

IONIZATION AND CHARGE TRANSFER
CROSS SECTIONS

FINAL REPORT ON TASK A, OF CONTRACT NO. AT-(40-1)-2591

Covering the Period

September 1, 1959 to February 28, 1970

By D. W. Martin

E. W. Thomas

Report No. ORO-2591-46

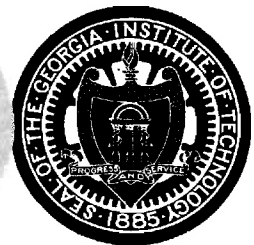
Contract No. AT-(40-1)-2591

U. S. ATOMIC ENERGY COMMISSION

OAK RIDGE, TENNESSEE

17 April

1970



School of Physics
GEORGIA INSTITUTE OF TECHNOLOGY
Atlanta, Georgia

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I. Title

Ionization and Charge Transfer Cross Sections: Final Report

II. Introduction

This report summarizes a program of experimental studies of collisional ionization and charge transfer processes that has been conducted at the Georgia Institute of Technology under Contract No. AT-(40-1)-2591 with the U. S. Atomic Energy Commission. This work was initiated on September 1, 1959 as the sole subject of that contract. On March 1, 1965 the scope and financing of the A. E. C. support were increased (Modification No. 7 of the Contract) to include additional experimental studies of collisionally induced emission and excitation processes. Since that time, the continuing studies of ionization and charge transfer have been designated as Task A of the Contract, while the new program of excitation and emission studies was designated Task B. The programs under tasks A and B have remained essentially distinct; that of Task A has in recent years been under the direction of Dr. D. W. Martin, while that of Task B has from its origin been under the direction of Dr. E. W. Thomas.

Support under this contract for the ionization and charge transfer studies of Task A has been terminated as of February 28, 1970. The present report is provided as a terminal summary report of the achievements of this program over its entire history, and of its status as of the termination date.

Support for the emission and excitation studies, formerly Task B, is at present continuing under the same contract (modification No. 12),

under the direction of Dr. E. W. Thomas. Since a status report¹ has recently been issued concerning this program, there will be no further mention of these continuing studies in the present report.

III. General Objectives of This Program

The broad program of absolute cross section measurements for charge rearrangement collisions of fast hydrogen and helium ions and atoms in gaseous targets has included studies with all of the five incident-particle states H^0 , H^+ , He^0 , He^+ , and He^{++} . The energies of the incident particles, obtained from a 1-MV Van de Graaff positive ion accelerator, lie in the range 0.15 to 1.00 MeV. This range extends to higher energies than those of most previous similar measurements elsewhere, and reaches well into the range where theoretical computations in the Born approximation may be expected to be valid, if they are properly formulated and make use of sufficiently accurate wave functions.

Exact wave functions are of course available for all of the simple projectiles used here, with the exception of the neutral He^0 atoms, but exact functions are not available for any molecule that is stable in a static gas at room temperature. Born calculations actually exist for only a few of the simplest possible target molecules, and, for the most part, only for the simplest rearrangement processes, in which only one electron in the whole projectile-target system changes its state. Although for some cases the results were rather confidently expected to be valid for sufficiently large energies, it has not really been possible to predict theoretically the minimum energy below which the approximations will become inadequate. It has been clear that progress in the fundamental understanding

of rearrangement collisions requires that every new development in theory be subjected to detailed experimental examination, to establish the bounds of its validity, and point the way for further developments.

It has always been one of the main objectives of this experimental program to perform just this kind of role, by including target molecules for which some theoretical work has been done, and by attempting to determine, as far as possible, the absolute cross sections for single, well-defined, and simple rearrangement processes. In addition, we have sought to cover systematically a sufficient variety of cases to reveal any general patterns in the dependence of the cross sections on the general properties of the projectile and target particles, and to provide an empirical basis for extrapolation, for the estimation of probable cross section values for a larger variety of cases. This was the basic idea behind the use as projectiles, in the first phase of this work, of both hydrogen and helium, in all of their possible charge states, and in the choice of the array of target gases studied. The latter has included He and Ar, as both the simplest noble gas molecule and another heavier and more complex one, and H_2 and N_2 , representing the simplest diatomic molecule and one heavier one. A few additional target molecules have been included in the studies involving some, but not all, of the five different projectile states.

IV. Total Cross Section Measurements

In the first phase of the present program, the quantities which were directly measured, by a simple total-charge-collection method using a parallel-plate geometry, were the cross sections for the gross total

production by the fast projectiles of slow positive ions and of free electrons and/or negative ions. From our results and from such information as was available on the same cases from elsewhere on the cross sections for (a) electron stripping from the fast projectile, (b) electron capture by the fast projectile, and (c) the relative production rates of multiply-charged recoil ions, we made estimates of the cross sections for pure ionization collisions. These results were compared with all available theoretical results, with similar experimental results from elsewhere, and with experimental results for electron bombardment of the same targets; finally, the results were further compared with each other, for all of the several projectile states and target molecules studied.

All of the results from these early studies of total collision cross sections have been published in the open literature²⁻⁶ and will not be repeated in detail here. However, the following general remarks are offered in summary of some of the conclusions which were drawn from them.

Our results for five different projectile-particle states and a variety of target molecules provided a standard set of data against which one could test the general theoretical predictions of the Born and Bethe-Born Approximations, as well as the specific predictions in those cases for which they were available. By comparison of the cross sections for simple ionization by H^+ and He^{2+} , together with published results for electrons, it was shown²⁻⁴ that at sufficiently high impact velocities the cross sections for these point-charge ions depend only on the impact velocity and the square of the ion charge, and are independent of the mass of the ion. This finding confirmed a well known general prediction of the

theory for high energies, and the data established the lower bound of the energy range for its validity. By further comparison with these cross sections of those for incident H^0 and He^+ particles, it was shown that the simple ionization produced by the latter projectiles was equivalent, in a variety of target gases, to the predicted ionization by point-charge ions with "effective charges" of 0.80 and 1.2 times the proton charge, respectively. These findings provide a very effective extrapolation scheme for predicting the high-energy ionization cross sections for all five projectiles, on a given target, provided that measurements are available for any one of them.

The ionization cross sections obtained for He^0 did not, by contrast, suggest the validity of an effective charge concept for this projectile. Although the energy dependence of the cross sections in various targets were in general similar to those of the other projectiles at the same impact velocity, the appropriate value of the effective charge varied with the target in a manner that did not seem to be systematic in any way. It is presently thought to be possible that the singular behavior observed in this one case might have been related to a contamination of the projectile beam with metastable particles. Several tests which were performed were believed, at the time, to be adequate to demonstrate that there were no effects of such particles present; however, more recent work elsewhere has indicated that He^0 beams formed by charge-transfer neutralization at these energies may contain a substantial fraction of such metastable particles.

For those cases for which detailed calculations of ionization cross sections were available, absolute comparison with our measurements were made. It was shown³ that, in general, the Born Approximation is quite accurate for H^+ ionization for energies above about 400 keV.

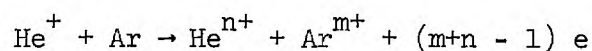
V. Measurements of Multiple Ionization and Scattering

The "pure ionization" cross sections referred to above were actually called "apparent" ionization cross sections because the method used, in which only the total current of the slow positive ions produced was measured, did not distinguish the fractions of this current due to doubly-charged, triply-charged, etc., slow ions, and hence the cross section obtained was a weighted sum of the true cross sections for the ionization events producing slow ions of various charges. In addition, quite apart from the matter of experimental errors, our method could produce only an estimate of even the apparent ionization cross section. The reason is that, in subtracting from the total (positive or negative) ion production that part due to charge-changing collisions, in which the fast projectile either gained or lost electrons, it was never known from the available information (except within certain limiting bounds) what fraction of the charge-changing events were accompanied by the simultaneous ejection of extra electrons from the target. The extra contribution to the total ion currents from any such events should also be subtracted, if the remainder was to represent even the pure apparent ionization, as defined above. Except for upper bounds that could be set from measurements of the total production cross sections for multiply-charged slow positive ions, information on the cross sections for simultaneous ionization and

charge transfer, or simultaneous ionization and projectile stripping, can be obtained only through an experiment in which one determines the final charge states of both of the partners from a single collision. This obviously requires a coincidence experiment.

In any collision process the direction of the projectile is changed as a result of the interaction. Similarly the target particle will recoil in a definite direction related to the angular scattering of the projectile and the inelastic energy loss associated with the collisional event. Determination of these angles in an experiment designed to detect, in coincidence, the scattered projectile and recoiling target, can provide considerable insight to the mechanism of the inelastic event.

The project has embarked upon the development of an experimental arrangement to allow determination of the charge state and direction of motion, after the collision, of both the recoiling target and scattered projectile systems. There is provision for the capability of detecting both particles in coincidence. As an example of the type of process which can be readily studied with this type of apparatus one may consider the charge rearrangement mechanisms which can occur as the result of the impact of He^+ on a target of Ar. The full rearrangement equation can be most simply written as:-



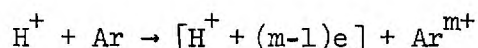
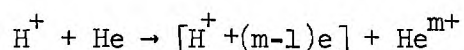
The new apparatus allows the determination of the charge states and direction of motion of both post collision systems. In contrast the total ionization measurements performed earlier in this program, and

described in IV above, provide only a weighted sum of the cross sections for all processes leading to the formation of ionized argon, Ar^{m+} , without any information on the relative population of different states of ionization.

The apparatus involves analyzer systems for both the scattered projectiles and recoil target atoms. Each analyzer samples particles emerging from the interaction region in a particular direction. Each analyzer provides analysis of the charge state of the particle. Finally both systems are provided with single-particle detectors, the outputs of which may be counted separately and in coincidence. The analyzer systems may be rotated about a common axis providing data on the angular distribution of the post collision particles.

As a preliminary to the pursuit of coincidence studies it has proved advantageous to carry out some separate non-coincidence studies of angular and charge state distributions of the scattered projectiles and of the charge state distributions of the recoil target atoms. At the time of writing the final step of detecting both particles in coincidence has not yet been successfully completed.

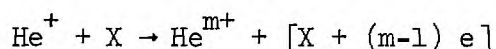
Studies of the recoiling target atoms have concentrated on the relative proportions of different charge states of target systems for the collision processes:-



The square brackets indicate that the present experiments provide no information on the state of ionization of the components contained within

them. The data provide information on the relative proportion of the recoiling target atoms in the different states of ionization. Rudimentary studies have also been made on the angular and velocity distributions of the recoiling products. This work has been accepted for publication and will appear shortly (See item A6 in Sec. VI. Publications).

Recently we have completed a study of the angular and charge state distributions of He^+ projectiles scattered as the result of impact on targets of He, Ne and Ar. The reaction equation may be written:-



Our present experiment provides no information on the post collision state of the components contained within the square brackets. Our experiments show that the total cross section for the scattering of all projectiles, irrespective of charge, can be readily predicted by classical theoretical treatments provided that the potential of interaction between the projectile and target takes into account the screening of the nucleus by the electrons. The relative population of different charge states cannot be readily predicted by theory but some useful correlations may be provided by considering a statistical distribution of the available electrons between the various possible bound and continuum states. These studies have been fully documented in our most recent annual report⁸ and are presently being prepared for publication (See item A7 in Sec. VI. Publications).

The final stage in the development of this technique, the detection of the recoil target particle and the scattered projectile in coincidence, is still being developed at the present time. Recent work has centered on

improvements in the recoil-ion analyzer, involving the additions of magnetic shielding and an improved type of detector, to extend its sensitivity to lower recoil-ion energies. Evaluation of the reassembled analyzer should be accomplished shortly. There would then appear to be no insurmountable difficulties in going on to perform the type of coincidence studies described above.

VI. Publications

All the results of this work have been fully documented in periodic technical reports. Also the majority have been published in the open literature; the few remaining results are being prepared for publication at the present time. We have deliberately refrained from including detailed presentation of data in this terminal report since all of the work has been previously written up elsewhere.

The following is a listing of all publications and major reports that have been generated by this project. We list first full length articles published in The Physical Review, including a final article still in preparation. This is followed by a list of several papers presented orally at major international conferences for which there is a generally available volume of proceedings. While the texts published therein have, in most cases, been technically designated as "abstracts", they were of substantial length and detail, and included figures with graphical presentations of results. Finally we list four Ph.D. theses based on this research, which were submitted by their respective authors in partial fulfillment of the requirements for the Ph.D. degree (all were subsequently awarded this degree by the Georgia Institute of Technology).

Each of these theses was also adapted and issued by Georgia Tech as a Technical Status Report or a Technical Report, as is indicated in each case.

Excluded from this list are a few additional minor conference presentations for which only a brief abstract was published; in all such cases the data presented were later published elsewhere in one of the listed papers.

A. Journal Articles:

- A1. "Ionization Cross Sections for Protons on Hydrogen Gas in the Energy Range 0.15 to 1.10 MeV," by J. W. Hooper, E. W. McDaniel, D. W. Martin, and D. S. Harmer, Phys. Rev. 121, 1123 (1961).
- A2. "Comparison of Electron and Proton Ionization Data with the Born Approximation Predictions," by J. W. Hooper, D. S. Harmer, D. W. Martin, and E. W. McDaniel, Phys. Rev. 125, 2000 (1962).
- A3. "Cross Sections for Ion and Electron Production in Gases by Fast Helium Ions (0.133-1.0 MeV). I. Experimental," by R. A. Langley, D. W. Martin, D. S. Harmer, J. W. Hooper, and E. W. McDaniel, Phys. Rev. 136, A379 (1964).
- A4. "Cross Sections for Ion and Electron Production in Gases by Fast Helium Ions (0.133-1.0 MeV). II. Comparison with Theory," by D. W. Martin, R. A. Langley, D. S. Harmer, J. W. Hooper, and E. W. McDaniel, Phys. Rev. 136, A385 (1964).
- A5. "Cross Sections for Ion and Electron Production in Gases by 0.15-1.00 MeV Hydrogen and Helium Ions and Atoms," by L. J. Puckett, G. O. Taylor, and D. W. Martin, Phys. Rev. 178, No. 1, 271 (1969).
- A6. "Analysis of Recoil He^+ and He^{++} Ions Produced by Fast Protons in Helium Gas," by L. J. Puckett and D. W. Martin, Phys. Rev. A (in press, scheduled for May, 1970).
- A7. "Scattering of He^+ Ions by Noble Gases at High Energies," by G. O. Taylor, E. W. Thomas, and D. W. Martin, (in preparation for submission to Physical Review A, 1970).

B. Papers Presented at International Conferences:

- B1. "Ionization Cross Sections for Protons Incident on He, Ne, Ar, H₂, N₂, O₂, and CO," by J. W. Hooper, E. W. McDaniel, D. W. Martin, and D. S. Harmer, Abstracts of Papers of the 2nd International Conference on the Physics of Electronic and Atomic Collisions, W. A. Benjamin, Inc., New York (1961), p. 67.
- B2. "The Ionization by H⁺ Ions in the Energy Range 0.15-1.1 Mev," by E. W. McDaniel, J. W. Hooper, D. W. Martin, and D. W. Harmer, Proc. Fifth International Conference on Ionization Phenomena in Gases, Munich 1961, North-Holland Pub. Co., Amsterdam (1962), p. 60.
- B3. "Production of Slow Electrons and Positive Ions in He, Ne, Ar, H₂, N₂, O₂, and CO by Energetic Helium Ions," by D. W. Martin, R. A. Langley, J. W. Hooper, D. S. Harmer, and E. W. McDaniel, Proc. 3rd International Conference on the Physics of Electronic and Atomic Collisions, London 1963, North-Holland Pub. Co., Amsterdam (1964), p. 679.
- B4. "Ionization and Charge-Transfer Cross Sections for Fast Neutral Helium Atoms (0.15 to 1.0 MeV)," by D. W. Martin, L. J. Puckett, and G. O. Taylor, Abstracts of Papers of the IVth International Conference on the Physics of Electronic and Atomic Collisions, Quebec 1965, Science Bookcrafters, New York (1956), p. 280.
- B5. "Analysis of the Recoil Ions Produced by Fast Protons," by D. W. Martin, L. J. Puckett, and G. O. Taylor, Abstracts of Papers of the Vth International Conference on the Physics of Electronic and Atomic Collisions, Leningrad 1967, Publishing House "Nauka", Leningrad Section (1967), p. 207.
- B6. "Investigations 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, Abstracts of Papers of the Sixth International Conference on the Physics of Electronic and Atomic Collisions, M.I.T. 1969, MIT Press, Cambridge (1969), p. 231.

C. Ph.D. Theses Issued by Georgia Tech as Reports:

- C1. "Ionization Cross Sections for Protons Incident on Helium, Neon, Argon, Hydrogen, Nitrogen, Oxygen, and Carbon Monoxide in the Energy Range 0.15-1.10 MeV," by John William Hopper (1961).
Adapted as Technical Status Report No. 7, Project B-176,
"Ionization and Charge Transfer Cross Sections, Phases I and II: H⁺ Ions Incident of He, Ne, Ar, H₂, N₂, O₂, and CO Targets," by E. W. McDaniel, J. W. Hooper, D. W. Martin, and D. S. Harmer (June 1, 1961).

(Ph.D. Theses Issued by Georgia Tech as Reports, cont'd):

- C2. "Total Cross Sections for the Production of Positive Ions and Free Electrons in Gaseous Targets," by Robert Archie Langley (1963).
Adapted as Technical Status Report No. 15, Project B-176, "Ionization and Charge Transfer Cross Sections: Summary of Results for Fast Helium Ions," by R. A. Langley, D. W. Martin, D. S. Harmer, J. W. Hooper, and E. W. McDaniel (June 1, 1963).
- C3. "Absolute Total Apparent Ionization, Electron Stripping, Electron Capture, and Partial Ionization Cross Sections in the Energy Range 0.15-1.00 MeV," by Lawrence Jackson Puckett (1967).
Adapted as a Technical Report, "Ionization and Charge Transfer Cross Sections for H^0 , He^0 , and He^{++} Beams in the Energy Range 0.15-1.00 MeV, and the Design of a Differential Recoil-Ion Analyzer," by L. J. Puckett, D. W. Martin, and G. O. Taylor (AEC Document No. ORO-2591-35, November 1, 1967).
- C4. "Scattering of He^+ Ions by Noble Gases at High Energies," by George Ormsbee Taylor, Jr. (1969).
Adapted as a Technical Report of the same title, by G. O. Taylor, Jr., D. W. Martin, and E. W. Thomas (AEC Document No. ORO-2591-42, September, 1969).

VII. Summary

This project has long been the major program in the USA designed to study atomic collision phenomena at high energies. The objective throughout has been to provide accurate data on well defined processes that can be compared with theoretical predictions. The chief value of this program to the understanding of collisional phenomena has been that it concentrates on studies at high impact energies where tractable theoretical predictions are expected to be valid. It has been shown that the well known Born approximation can provide good theoretical predictions of the observed single ionization processes in the target. Classical predictions serve to provide accurate predictions of the angular scattering of fast projectiles. It has also been shown that theoretical predictions

of multiple ionization processes by statistical methods are surprisingly successful.

The final objective of this program has been to carry out measurements of the post collision states of ionization and directions of motion of both projectile and target atoms in coincidence. Although this objective has not been achieved during the tenure of this contract it is hoped that the full potentialities of this technique can be realized in the near future.

VIII. References

1. E. W. Thomas, J. L. Edwards, J. C. Ford, "Emission and Excitation Cross Sections," Progress Report No. 5, AEC Report No. ORO-2591-44, 30 November 1969.
2. J. W. Hooper, et al., Phys. Rev. 121, 1123, 1961.
3. J. W. Hooper, et al., Phys. Rev. 125, 2000, 1962.
4. R. A. Langley, et al., Phys. Rev. 136, A379, 1964.
5. D. W. Martin, et al., Phys. Rev. 136, A385, 1964.
6. L. J. Puckett, et al., Phys. Rev. 178, 271, 1969.
7. A. B. Wittkower, et al., Proc. Phys. Soc. 91, 862, 1967.
8. L. J. Puckett, et al., Phys. Rev. (To be published).
9. D. W. Martin, G. O. Taylor, "Ionization and Charge Transfer Cross Sections" Progress Report No. 22, AEC Report No. ORO-2591-43, 30 November 1969.