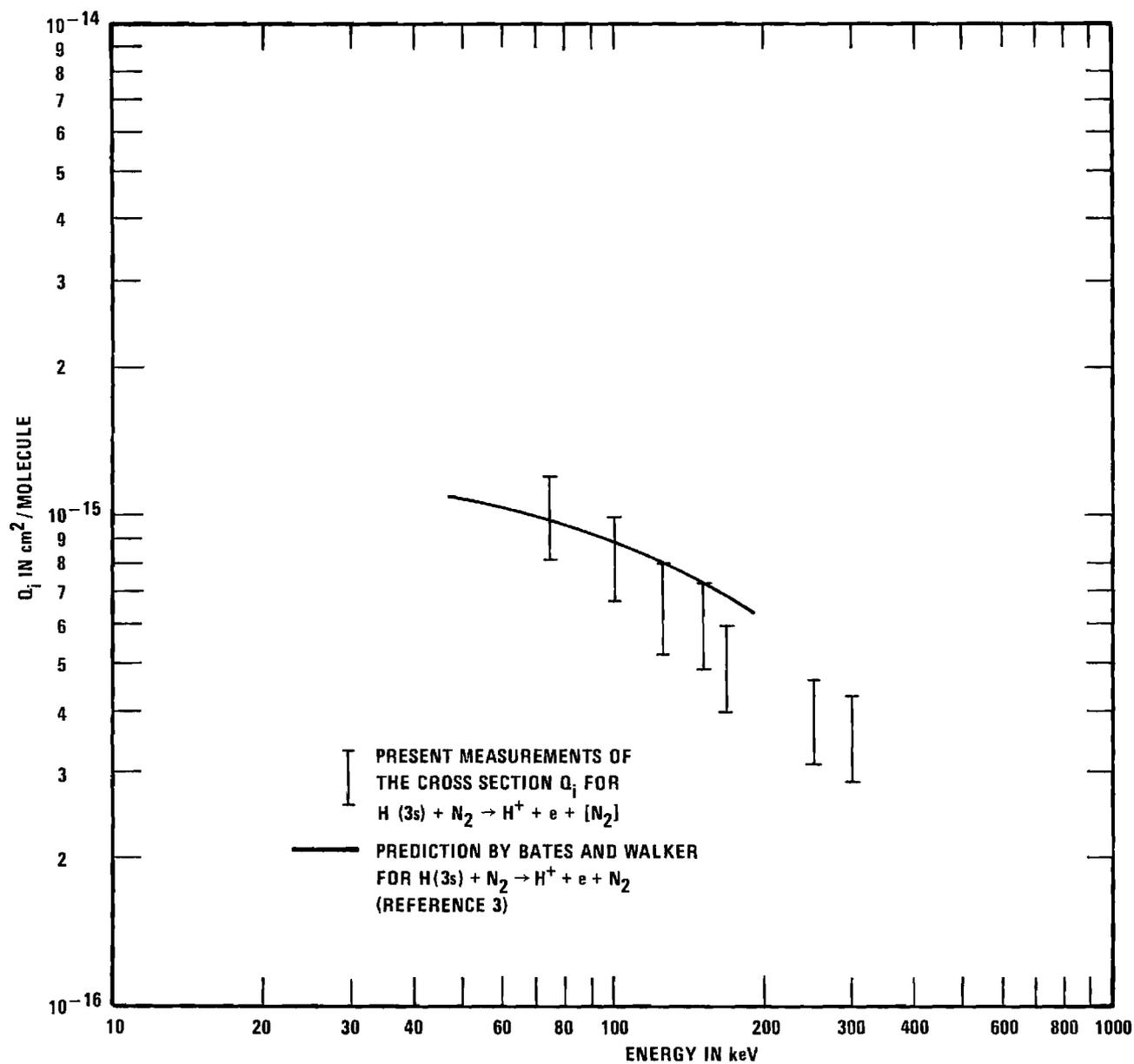


Figure 12. Cross Section Q_1 for Collisional Destruction of H Atoms in the 3s State by Impact on a Target of Nitrogen.

and 3d state populations. The energy is 125 keV.

TABLE I

Relative Populations of Excited States

Target State	He	Ar	N ₂	H ₂
3s	78%	83%	75%	
3p	21%	12%	17%	
3d	1%	5%	8%	
n=3 (total relative to N ₂)	0.323	0.819	1.00	0.258

Clearly there is no very great difference between the relative distributions among the three levels for these three rather different target cases. Butler and May¹¹ have used one electron wave functions to predict that the relative cross sections for the formation of the ns, np and nd states at 125 keV should be 50%, 43% and 7%, respectively. Clearly this prediction seriously overestimates the fraction in the 3p state for all three cases shown above. This is in line with our previous discussion of the detailed measurements for the 3p state where it was noted that although theory and experiment agreed for the 3s and 3d states, theory seriously overestimated the 3p state cross section.

VII. Program for the Remainder of the Contract Year

Measurements of charge transfer into the excited state and collisional stripping of the excited electrons will be completed for targets of He and

N_2 . This work is already well advanced. We require only to improve the statistics of the stripping measurement and to improve the details of our analytical procedures. This will certainly be completed within the available time period. In the remaining time detailed studies will be started on the formation and collisional destruction of excited H atoms as protons are incident on targets of H_2 and Ar. The method to be used for these studies will involve the target cell with observation in the evacuated flight region. It is expected that the analysis of the data will be simpler than for the measurements within the collision region itself, which has been the procedure for most of the studies accomplished thus far. Work on one of these gases will be completed during the present period.

VIII. Program for the Future

It is proposed to continue the study of the formation and collisional stripping of the $3s$, $3p$ and $3d$ states of H formed by proton impact on targets of H_2 and Ar. The gas cell approach will be used for these studies. Particular attention will be paid to the pressure dependence of the emission from the various states in order to search for collisional transfer from one angular momentum state to another. A detailed study will be made of the influence of electric fields in the collision region with three basic objectives: (a) to determine whether stray fields in the collision region can cause appreciable Stark mixing of the $3p$ and $3d$ excited states, (b) to determine whether the presence of a field alters the collision cross sections, (c) to determine the population distribution among the three angular momentum states for high field conditions. Finally, it is intended to carry out a detailed study of processes of collisional dissociation of

H_2^+ and H_3^+ on targets of He, H_2 , Ar, and N_2 which lead to the formation of the 3s, 3p and 3d states of hydrogen in the dissociation fragments.

IX. Publications and Travel

Two reports have been published during the present reporting period:

- (1) "Excitation Induced by 0.15-1.0 MeV Protons in Argon", by E. W. Thomas, *J. Phys. B.* 2, 625, 1969. AEC Report No. ORO-2591-40
- (2) "The Formation and Destruction of Excited Hydrogen Atoms at High Impact Velocities", by E. W. Thomas, J. L. Edwards, J. C. Ford, *Proc. Sixth International Conf. on the Physics of Electronic and Atomic Collisions*, (MIT Press, 1969), page 462. AEC Report No. ORO-2591-39.

In addition to these publications the principal investigator for this program has been associated with two reports concerning work on the ionization and charge transfer studies that are carried out under the same research contract.¹

- (3) "Investigation of the Scattering of He^+ by Noble Gases at High Energies" by G. O. Taylor, B. W. Griffiths, D. E. Troyer, E. W. Thomas and D. W. Martin, *Proc. Sixth International Conf. on the Physics of Electronic and Atomic Collisions*, (MIT Press 1969), page 231.
- (4) "The Detection Efficiency of a Channel Electron Multiplier", by J. N. Fox, R. L. Fitzwilson and E. W. Thomas, *Journal of Scientific Instruments*, (Journal of Physics E). To be published.

Dr. Thomas, Mr. J. L. Edwards and Mr. J. C. Ford attended the "Sixth International Conference on the Physics of Electronic and Atomic Collisions" in Boston, Massachusetts (July 1969). Visits have also been made to the Oak Ridge National Laboratory for the purpose of consulting with research

workers in the Thermonuclear Division.

X. Personnel

The work described in this report was carried out as part of AEC Contract No. AT-(40-1)-2591 and has been under the jurisdiction of Dr. Thomas, Principal Investigator. He has devoted one-quarter time during the academic year to this project and full time during the summer.

Mr. Lee Edwards continues to work full time on this project and draws half time support. The work on charge transfer and collisional stripping reported in this document has been primarily his responsibility. This work is currently being analyzed and will form the basis for a thesis that will be completed during the coming contract period.

Mr. John Ford continues to work full time on this project and draws half time support. He has been responsible for the design, building and testing of the new target gas cell configuration of the experiment. It is expected that the work carried out under this program in the coming contract year will provide the basis for his Ph.D. thesis.

During the course of the present year, assistance has also been received from a number of undergraduates: Mr. Floyd Richie, Mr. David Jacobi,* Mr. Mordechai Shacham, Mr. John Dickey, Miss Tana Sims.*

XI. Accelerator Usage

Use of the Van de Graaff accelerator is shared with a project to measure ionization cross sections, also under AEC contract No. AT-(40-1)-2591.

*Employed under the "Work Study Program" at no cost to this contract.

The accelerator is designed so that it may be rapidly switched from one experiment to another. A period of one day suffices to make the required mechanical connections and to align the ion beam with the experiment.

XII. Incident Report

There have been no incidents for which a report is required during the performance of the research under this contract in the present reporting period.

FORMATION OF EXCITED HYDROGEN ATOMS
BY CHARGE TRANSFER AND DISSOCIATION

ORO-2591-69

PROGRESS REPORT NO. 8

Covering the Period

December 1, 1971 to November 30, 1972

By E. W. Thomas

J. C. Ford

I. Sauers

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Report No. ORO-2591-69

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U. S. ATOMIC ENERGY COMMISSION

OAK RIDGE, TENNESSEE

1972



School of Physics
GEORGIA INSTITUTE OF TECHNOLOGY
Atlanta, Georgia

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TABLE OF CONTENTS

	Page
I. Title	1
II. Introduction	1
III. Abstract	1
IV. Objective	4
V. Organization of the Report	7
VI. High Energy Total Cross Section Measurements	8
VII. Low Energy Differential Cross Section Measurements	17
VIII. Excitation Induced by Particle Impact on Solids	23
IX. Program for the Remainder of the Contract Year	29
X. Program for the Future	31
XI. Publications and Travel	32
XII. Personnel	34
XIII. Incident Report	35

I. Title

Formation of Excited Hydrogen Atoms by Charge Transfer and Dissociation

II. Introduction

This report summarizes work performed on excitation phenomena under contract AT-(40-1)-2591 for the U. S. Atomic Energy Commission. The present report covers the period 1 December 1971 to 30 November 1972 which corresponds to the first 9 months of the 12 month period covered by modification No. 14 to this contract, plus the final three months of the preceeding contract period.

III. Abstract

Studies have been made of excited hydrogen atom formation by collisional neutralization of protons, collisional dissociation of molecular hydrogen ions, and collisional excitation of hydrogen atoms. The bulk of the work involves study of excited state formation as H , H^+ , H_2^+ and H_3^+ traverse various gaseous targets; from these experiments we derive collision cross sections as a function of projectile impact energy and also as a function of the angle through which the projectile is scattered. A new line of studies, initiated very recently, seeks to investigate excited hydrogen atom formation when these same projectiles strike solid metal targets.

The processes of interest may be described by the following reaction equations;

charge transfer neutralization



excitation of H atoms



dissociation of H_2^+



dissociation of H_3^+



The target is represented by X; this is a low density gas in most experiments but a solid metal has also been used for some studies. Our experiments provide no information on the state of excitation, ionization or molecular association of the systems shown in square brackets; our experiments sample only the post collision status of the hydrogen atom.

The total cross sections for charge transfer [Eq. (1)] and dissociation [Eqs. (3) and (4)] have been studied for H^+ , H_2^+ and H_3^+ impact on various target gases at projectile energies ranging from 75 to 1000 KeV. Interest is in the formation of the 3s, 3p and 3d excited states of hydrogen. Principal targets include H_2 , He, Ne, Ar, Kr, Xe, CO, CO_2 , O_2 , CH_4 , C_2H_4 , C_2H_6 and C_3H_8 . The cross sections for charge transfer decrease rapidly with projectile energy while dissociation remains relatively constant. We have shown that the charge transfer cross sections for noble gas and certain molecular gas targets may be scaled together by a semi-classical theory of Gryzinski. The "universal curve" so generated may permit the prediction of unknown cross sections for cases that have not been experimentally studied. In dissociation [Eqs. (3) and (4)] the cross sections for formation of 3s, 3p and 3d levels are approxi-

mately equal at any given energy. Although systematic variations of cross section with target have been observed we have been unable to formulate a mathematical prediction of the behavior.

In a lower energy experiment we are studying the angular distribution of metastable hydrogen atoms created in the direct excitation of neutral H atoms [Eq. (2)] and in dissociation [Eqs. (3) and (4)]; targets used include H_2 , He and Ar. We have also studied the distributions of H^+ and H^0 formed in these same collisions. The angular distributions in dissociation are caused primarily by the potential energy released in fragmentation. All distributions in H_2^+ dissociation indicate that the cross section is greatest when the molecular internuclear axis is parallel to the direction of H_2^+ motion; in one case the predictions of theory are in agreement with experiment. Since there is little similarity between dissociation on different targets, it seems likely that the mechanism for dissociation is related to an atomic complex formed by the projectile and target. Direct excitation of H projectiles is now under study. It appears that the angular distribution of the scattered projectiles is predictable by a classical scattering mechanism using screened coulomb interactions between the projectile and target. Relative angular distributions of H^0 and $H(2s)$ are essentially the same.

Some studies have also been made of how projectiles reflect from a metal target. The experimental technique involves study of light emission induced by the projectile impact on a surface (Cu, Ni or Au); spectral lines corresponding to neutralized projectiles are observed. These lines are found to be Doppler broadened and from the line shape we can deduce the velocities and angular distributions of the scattered particles. For impact of H^+ and He^+ ions at energies from 3 to 10 KeV the scattered excited projectiles appear

to be elastically scattered from the first few surface monolayers; the scattering cross section is approximately that given by a Rutherford formula. At energies of 100 to 1000 KeV the width is less than that predicted for elastic scattering, showing that the projectiles suffer a considerable energy loss. This may indicate that high energy projectiles are scattered from some rather deep layer in the target. In none of these observations do we have a definite indication of thermal energy H atoms being ejected from the surface. In addition to the line emissions from scattered projectiles we also observe very intense broad band emissions that one cannot relate to the atomic structure of ejected target atoms or to reflected projectiles. The observed emission is neither bremsstrahlung nor transition radiation; the origin of the band emission requires some further study.

IV. Objective

The overall objective of this program is to come to an understanding of the mechanisms by which excited hydrogen atoms are formed in processes of charge transfer and dissociation. Much of the work involves simple gaseous targets; for such cases detailed theoretical predications should be tractable. For complicated gaseous targets there are generally no detailed theoretical predictions available; in such cases we are interested in finding scaling relations that will permit the estimation of unknown cross sections from available experimental data. In the case of solid metal targets there is virtually no existing experimental or theoretical data available. We are here probing the basic understanding of the process in order to provide some guidelines within which a theoretical understanding might be attempted.

Our research effort is directed primarily towards formation of excited atoms in the $n = 2$ and $n = 3$ states; such atoms may be detected fairly readily by the photons emitted as they decay. We believe that there are good reasons for concentrating on these states rather than studying total fluxes of neutral atoms produced in some collision process. A neutral atom flux will contain particles in all states of excitation; it may sometimes be the case that a very large fraction of the atoms are excited. Thus the measurement of a neutral flux is ambiguous in that the state of the atom is undefined. Consequently we believe that the study of excited states is the best way of carrying out a fundamental study. In general, once a fundamental understanding has been achieved of how a specific state is formed then one can readily extend the theory to the prediction of other reaction channels in the same mechanism; at that stage one can assess the behavior in situations of practical importance.

The principal areas studied with gaseous targets may be considered in two parts. First there are measurements of total cross sections for the formation of the $3s$, $3p$ and $3d$ states of hydrogen measured at high energies (75 - 1000 KeV) and secondly, differential (in-angle) cross sections for formation of metastable ($2s$) hydrogen at low energies (5 - 30 KeV). The total cross sections are designed to test detailed theoretical predictions in the Born approximation at high energies; also we hope to develop scaling laws to relate cross sections for reactions with different atomic and molecular targets. The low energy studies are to cover an energy region where Born approximations are incorrect and where sophisticated coupled state calculations should be necessary. It is always desirable to test theory by comparison with differential (in-angle) cross section measurements; however for practical reasons differential cross sections can be measured only in low velocity regions.

Our research with solid metal targets involves an area where there is little prior information. The motivations are principally to provide a collection of empirical data that might provide a basis for understanding reflection of heavy particles at surfaces. In many respects the solid target might be considered as a "thick" form of a gas target with some modifications to the energy level scheme of the outermost electrons; it might be expected that the fundamental understanding of collision processes in gaseous targets should be applicable to the multiple collision events that one will find in a solid target.

The specific objectives for the period covered by the report were the following.

- (i) To complete the studies of high impact energy (100 - 1000 KeV) charge transfer and dissociation events that lead to formation of H(3s), H(3p) and H(3d) excited states. This program has been completed and prepared for final publication; no further work on these reactions is planned.
- (ii) To study at low impact energies (5 - 30 KeV) the angular distribution of H(2s), H⁰ and H⁺ formed when H₂⁺, H₃⁺ and H projectiles strike gaseous targets. The study of the dissociation events is completed. Studies of H excitation required some modifications to the apparatus; those modifications are complete and the experimental investigations are in progress.
- (iii) To make preliminary studies of the light emission induced by projectile impact on various solid targets. This program has the nature of a feasibility study to determine whether one can

get useful information on scattering of particles from surfaces by this method. The work so far completed indicates that the angular distribution and velocities of scattered projectiles can be usefully studied in this manner. It is proposed that this program should be further extended. Preliminary data from this part of the program are discussed in the report.

We will not repeat here the various justifications for this program in terms of the AEC's effort in thermonuclear research; those considerations are discussed in our proposals. It will suffice to note that the areas of neutral beam injection into plasmas, plasma diagnostics and particle loss mechanisms all involve charge transfer and dissociation mechanisms; recycling of hydrogen emanating from the containment vessel walls is important in that it may severely limit attainable plasma temperatures. We suggest that the understanding of these mechanisms is important to the AEC's overall thermonuclear research program.

V. Organization of the Report

The report is divided into three parts. First, we discuss high energy measurements of $3s$, $3p$ and $3d$ excited state formation. Second, there is a report on the low energy studies of how metastable hydrogen formation varies with the scattering angle. Third, there is a report on our preliminary studies of excited state formation induced by particle impact on surfaces.

Most of the cross sections measurements have been submitted for publication or are now being prepared for publication. For economy and efficiency we will not repeat that material here but will refer the reader to the impending publications. This present report will concentrate on a brief statement of the experimental method followed by a summary of the more important conclusions.

VI. High Energy Total Cross Sections Measurements

(a) Introduction

These experiments study formation of hydrogen in the 3s, 3p and 3d states induced by H^+ , H_2^+ and H_3^+ impact on various targets; projectile energies range from 75 to 1000 KeV. The complete details of the experimental technique have already been published.¹

The experimental method is to fire the projectile beam through a cell containing the target gas and then into an evacuated flight tube; observations are made of light emitted as the beam traverses the flight tube. If light is emitted by a single decay mechanism, then the intensity of the emission will decrease exponentially with distance from the gas cell exit. The intensity $I(x)$ as a function of distance, x , from the target cell exit is given by:

$$I(x) = I(0) e^{-x/v\tau} \quad (5)$$

The velocity of the excited atom is v , the lifetime of the excited state τ ; $I(0)$ is the intensity at the exit from the cell and may be related to the cross section for the formation of the excited state. Measurement of $I(x)$ at various x permits determination of $I(0)$ and hence of the cross section of interest. Now, in the present experiments we are studying the formation of the 3s, 3p and 3d states by measurement of the hydrogen Balmer alpha spectral line intensity. This line contains contributions from three transitions, $3s \rightarrow 2p$, $3p \rightarrow 2s$ and $3d \rightarrow 2p$. Thus the measured intensity is represented by a sum of three equations like equation 5; each equation involving different

1) J. C. Ford and E. W. Thomas, Phys. Rev. A 5, 1694 (1972). Assigned AEC Report No. ORO-2591-58.

values of $I(0)$ and τ . The procedure is to measure $I(x)$ and de-convolute this into three exponential decays having the known characteristic lifetimes of the $3s$, $3p$ and $3d$ states; the three characteristic values of $I(0)$ so derived are then used to find the cross section for the formation of the excited states. It is to be noted that the three transitions that contribute to the Balmer alpha line (namely $3s \rightarrow 2p$, $3p \rightarrow 2s$, and $3d \rightarrow 2p$) all exhibit essentially the same wavelength and cannot be resolved spectroscopically in a simple manner. The technique described here utilized the characteristic lifetimes of the three states to provide a separation of contributions that cannot be carried out by spectroscopic methods.

The charge transfer measurements have been written up in a total of three papers. In the first¹ we discussed the experimental procedure and the results for charge transfer in helium and argon. In the second paper² we consider charge transfer in molecular targets including H_2 , N_2 , NO , O_2 , CO , CO_2 , CH_4 , C_2H_4 , C_2H_6 and C_3H_8 . A third and final paper³ includes data for charge transfer in Ne , Ar , Kr and Xe . Data on the dissociation mechanisms are to be found in three papers^{3,4,5}; it includes data on H_2^+ and H_3^+ dissociation

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- 2) J. C. Ford, and E. W. Thomas, Phys. Rev. A, 5, 1701 (1972). Assigned AEC Report No. ORO-2591-59.
 - 3) R. J. Conrads, T. W. Nichols, J. C. Ford, and E. W. Thomas, Phys. Rev. (submitted for publication October 1972). Assigned AEC Report No. ORO-2591-67.
 - 4) J. C. Ford, F. M. McCoy, R. Conrads, and E. W. Thomas, Phys. Rev. A, 5, 1705 (1972). Assigned AEC Report No. ORO-2591-60.
 - 5) R. J. Conrads, J. C. Ford, and E. W. Thomas, J. Chem. Phys. (submitted for publication October 1972). Assigned AEC Report No. ORO-2591-68.

in targets of He^4 , H_2^3 , D_2^3 , N_2^3 , Ne^3 , Ar^3 , Kr^3 , Xe^3 , O_2^5 , CO^5 , CO_2^5 , CH_4^5 , C_2H_6^5 and C_3H_8^5 . This series of papers provides a very complete survey of the charge transfer and dissociation mechanisms. For complete details the reader is referred to the relevant articles; further remarks in this report will be confined to a general survey of the various unifying features to be found in the data.

(b) Behavior of Charge Transfer

All the charge transfer cross sections decrease very rapidly with projectile kinetic energy. Values for $\text{H}(3s)$ formation range from the order of $5 \times 10^{-18} \text{ cm}^2$ at 75 KeV to the region of 10^{-21} cm^2 at 1000 KeV. Cross sections for $\text{H}(3p)$ formation exceed those for $\text{H}(3d)$ production and are less than those for the $\text{H}(3s)$ level. It is quite clear that the cross section decreases with the angular momentum of the final state. It has sometimes been assumed, by others, that the cross section should vary with the statistical weight of the final state and should therefore increase with angular momentum; clearly our data show this assumption to be wrong.

For a helium target there are theoretical predictions available based on the Born approximation.⁶ There is a good general agreement between theory and experiment but some specific differences exist for the $3p$ cross section which cannot be explained by experimental error;¹ undoubtedly the theory needs improvement. There are a couple of general theoretical formulations of the charge transfer problem^{7,8} that have often been used for design purpose in neutral beam injectors; one formulation treats all types of targets as single

6) R. A. Mapleton, Phys. Rev. 122, 525 (1961).

7) J. R. Hiskes, Phys. Rev. 137 A 361 (1965); Phys. Letters 17, 263 (1965); Phys. Rev. 180, 146 (1969).

8) S. T. Butler and R. M. May, Phys. Rev. 137, A 10 (1965).

electron atoms (i.e. as H)⁷ while the other treats all targets as two electron atoms (i.e. as He)⁸. These theories are in general agreement with experiment for targets as H and He; this is to be expected. However they show only qualitative agreement when applied to the more complex targets. It is concluded that when the one and two electron formulations are applied to complex atoms they should be expected to give order of magnitude estimates and then only at high energies (say greater than 300 KeV).

In our most recent article³ we have attempted to find some unifying relationship to describe the charge transfer process. We have had some moderate success with a formulation by Garcia et al. that is in turn based on a classical prescription by Gryzinski.¹⁰ The relationship is as follows:

$$\sigma \frac{E \lambda^3}{U_B} = \frac{2\pi e^4}{3} \left(\frac{\lambda E}{U_A} \right)^2 \left[\frac{7 + 3(\lambda E/U_A)}{(\lambda E/U_A + 1)^2} \right] \quad (6)$$

Here σ is the cross section for charge transfer when a singly ionized projectile of energy E captures an electron; the binding energy of the electron in the target atom before the collision is U_A , the binding energy of the electron in the projectile atom after the collision is U_B . The factor λ is the ratio of electron mass to projectile mass. The cross section is per electron in the outer shell of the target. This relationship suggests that $\sigma(E\lambda)^3/U_B$ is a function only of $E\lambda/U_A$. Thus cross sections σ measured as a function of impact energy E , for a variety of different targets may be scaled together.

9) J. D. Garcia, E. Gerjuoy and J. E. Welkner, Phys. Rev. 165, 72 (1968).

10) M. J. Gryzinski, Phys. Rev. 138, A326 (1965).

Garcia et al.⁹ test the effectiveness of Eq. (6) and find that it does indeed provide a scaling of various cross sections for H^+ neutralization in rare gases and in potassium vapor. It was suggested⁹ that this scaling procedure should permit the prediction of unknown cross sections.

We can attempt to apply this same scaling procedure to the measured cross sections for excited hydrogen formation by charge transfer. Mapleton¹¹ points out that the cross section σ to be used in Eq. (6) is that for a capture process with a specific change of the electron energy and therefore includes capture into all angular momentum substates of a given principal quantum number. Thus we will use the sum of cross sections for the formation of the $3s$, $3p$ and $3d$ levels; let us call this $\sigma(n = 3)$. Figure 1 shows cross sections scaled according to Eq. 6. One axis is the quantity $\sigma(n = 3) (E\lambda)^3/U_B$; E is the projectile impact energy (in eV), U_B the binding energy of the captured electron in the $n = 3$ state of hydrogen (1.511 eV), and λ is $1/1836$. The horizontal axis is $E\lambda/U_A$. Here U_A is the binding energy of the electron before capture; this is to say U_A is the ionization potential of the target. It is not clear that this scaling procedure should be applicable to molecular targets but nevertheless we do include data for H_2 , O_2 and N_2 . In the molecular cases the cross section for the atom is assumed to be half the measured cross section for the diatomic molecule. For the molecules, H_2 , O_2 and N_2 , we have taken U_A to be the ionization potentials of atomic H, O and N; there is some justification for using instead the molecular ionization potentials but this does not in fact appreciably change the plotted position of the data points. There is one further factor that must be included. The cross sections pre-

11) R. A. Mapleton. "Theory of Charge Transfer" (Wiley-Interscience, New York 1972) page 194.

dicted by Eq. 6 are per electron in the outer shell of the target. Thus the cross sections for Ne, Ar, Kr and Xe are for 6 electrons in the outer shell. The measured cross sections for He and the estimated cross sections for H, O and N have been multiplied by factors of 3, 6, 1.5 and 2 respectively to give the cross sections for six equivalent electrons in each case. The data plotted according to this prescription in Fig. 1 have been obtained from our own work and from that of Hughes et al.¹². In view of crudeness of the approximations made in the derivation of Eq. 6 it is gratifying that the data points shown in Fig. 1 agree as well as they do. The scatter is certainly less than that exhibited when Garcia et al.⁹ applied this scaling procedure to total cross sections. Following Garcia et al.⁹ we would suggest that equation (6) may be used to scale measured cross sections in order to predict unknown cross sections for excited state formation; the accuracy would appear to be within a factor of 3 or better.

Strictly speaking a cross section determination for a final state of any principal quantum number n should fit this curve by suitable choice of U_B in equation (6). It has however been pointed out¹¹ that this will cause the cross section to vary as n^{-2} while experiment shows definitely¹² that it varies as n^{-3} . On a purely empirical basis one can state that the curve generated by Eq. 6 will represent formation of any excited state provided we multiply cross section by n , the principal quantum number of that state.

In summary we may state this conclusion. The cross section $\sigma(n)$ obeys the relationship

$$\frac{\sigma(n)}{N} \frac{(E\lambda)^3}{U_B} n = f\left(\frac{\lambda E}{U_A}\right) \quad (7)$$

12) R. H. Hughes, C. A. Stigers, B. M. Doughty and E. D. Stokes, Phys. Rev. A, 1, 1424 (1970).

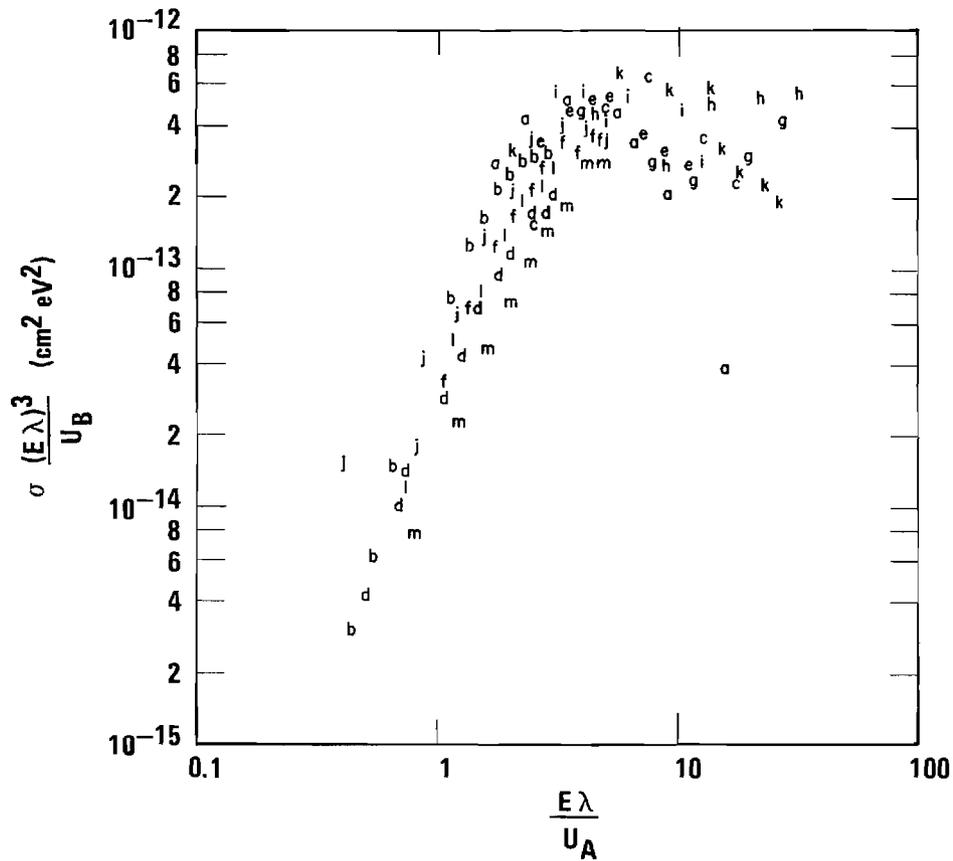


Figure 1. Scaling of cross section determinations using the prediction of Gryzinski; $\sigma(n = 3) (E\lambda)^3/U_B$ shown as a function of $E\lambda/U_A$. Cross sections are for the reaction $H^+ + X \rightarrow H^+ (n = 3) + X^+$. The sources of data points are as follows; (a), (e), Ford et al.¹; (b), (k), Ford et al.²; (c), (g), (h), Conrads et al.³; (b), (d), (f), (j), (l), (m), Hughes et al.¹². Targets include He [(a) and (b)]; Ne [(c) and (d)]; Ar [(e) and (f)]; Kr [(g)]; Xe [(h)], H [(i), (j)], N [(k), (l)] and O [(n)].

Here N is the number of valence electrons in the atom (the rest of the symbols are as previously defined). A cross section measured for some target, or for a group of targets, may be used to establish the form of the function $f\left(\frac{\lambda E}{U_A}\right)$ of eq. (7). The cross section for some other state n or some other atomic target may then be estimated with a fair degree of reliability. This scaling procedure has been established by our data only for values of $E\lambda/U_A$ ranging between 0.15 and 15; it does not necessarily apply for conditions outside this range.

(c) Behavior of Dissociation

A consistent feature of all our dissociation studies is that, in a given collision situation, the cross sections for $H(3s)$, $H(3p)$ and $H(3d)$ formation are almost equal. Cross sections all decrease slowly with increasing impact energy; a decrease by a factor of two between 100 and 700 KeV is typical. In figure 2 we show some representative data for $H(3s)$ formation induced by H_2^+ impact on various noble gas targets; for a full presentation of data the reader is referred to our publications.^{3,4,5} Figure 2 shows that cross sections generally increase with the complexity of the target atom; there also a definite indication of structure in the cross section curves. By comparing our measurements with previous data on H^0 atom formation by dissociation we find that the proportion of H atoms formed in the $3s$, $3p$ and $3d$ states may be as high as 15% under some circumstances.³ This is to be compared with excited state components of only 3% in formation of H^0 by charge transfer.¹ Thus we have the very definite conclusion that the highest excited state content of H beams is achieved when the hydrogen atoms are formed by dissociation of molecular ions rather than by neutralization of protons. The fractional excited state content does not vary greatly with

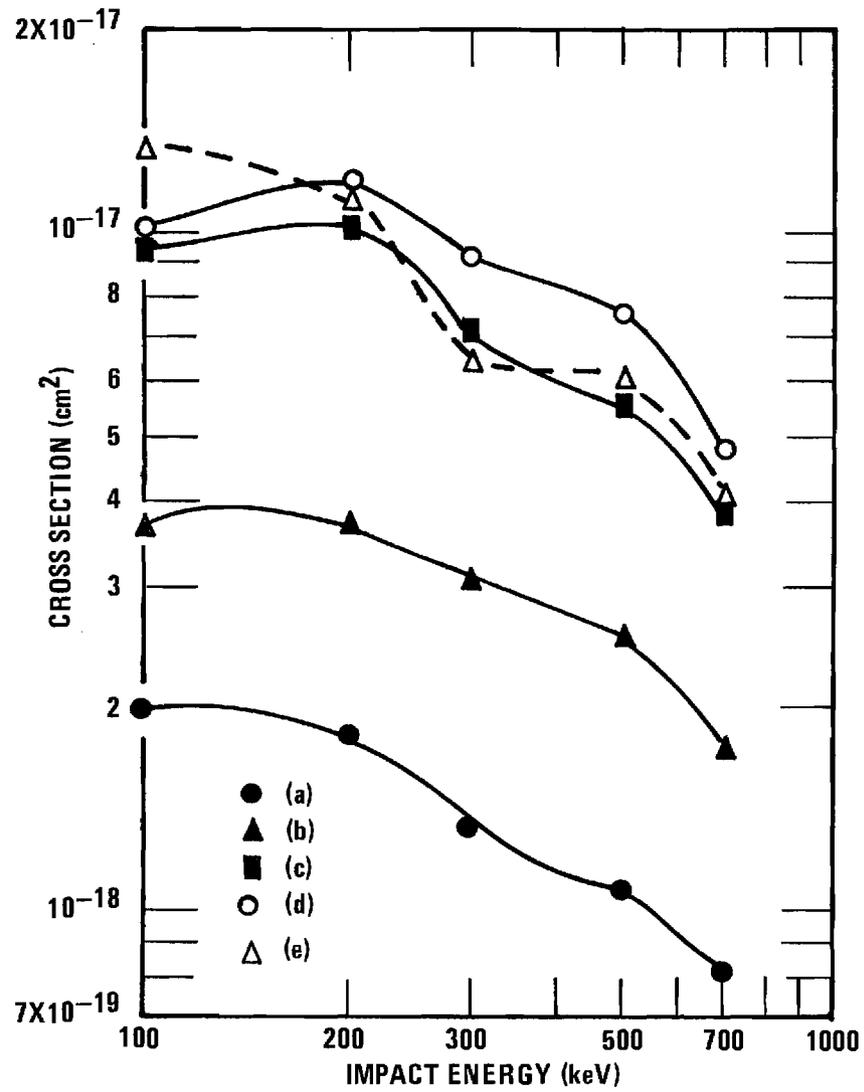


Figure 2. Cross sections for the formation of H(3s) by collisional dissociation of H_2^+ in various noble gas targets. (a) He target; (b) Ne target; (c) Ar target; (d) Kr target; (e) Xe target.

the nature of the target but the net flux of excited particles does generally increase with the complexity of the target. Thus, in very general terms, if one wishes to prepared a high flux of excited H atoms one should do this by dissociation of H_2^+ or H_3^+ in a high molecular weight target gas. (e.g. Ar, Kr, Xe or one of the hydrocarbons).

We have attempted to find some unifying feature in this data that would allow us to relate cross sections for different targets; some formulation like Eqs. 6 and 7 for the charge transfer problem would be very useful. No such feature has been found.³

(d) Future Plans

This work is now considered finished and all the data has been published or submitted for publication. We believe that our work represents a rather complete survey of the excitation mechanisms and no further experimental work is called for.

VII. Low Energy Differential Cross Section Measurements

(a) Introduction

Here we study the angular distribution of metastable (2s) hydrogen atoms formed as H^+ , H_2^+ and H_3^+ ions and H atoms traverse various targets. Again the intention is to provide information that will assist with the fundamental understanding of such processes. It can be shown that the distance of closest approach during a collision is (at least approximately) a linear function of the product between projectile energy, E, and scattering angle, θ . Thus, keeping E fixed and varying θ one can observe how the collision mechanism varies with the colliding atoms. The prediction of cross section behavior is in terms of the potential energy curves and the minimum separation distance between the colliding partners. It follows that

studies of angular scattering permit the most direct comparison between theory and experiment. At the energies of this experiment the simple one-state theories, like the Born approximation, are not satisfactory. The so-called coupled state calculations are necessary at energies below the limit of the Born approximation's applicability; this experiment is designed specifically to assist with the formulation of coupled state calculations.

The ion beam for this experiment is provided by a 5 - 30 KeV accelerator with an RF ion source. The beam is collimated to an angular width of $\pm 20^\circ$ and then directed into a target gas cell.

A slit system selects a small part of the scattered particle flux and permits it to enter a detection region. Facilities are provided for detection of ions, neutrals and metastable atoms. By rotating the slit system about the center of the target cell one may change the scattering angle of the detected particles. From the measurement of scattered particle flux as a function of angle one may determine the cross sections for the scattering of charged, neutral, and metastable atoms.

The ions are detected quite simply as a current by a Faraday cup arrangement. The neutrals are detected by a secondary emission detector. Metastables are monitored through the emission of Lyman alpha photons when an electric field is applied to the flux of scattered particles; a field mixes the 2s with the 2p state causing a Lyman alpha photon to be emitted through the $2p \rightarrow 1s$ decay.

We have written a rather complete description of the basic apparatus in a configuration for measurement of neutral and charged particle fluxes.¹³⁾

13) R. L. Fitzwilson and E. W. Thomas, Rev. Sci. Inst. 42, 1864 (1971).
Assigned AEC Report No. ORO-2591-57.

There is also a separate publication wherein the metastable hydrogen detector is described in detail.¹⁴

(b) Data for Charge Transfer

We have already published data for H^+ traversing an the target¹⁵ and shown that a coupled state theoretical calculation provides a good prediction of the cross section behavior. Also the total flux of scattered particles, ions and atoms, scattered at a particular angle can be adequately predicted by a Rutherford type cross section with inclusion of screening of the target nucleus by its electron.

(c) Dissociation of H_2^+

Considerable work has been done on the dissociation of H_2^+ by impact on targets of He, Ar, H_2 and N_2 ¹⁶; data includes the differential cross sections for formation of H^+ , H^0 and $H(2s)$.

The data indicates¹⁶ that the dissociation mechanism involves excitation of the H_2^+ to some repulsive state followed by separation of the fragments. Following the Franck-London principal one would expect that the angle φ between the H_2^+ internuclear axis and the direction of H_2^+ motion will not change during the collision. From the known potential energy curves of H_2^+ one can deduce the potential energy, Q , released as the ion fragments; this energy imparts kinetic energy to the fragments. One may show¹⁶ that the angle θ at which a fragment is observed in the laboratory frame may be related to Q and φ by

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- 14) R. L. Fitzwilson and E. W. Thomas, Phys. Rev. A 3, 1305 (1971).
15) R. L. Fitzwilson and E. W. Thomas, Phys. Rev. A 6, 1054 (1972).
Assigned AEC Report No. ORO-2591-62.
16) I. Sauers, R. L. Fitzwilson, J. C. Ford and E. W. Thomas, Phys. Rev. A, 6, 1418 (1972). Assigned AEC Report No. ORO-2591-63.

$$\theta = \left(\frac{Q}{E}\right)^{\frac{1}{2}} \text{Sine } \varphi \quad (8)$$

where E is the energy of the H_2^+ ion before collision. Using this relationship one can take the cross section measured in the laboratory frame as a function of θ and transform it to the center of mass frame (of the H_2^+) as a function of φ . The result of this transformation may then be examined to determine how the cross section for dissociation varies as a function of the angle φ between the internuclear axis and the initial direction of H_2^+ motion. In all cases studied we find that the cross section peaks for φ close to zero (i.e. internuclear axis parallel to direction of motion) giving rise to a strongly peaked forward scattering in the laboratory frame of reference. In the case of a helium target the angular distribution of $H(2s)$ is given quite accurately by a cross section that varies as $\cos^2 \varphi$; this confirms a detailed theoretical prediction by Green and Peek.¹⁷ Our work represents the first experimental data on the angular dependence of cross section for formation of a specific final state. Previous work involved only measurement of the total neutral flux; consequently there is ambiguity as to what repulsive state (or states) were concerned in the dissociation mechanism.

The dissociation cross sections for Ar and N_2 targets are even more strongly peaked than the data for helium; contrary to the predictions by Green and Peek¹⁷ the relative angular distribution is not the same as that for helium. This suggests that the nature of the target strongly influences the excitation mechanism; perhaps an explanation of the observations will require consideration of a temporary complex molecule involving both the projectile and the target.

17) T. A. Green and J. M. Peek, Phys. Rev. 183, 166 (1969).

(c) Excitation of Neutral Atoms

We have recently started work on the angular distribution of H(2s) formed by direct excitation of ground state H; targets used for this work so far are He and Ar. The excitation of H is a very important problem because of the contribution that it can make to the development of theory. A difficult theoretical problem in ion-atom collisions is the consideration of the long range Coulomb force due to the charged particle. For collisions between neutral particles this long range force component is of course absent and the theoretical problem is much simplified.

To carry out this study we have modified the apparatus by introduction of a gas cell to neutralize H^+ ions and so form our H^0 projectile beam. Following the neutralizer is an electric field which removes any residual H^+ component and also Stark quenches H(2s) metastables; the distance between the neutralizer and target cell is sufficient to permit most excited states of H to decay before impact on the target.

Data from this experiment are still preliminary and further analysis is necessary. In Fig. 3 we show a sample of the data in the form of cross sections for scattering of H^0 , H(2s) and H^+ as a function of scattering angle for a helium target. The data are relative values only and we are still in the process of establishing absolute cross sections. It is observed that the cross sections all show the same relative variation with angle. This is perhaps surprising since the H^0 flux is principally due to elastic scattering, while the H(2s) and H^+ fluxes involve an inelastic event. One of our faculty, Prof. M. R. Flannery, has previously carried out calculations of total cross

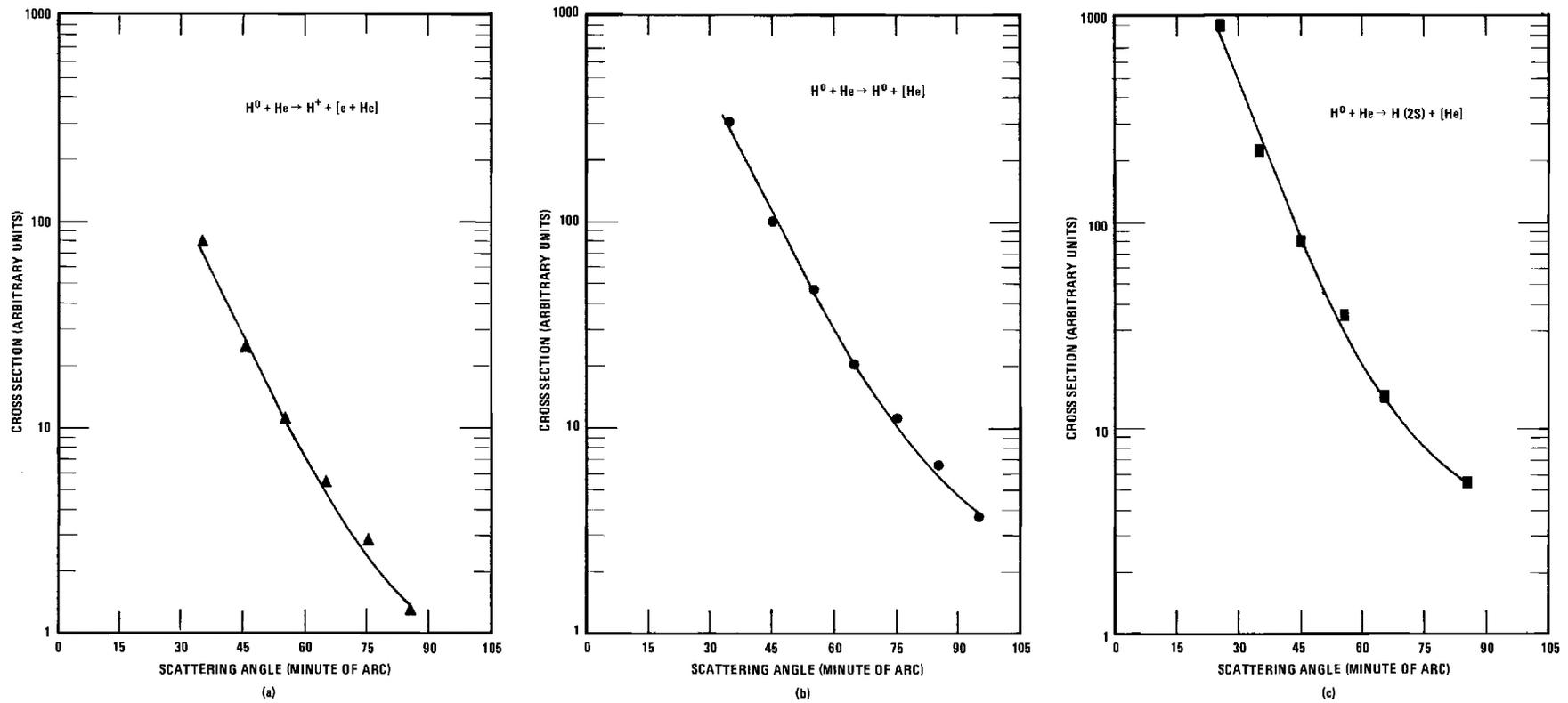


Figure 3. Differential cross sections for production of H^+ , H^0 and $H(2s)$ when 15 keV H^0 atoms are incident on a helium target.

sections for this problem¹⁸; he is now modifying his calculations to permit predictions of differential cross sections.

(d) Future Plans

It is intended that this study will be continued to provide absolute values of the various cross sections involved. We intend also to further modify the apparatus to permit measurement of H^- cross sections for the various collision combinations discussed above.

VIII. Excitation Induced by Particle Impact on Solids

We have embarked upon a study of particle scattering from surfaces. Again we are concentrating on excited state formation; the experimental technique involves a study of the light emission induced by projectile beam impact on a surface. It is expected that a study of emission from the projectiles will give information on how they are scattered by the surface.

We will describe here the results of two experimental studies. The first, and most complete, is work carried out by Dr. E. W. Thomas (Project Director) while on leave of absence at the FOM Institute for Atomic and Molecular Physics in Amsterdam (The Netherlands). This work received no support from the AEC contract except that some time was devoted to analysis of data after Dr. Thomas returned to the U.S.A. and was again being supported financially by the AEC contract. The work done in Amsterdam was, however, designed to be the first stage of our research program in this area by establishing the feasibility of this type of study. We therefore include a discussion of the results in this report. The second part of the research is the work done at Ga. Tech on this same phenomenon; that work has only

18) M. R. Flannery, J. Phys. B 2, 913 (1969) and J. Phys. B. 2, 1044 (1966).

recently been started. The apparatus and scientific objectives of both studies are in fact similar in all essential respects. Much of the work done in Amsterdam has been submitted for publication^{19,20}; the data obtained at Ga. Tech are considered to be preliminary and not yet of publishable quality.

Fig. 4 shows diagrammatically the apparatus used in both studies. The projectile beam is prepared in the usual manner and directed onto a solid metal target. A scanning monochromator is placed with its axis at 90° to the ion beam path and views the emission at the point of projectile impact on the surface. The angle between projectile direction and the surface normal, ϕ , may be varied from 0 to 90° . Behind the target is a Faraday cup; to monitor the projectile ion beam current directly, the target is withdrawn from the beam line and the projectile flux is monitored on the Faraday cup in a conventional manner. It has been shown that the measured current on the target itself is not an accurate indication of incident projectile flux; copious quantities of secondary ions and electrons are ejected from the target and cannot be completely suppressed.

The apparatus used in Amsterdam had a background pressure of 10^{-9} mm. The targets of Cu, Ni and Ar were single crystals of known orientation; prior to the commencement of measurements the targets were cleaned by electron bombardment heating and by sputtering with an Ar^+ beam. The targets used in the Amsterdam work were believed to be atomically clean. In contrast

19) C. Kerkdijk and E. W. Thomas, Physica (to be published). Assigned AEC Report No. ORO-2591-65.

20) C. Kerkdijk and E. W. Thomas, Surface Science (to be published). Assigned AEC Report No. ORO-2591-66.

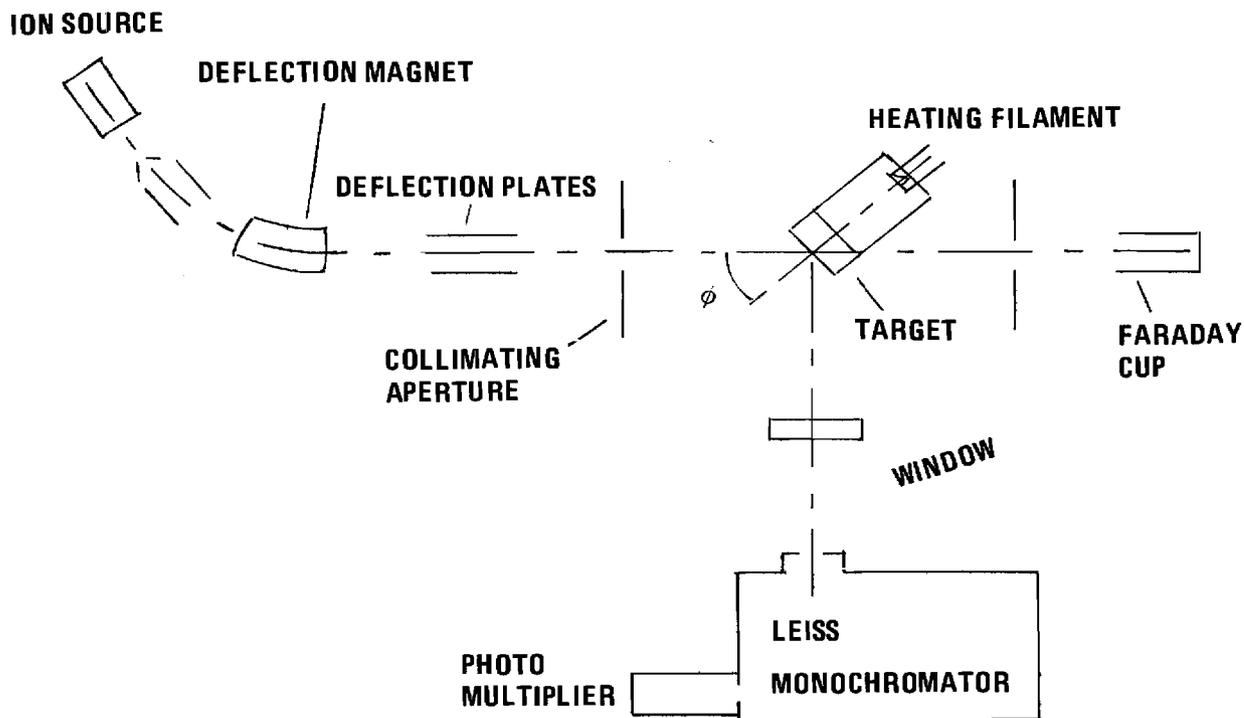
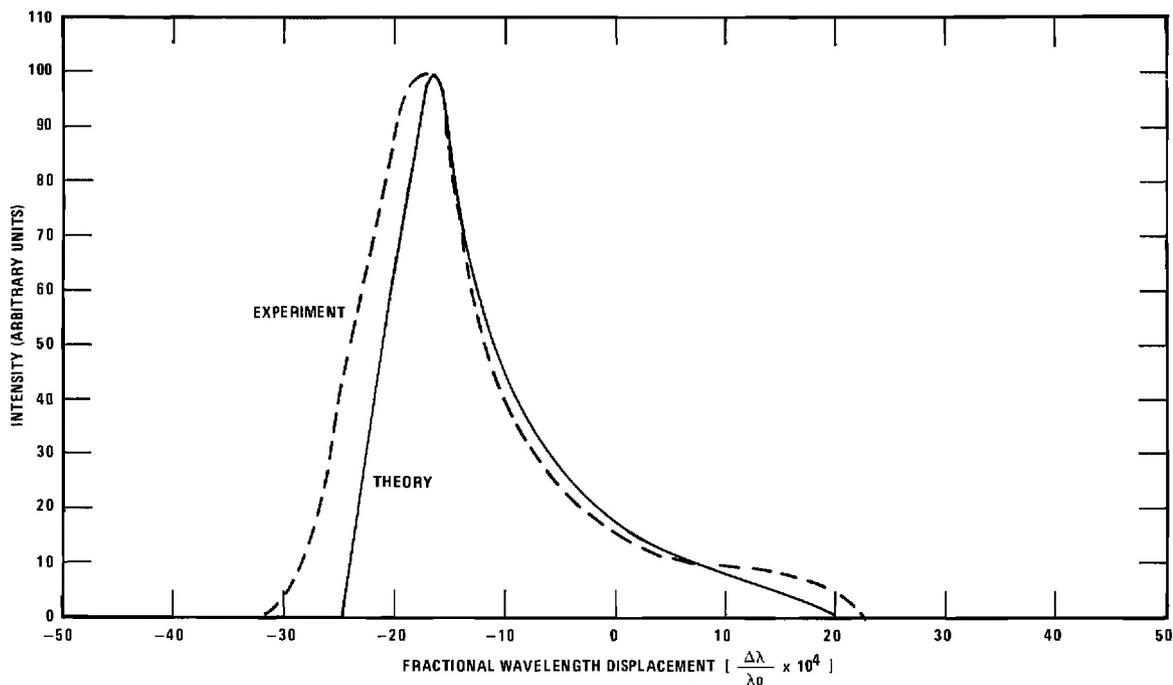


Figure 4. Schematic diagram of the apparatus used for study of light emission induced by ion impact on solid targets.

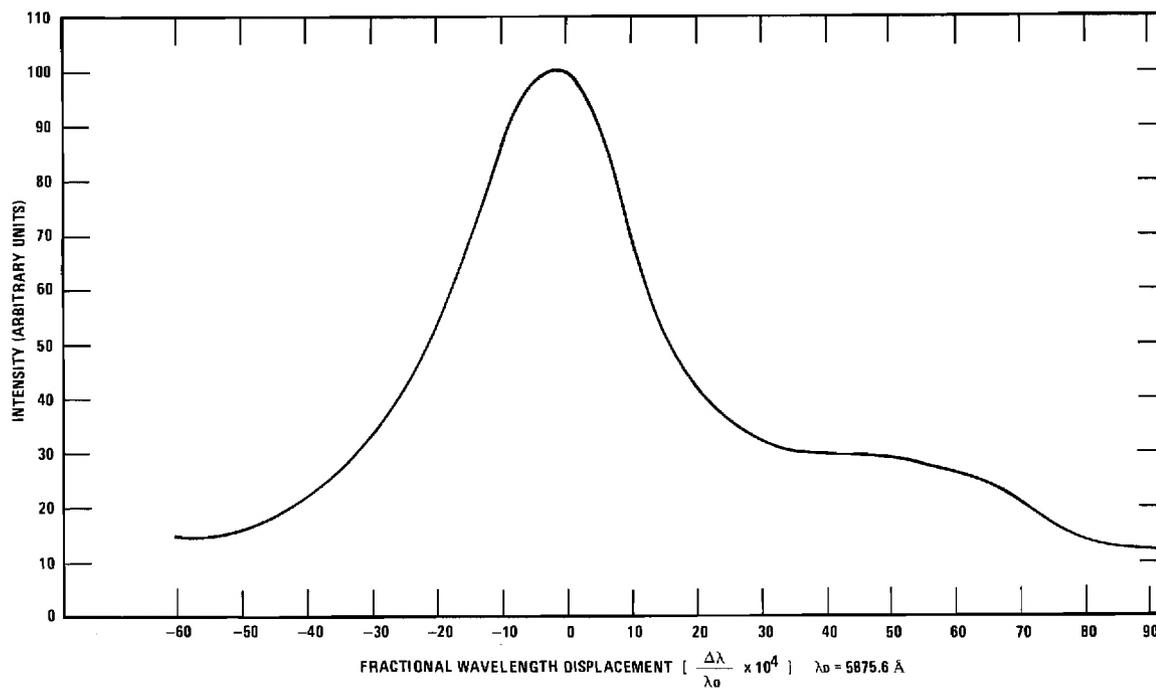
the targets used in our preliminary studies with the Ga. Tech apparatus were polycrystalline and situated in a background gas pressure at 10^{-7} mm; no serious attempt was made to clean the targets except for the sputtering action of the projectile beam itself. Thus the preliminary results obtained at Ga. Tech may be in error due to monolayers of contaminants on the surface. Modifications are now under way to provide the Ga. Tech apparatus with a better environment so that atomically clean surfaces may be utilized.

The research in Amsterdam involved a study of emissions induced by 2 to 10 KeV beams of H^+ and He^+ incident on targets of copper, nickel and gold. The work at Ga. Tech has thus far involved 100 - 1000 KeV beams of H^+ and He^+ incident on a polycrystalline target of copper; studies with tantalum and molybdenum are also under way at the present time. In all experiments we observe spectral lines of neutralized reflected projectiles; for He^+ beams the emission is from the triplet system of He, for H^+ beams we see the Balmer series of hydrogen. In addition to these spectral lines one also observes some broad band emissions whose origin is unclear.

The line emissions associated with the scattered projectiles are very broad, as much as 60 \AA in some cases. Fig. 5 shows two such line shapes for helium emission, one derived from the work in Amsterdam with a 10 KeV He^+ beam on copper the second from the experiment at Ga. Tech using a 200 KeV He^+ beam on a copper target. The presentation of Fig. 5 shows intensity (in arbitrary units) measured at some wavelength λ_1 plotted as a function of $\Delta\lambda/\lambda_0$; here λ_0 is the wavelength of the line emitted when the He atom is stationary and $\Delta\lambda$ is equal to $\lambda_1 - \lambda_0$. Thus the figure shows intensity as a function of fractional wavelength displacement from



(a)



(b)

Figure 5. Line shapes induced by He^+ impact on a copper surface
 (a) 3889 \AA ($3^3\text{P} - 2^3\text{S}$) line induced by 10 KeV He^+ at an incidence angle of 45° .
 (b) 5876 \AA ($3^3\text{D} \rightarrow 2^3\text{P}$) line induced by 250 KeV He^+ at an incidence angle of 65° .

the true wavelength emitted by a stationary atom. The work at low energies^{19,20} indicated that the breadth of the line varied with projectile energy; it was readily concluded that the line breadth was caused by the Doppler effect as excited atoms were reflected in various directions with little loss of kinetic energy. We have made predictions of these line shapes under the following assumptions;

(a) that the excited projectiles are reflected by elastic collisions on the surface monolayer;

(b) that the angular distribution of scattered excited projectiles is given by a Rutherford type cross section; that means the scattering cross section varies with scattering angle θ as $\text{sine}^{-4}(\theta/2)$;

(c) that particles scattered into the surface do not contribute to emission;

(d) that the decay of the excited state is by a normal radiative process. A line shape predicted on this basis for 10 KeV He^+ impact on copper is shown in figure 5a. Experiment and theory are in surprisingly good agreement. These assumptions also permit prediction of how the total line intensity varies with angle of projectile incidence on the surface; again prediction agrees well with experimental measurement^{19,20}, for projectile energies up to 10 KeV. In the higher energy data (Fig. 5b) for 200 KeV He^+ impact on copper we find that the line width is fairly symmetrical about a shift of zero; this indicates that most excited atoms are moving slowly. This suggests that the excited atoms are scattered at some considerable depth

inside the target and that they lose appreciable amounts of energy as they enter and as they leave again. This situation is generally consistent with our previous work reported last year²¹ from which we concluded that the velocities of the surface-scattered 200 KeV projectiles was only of the order 10 KeV. These observations do indicate that the measurement of line broadening can provide information on the velocity distribution of the scattered projectiles.

The other spectral feature observed when projectiles strike surfaces is a broad continuum emission extending over some hundreds of Angstroms. Fig. 6a shows the band observed with 10 KeV H^+ and He^+ incident on Gold. Fig. 6b shows the band observed for 100 KeV He^+ on copper. The origin of this band emission is not clear. It certainly is not due to scattered projectiles or atoms of target material ejected from the solid. We have examined the possibility of this emission being due to bremsstrahlung, transition radiation or interband transitions; none of these possibilities seems to fit the observations.^{19,20}

IX. Program for the Remainder of the Contract Year

We will continue our studies of neutral H atom excitation by impact at 5 - 30 KeV on targets of He and Ar [Eq. (2)]. Data will be in the form of differential cross sections as a function of scattering angle. In addition to the data for H(2s) production we will measure elastic scattering of H, ionization of H to H^+ , and charge transfer production of H^- .

The work on scattering of projectiles from solids will also be continued. The first objective is to provide an ultra-high-vacuum environment

21) E. W. Thomas, et al. Progress Report No. 7, AEC Report No. ORO-2591-61.

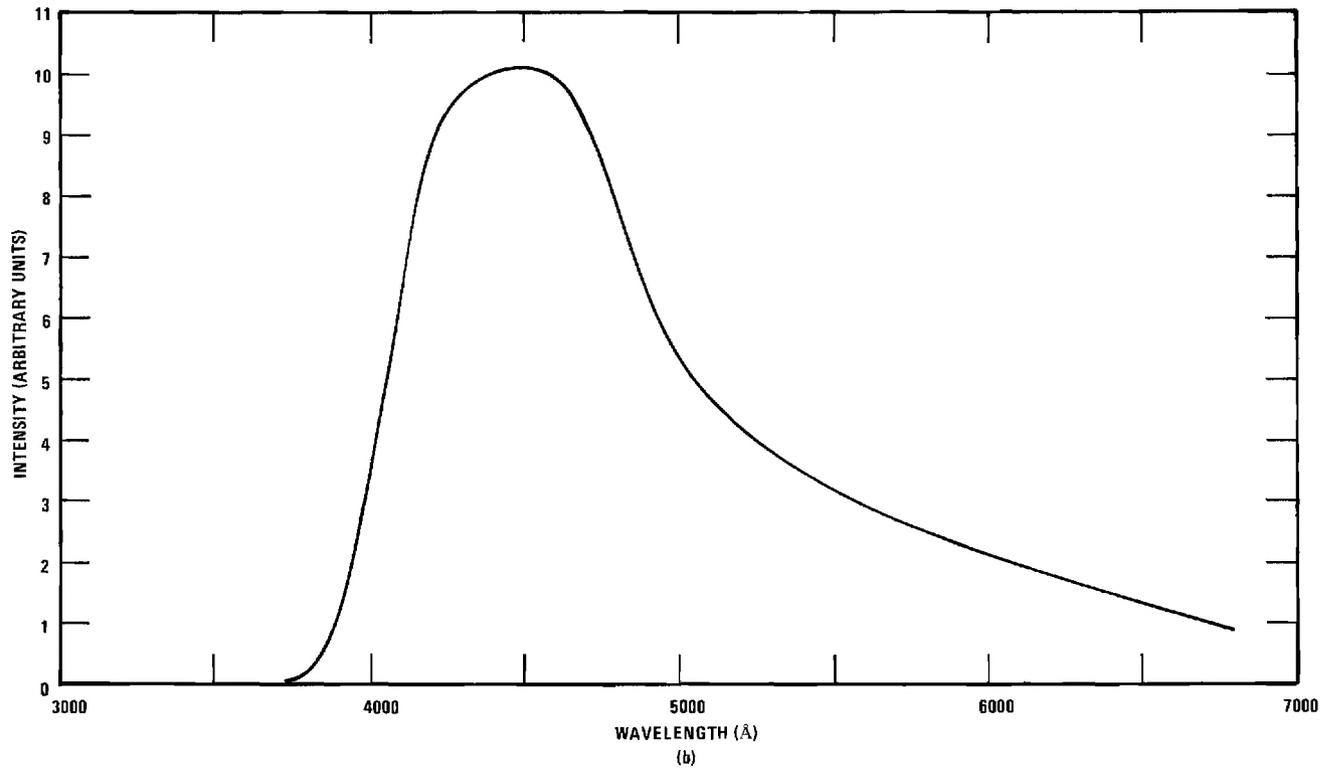
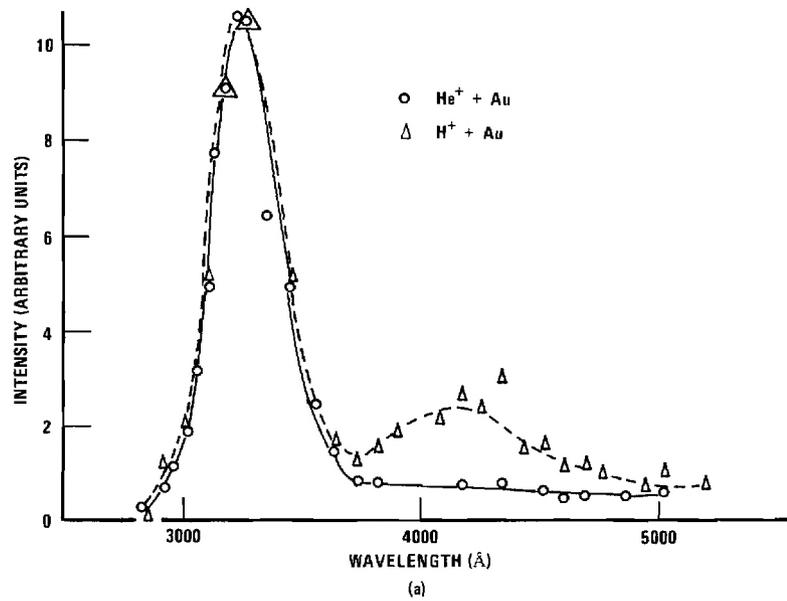


Figure 6. Broad band emission observed for He^+ impact on a metal target.

(a) 10 KeV energy at an angle of 45° on Au.

(b) 100 KeV energy at an angle of 65° on Cu.

[Note - Atomic line emissions have been deliberately removed from the trace. Data below 4000 Å on curve (b) should be disregarded].

for the solid targets; it will then be possible to guarantee atomic cleanliness of the target. It is expected that this work will occupy much of the remainder of the contract year.

X. Program for the Future

Most of the emphasis will be on the study of particle impact on solids. H^+ and He^+ projectiles will be directed on polycrystalline Cu, Mo and Ta; projectile energies from 5 to 1000 KeV will be available using two different accelerators. Detailed study of spectral line shapes should permit determination of the scattered projectile's velocity distribution. A limited amount of effort will be devoted to the broad continuum bands, principally to determine their origin. Detailed study of these bands will be carried out only if they appear to have some bearing on phenomena that may be relevant to a controlled thermonuclear device.

Further studies of atomic collision cross sections will be principally directed to collisions of H^0 with a variety of targets at impact energies for 5 to 30 KeV. Measurements as a function of angle, will be made of the scattered H^0 , $H(2s)$, H^+ and H^- . This represents a continuation of the work already in progress and should be finished within twelve months. If time permits, an attempt will be made to study differential cross sections for $H(2s)$ formation as H^+ traverses a target of cesium. This is an interesting resonant reaction that gives rise to very high cross sections. It offers a possibility of producing an H beam with more excited atoms than ground state atoms; this constitutes an inverted population and might provide a basis for laser action. This reaction is also expected to be one of the most efficient methods of producing H^0 beams.

XI. Publications and Travel

A total of ten papers have been published or submitted for publication during the present reporting period.

- (i) "Measurement of Differential Atomic Collision Cross Sections" by R. L. Fitzwilson and E. W. Thomas. Rev. Sci. Inst. 42, 1864 (1971). AEC Report No. ORO-2591-57.
- (ii) "Formation of Fast Excited H Atoms, I. Charge Transfer Neutralization of H^+ in He and Ar" by J. C. Ford and E. W. Thomas. Phys. Rev. A. 5, 1694 (1972). AEC Report No. ORO-2591-58.
- (iii) "Formation of Fast Excited H Atoms, II. Charge Transfer of H^+ on Molecular Gases" by J. C. Ford and E. W. Thomas. Phys. Rev. A. 5, 1701 (1972). AEC Report No. ORO-2591-59.
- (iv) "Formation of Fast Excited H Atoms, III. Collisional Dissociation of H_2^+ and H_3^+ on Helium" by J. C. Ford, F. M. McCoy, R. Conrads and E. W. Thomas. Phys. Rev. A. 5, 1705 (1972). AEC Report No. ORO-2591-60.
- (v) "Angular Distribution of Metastable Hydrogen Formed by Electron Capture in a Helium Target" by R. L. Fitzwilson and E. W. Thomas. Phys. Rev. A. 6, 1054 (1972). AEC Report No. ORO-2591-62.
- (vi) "Angular Distribution of Metastable Hydrogen Formed by Dissociation of H_2^+ " by I. Sauers, R. L. Fitzwilson, J. C. Ford and E. W. Thomas. Phys. Rev. A. 6, 1418 (1972). AEC Report No. ORO-2591-63.
- (vii) "Light Emission Induced by H^+ and He^+ Impact on a Clean Copper Surface" by C. Kerkdijk and E. W. Thomas. Physica (to be published). AEC Report No. ORO-2591-65.

- (viii) "Photon Emission Induced by Impact of Fast Ions on Metal Surfaces" by C. Kerkdijk and E. W. Thomas. Surface Science (to be published). AEC Report No. ORO-2591-66.
- (ix) "Formation of Fast Excited H Atoms Induced by Impact on H^+ , H_2^+ and H_3^+ Atoms on Molecular and Noble Gas Targets" by R. J. Conrads, T. W. Nichols, J. C. Ford and E. W. Thomas. Phys. Rev. (submitted for publication). AEC Report No. ORO-2591-67.
- (x) "Collisional Dissociation of H_2^+ and H_3^+ on Molecular Targets Leading to Formation of Excited Hydrogen Atoms" by R. J. Conrads, J. C. Ford and E. W. Thomas. J. Chem. Phys. (submitted for publication). AEC Report No. ORO-2591-68.

Two of these papers [Nos. (vii) and (viii)] represent work carried out while one of us (EWT) was at the FOM Institute for Atomic and Molecular Physics in Amsterdam. Although no AEC funds were expended in the research the preparation of the material for publication was in part carried out after Dr. Thomas returned to the U.S.A.; the time used in this final preparation was supported by the present contract.

Four papers were presented at conferences.

- (i) "Formation of Metastable Hydrogen by Collisional Dissociation of H_2^+ and H_3^+ " by I. Sauers and E. W. Thomas. Annual Meeting of the Atomic and Molecular Physics Section of the Physical Society (London), University of Sussex, April 1971.
- (ii) "Photon Emission Induced by Impact of Fast Ions on Metal Surfaces" by C. Kerkdijk and E. W. Thomas. Conference on Ion-Surface Interactions, Garching, September 1972.

- (iii) "Angular Distribution of H(2s) Formed by Collisional Excitation of Hydrogen Atoms" by I. Sauers, T. W. Nichols and E. W. Thomas. Annual Meeting of the Division of Electron and Atomic Physics. Menlo Park, November 1972.
- (iv) "Formation of H(3s) by Charge Transfer Neutralization of H⁺ and by Dissociation of H₂⁺ and H₃⁺" by R. J. Conrads, T. W. Nichols, J. C. Ford and E. W. Thomas. Annual Meeting of the Division of Electron and Atomic Physics, Menlo Park, November 1972.

During the year E. W. Thomas made visits to the Universities of Aarhus and Copenhagen (not at AEC expense) where seminars were given on the research conducted at Ga. Tech. Visits have also been made to the Oak Ridge National Laboratory and to the European Space Research Organization in Noordwijk (The Netherlands).

XII. Personnel

The work described in this report was under the jurisdiction of Dr. Thomas, Principal Investigator. For the period up till July 1972 Dr. Thomas was on leave of absence at the FOM Institute for Atomic and Molecular Physics, (Amsterdam, The Netherlands). During Dr. Thomas' absence the day to day direction of the program was in the hands of Dr. J. C. Ford, Co-Principal Investigator; however Dr. Thomas retained overall responsibility for the project. The arrangements for supervision of the project during Dr. Thomas' absence were made with the full agreement of AEC headquarters.

For the period starting 1 July 1972 Dr. Thomas devoted 20% of his time to this project during the academic year and 80% of full time during

the summer. Prior to the 1 July 1972 Dr. Thomas drew no financial support from this contract but continued to participate in the direction of the work and the writing of articles.

For the period up till June 1972, Dr. J. C. Ford devoted 40% of his full time to this project. Dr. Ford has left Ga. Tech and is now employed by the Radiation Division of Varian.

Mr. Isidor Sauers has been supported for half of full time on this contract. He has been responsible for the low energy experiments discussed in part VII of this report. It is expected that he will write a Ph.D. thesis on this work in about 9 months time.

Three other graduate students have been supported for about one quarter of full time; these were Messers R. Conrads, W. Hill and T. W. Nichols. Mr. Nichols continues to work on the project, devoting most of his effort to the study of light emission induced by particle impact on solids.

XIII Incident Report

There have been no incidents for which a report is required during the performance of the research under this contract in the present reporting period.