The Retardation and Neutralization of Argon Ions in Helium

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An apparatus is described for making direct measurements of the loss of forward velocity of argon ions in single encounters with helium atoms. Results are given in the form of collision cross sections for given percentage retardations, for ions of energies of 1000, 800, 600, 400, and 200 ev. The cross sections are zero for energy losses greater than 33.1 percent and increase both with decreasing percentage loss and decreasing beam speed. A correction to the readings necessary to obtain the results proves to be of unusual interest because the correction is itself the cross section for charge exchange between the argon ions and the helium atoms. Quantitative values of the cross sections for this unusual charge exchange are thus obtained. The values run roughly from 3.5 to 4.5×10^{-16} cm^2 per molecule in the energy range studied, compared with 28×10^{-16} cm^2 for A^+ ions in A and 63×10^{-16} cm^2 for N_2^+ ions in N_2. All the results are in sufficiently precise form to invite detailed theoretical analysis.

INTRODUCTION

INVESTIGATIONS of the collision of ions and atoms have led to a number of experimental procedures for studying the processes of scattering, neutralization, and retardation, which affect a beam of ions shot through a gas. Various experimenters[1] using the Dempster mass spectrograph technique, and in the majority of cases employing the alkali ions emitted by Kunsman filaments, have published data on the relative importance of these processes for different ion-gas combinations. The experimental procedures were frequently such that quantitative specification of cross sections for the various processes was not possible. This was particularly true when retarding collisions were present, wherein the projectile ions suffered loss of forward speed without undergoing appreciable deviations.

Theoretical considerations by A. V. Hershey[2] indicated that results of the retardation of ions in passing through a gas would involve fewer experimental difficulties and be easier to analyze than angular scattering results. Retardation measurements are most readily made by using ions very much heavier than the gas atoms of the retarding gas as such a combination of masses results in small angular scattering. One of the investigators of the Dempster group, J. S. Thompson,[3] sent Cs^+ ions through helium and hydrogen in apparatus similar to that employed by Mayer[4] in his classic studies of the absorption of electron beams in gases. He developed a method for deriving the cross sections for retardation from the resulting data on multiple collisions. The present paper describes an experiment devised to measure retardation cross sections directly for A^+ ions in helium gas under conditions such that only single collisions occur in the test region to any appreciable extent.

APPARATUS AND EXPERIMENTAL PROCEDURE

The circuit diagram of Fig. 1 illustrates the essential parts of the apparatus. "*S*" represents the ion source, which is a modification of the "High Efficiency Ion Source" described by

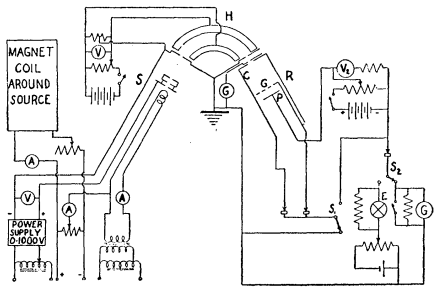

FIG. 1. Schematic diagram of the apparatus.

[1] A. J. Dempster, Phil. Mag. **3**, 115 (1927); R. B. Kennard, Phys. Rev. **31**, 423 (1928); I. W. Cox, Phys. Rev. **34**, 1426 (1929); K. H. Bracewell, Phys. Rev. **54**, 639 (1938).

[2] Private communication. The writer is pleased to acknowledge his indebtedness to Dr. Hershey for furnishing his memoranda on the subject of retardation measurements. Many of his suggestions are incorporated in the experimental procedure.

[3] J. S. Thompson, Phys. Rev. **35**, 1196 (1930).

[4] H. F. Mayer, Ann. d. Physik **64**, 451 (1921).

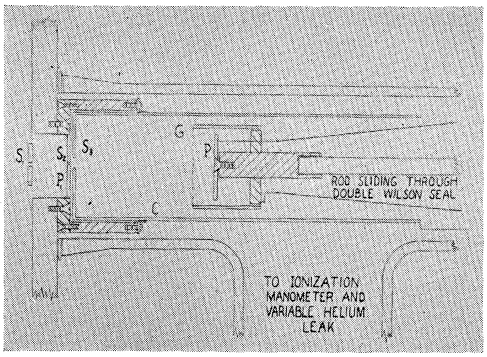

FIG. 2. Details of retardation chamber electrode assembly.

Finkelstein.[5] The source consists essentially of a combined magnetic and electric electron trap which maintains a high electron density in the source and serves the double purpose of using the electrons in a highly efficient way for ionization and of neutralizing the ordinarily inhibitory space charge of the positive ions. The source as used in this experiment is capable of currents of 2 to 10 ma of A^+ ions and has the highly desirable characteristic of operating at extremely low gas pressures. The beam energy used ranged between 200 and 1000 ev. Under these conditions the beam was not too sharply defined but was of such great intensity that it could be collimated with slits and homogenized with an electrostatic velocity selector to give a sharp beam of 10^{-7} amp., highly suitable for the present experiment.

"*H*" represents the velocity selector.[6] "*R*" is the retardation chamber, details of which appear in Fig. 2. The source and retardation electrodes were housed in flanged Pyrex pipe sections (diam. 3 in.) which were attached to a heavy steel box of diamond-shaped cross section containing the 127° selector. This steel box shielded the selector from the axial magnetic field used with the source and served to connect the entire system directly onto the mouth of a three-stage fractionating diffusion pump[7] of 80 liters per sec. speed. Rubber gasket technique was used throughout in making vacuum seals. Argon of 99.6 percent purity was fed into "*S*" through a variable leak,[8] and helium of 98.2 percent purity into "*R*." The gas pressures were measured by a D-79510 ionization manometer connected to a control and measuring unit similar to that described by Bowie.[9]

Power (a.c.) was supplied to the apparatus through a 1 kva Sola constant voltage transformer, and direct current for the source and source magnet coils was supplied by a generator maintained at constant voltage by a General Electric type CD stabilizer. General Radio "Variacs" were used for adjusting accelerating voltages and source-cathode heating current. A shelf of "*B*" batteries suspended by long insulating strips of "Amphenol *A*," a polystyrene derivative, supplied the retardation potentials. These batteries, the switches, and the associated wiring which carried the positive ion currents were well shielded and were supported where necessary with "Amphenol *A*."

The retardation electrode assembly shown in Fig. 2 consisted of a can, a grid, and a plate. The grid and plate were moved as a unit by the sliding motion of the supporting rod through a double Wilson seal.[10] The region between the two seals was kept filled with helium at atmospheric pressure. For most measurements the can and grid were grounded so that the space between S_3 and G was practically field-free. The circuit connections were so arranged that A^+ ions of a chosen energy, V_1, would be shot into He atoms in the field-free space where they would suffer their collisions along a path whose length could be varied at will, after which they would enter the analyzing region between the grid and the plate. The plate was held at a positive potential of V_2 volts with respect to ground so that all ions which had energy less than V_2 electron volts would be unable to reach the plate. Throwing switch S_1 (Fig. 1) upward connected the entire retardation electrode assembly as a Faraday cage by means of which the total ion current I_0 in the beam was measured.

Ideally, if the interaction of an A^+ ions with a helium atom caused the former to be so retarded that its residual energy was less than

[5] A. T. Finkelstein, Rev. Sci. Inst. **11**, 94 (1940).
[6] A. L. Hughes and V. Rojansky, Phys. Rev. **34**, 291 (1929).
[7] L. Malter and N. Marcuvitz, Rev. Sci. Inst. **9**, 92 (1938).
[8] R. D. Fowler, Rev. Sci. Inst. **6**, 26 (1935).
[9] R. M. Bowie, Rev. Sci. Inst. **11**, 265 (1940).
[10] R. R. Wilson, Rev. Sci. Inst. **12**, 91 (1941).

V_2 electron volts, the retarding field would prevent it from reaching the plate. When I_p, the plate current, is plotted as a function of V_2, any ordinate of the curve is proportional to the number of ions retaining energies equal to, or greater than, the corresponding voltage V_2. Energy and momentum considerations show that for the case of an argon ion colliding with a helium atom the minimum velocity retained by the ion is the fraction $(M-m)/(M+m)=0.8179$ of its initial velocity, M and m being the masses of the ion and atom, respectively. Hence the greatest percentage loss of energy that can occur at such an impact is 33.1 percent, this maximum loss corresponding to a perfectly elastic, head-on collision. Any possible inelastic phenomena such as excitation or ionization would have the effect of decreasing this maximum possible percentage loss. The maximum deflection that the ion can experience is given by: $\theta_{\max}=\sin^{-1}(m/M)$, or 5° 46′. The geometry of the electrodes was such that for the longest path lengths used (13 cm) all ions so deflected could reach the plate if they were not rejected by the analyzing field.

The graph of I_p against V_2 as it would appear under ideal experimental conditions is shown in Fig. 3 as a dashed curve. A typical actual experimental result is shown as a solid curve. The difference between the two curves may be analyzed as follows: The negative value of I_p at values of $V_2 \geqslant V_1$ could only result from the emission of secondary electrons from the grid by impact of the positive ions and by fast neutral atoms which might be present as the result of the Kallman-Rosen "Umladung" or charge exchange process. A choice of 40×40 mesh grid of 0.007-in. diameter nickel wire was made following preliminary studies on field penetration. At the large values of V_2, all ions of the beam eventually strike the grid because of its relatively negative potential, the ions passing between the meshes on the first passage being deflected back to it. All secondary electrons liberated are collected by the plate. Hence the plate current is solely due to the secondary electrons and is just equal to the correction to be made. At the other extreme of voltage when V_2 approaches zero, the ions which pass through the grid are not deflected back to it, so that only a fraction, 48/73, as many secondary electrons are liberated, the fraction being derived from the effective grid open-area to metal-area ratio. For intermediate V_2 values, the correction was taken as a linear function of V_2, varying between the secondary electron current observed at high V_2 and approximately two-thirds of this value. An assumption made in developing this correction procedure was that neutrals and ions of the same velocity were equally efficient in ejecting secondaries. Other studies[11] support this assumption.

The generous slit dimensions adopted to permit use of a 10^{-7}-ampere ribbon of ions and galvanometer measuring technique allowed some leakage of source gas into the retardation chamber despite the great speed of the pump. The effects of this argon impurity in the target gas (He) were eliminated by taking double sets of readings with and without helium present, which yielded corresponding "helium data" and "foreign gas data." The effect of a slight amount of helium leakage into the source chamber with the consequent generation of a He^+ ion beam

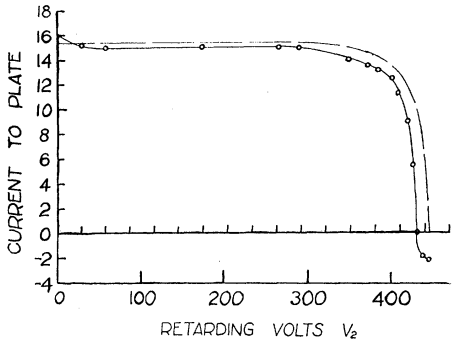

FIG. 3. Graph of the current reaching the plate P, as a function of the retarding voltage V_2. The dashed curve is an ideal one; the solid curve is experimental (helium pressure, 1.07×10^{-3} mm Hg; 430 v A^+ ions; for I_p, one division $=2.4\times10^{-9}$ amp.). The difference is attributed to the emission of secondary electrons from the grid G.

was considered negligible because of the relatively low ionization efficiency of He by electrons of optimum energy for ionizing argon.[12] For a similar reason the ion beam must have contained a negligible percentage of A^{++} ions.[13] No spectrographic investigation of beam quality was attempted.

[11] A. Rostagni, Zeits. f. Physik **88**, 69 (1934).
[12] P. T. Smith, Phys. Rev. **36**, 1293 (1930).
[13] W. Bleakney, Phys. Rev. **36**, 1303 (1930).

The following procedure was used in taking readings: Ions of energy V_1 were shot into the helium-free retardation chamber, and the retarding voltage between grid and plate was made to have values such that energy losses of p percent or greater were studied, where p was successively 0, 5, 10, 15, 20, 33, and also several values between 33 and 100. For each V_2 setting, values of the plate current were recorded as the grid-plate assembly was moved in the retardation tube so as to alter the path length from 4 to 13 cm. The current entering the can was maintained at a constant value for a given ion velocity by frequent measurement of its value as a Faraday cage current and by proper adjustment of the source-magnet current so as to increase or decrease its focusing action on the beam. Similar experiments were next performed with helium in the retardation chamber, the other variables being set at the exact values obtained when no helium was present.

TABLE I. Experimental values of σ, the cross section for specified percentage loss of energy, in units of 10^{-16} cm^2 per molecule.

Energy of ion	1000	800	600	400	200 ev
Energy loss (p% or more)					
33	3.41	3.46	4.02	4.32	4.93
25	3.599	3.747	4.411	4.900	5.635
20	3.996	4.204	4.902	5.770	7.830
15	4.42	4.72	5.49	6.86	—
10	5.08	5.57	6.53	8.67	—
5	—	6.94	—	—	—

The data obtained were analyzed in the following manner: Let I_0' represent the value of the Faraday cage current, the total entering beam strength where path length is zero, and let I_p' represent the plate current in the "foreign gas run" when no helium has been admitted to the retardation chamber. Let I_0 and I_p represent similar quantities in the experiments with helium present. Then, if sufficiently low pressure exists so that the use of single collision equations is justified, the foreign gas effect is given by: $I_p'=I_0'\exp(-\beta x)$, where β is the absorption coefficient representing chiefly the loss of A^+ ions in argon by exchange-neutralization and also by scattering. When helium is admitted to the tube, the law of the beam-intensity decrease becomes: $I_p=I_0\exp-(\beta+\alpha p)x$, where α is the cross section ($N_1\sigma$) of the atoms in 1 cm^3 of the helium at 1 mm Hg pressure for the ions of velocity determined by V_1, and for a percentage energy loss determined by V_2. Taking the logarithms of these equations and subtracting we find:

$$(\log I_p-\log I_p')+\text{constant}=-\alpha px=-N_1 px\sigma.$$

Hence the curves obtained by plotting ($\log I_p-\log I_p'$) as ordinate against x as abscissa should be straight lines and possess a slope of value $-\alpha p$, or $-N_1 p\sigma$.

RESULTS

Argon ions of energies 1000, 800, 600, 400, and 200 ev were shot through helium gas maintained at a pressure of 1.07×10^{-3} mm Hg. Observed values of I_0 and I_p for different path lengths and different V_2 were tabulated, and the values of I_p corrected for secondary electrons were calculated. A preliminary run was always made to determine the corresponding I_p' values. Curves of ($\log I_p'-\log I_p$) *versus* the path length x were then plotted. From such plots, the slopes which are proportional to the cross sections were derived. Table I gives the total cross section values derived from such graphs for the energy and loss criterion combinations of chief interest in this study. Cross sections remained constant when V_1-V_2 exceeded 33.1 percent.

INTERPRETATION OF DATA

At a single impact, an argon ion colliding with a helium atom cannot lose more than 33.1 percent of its energy. However, in no case did the slope of the ($\log I_p-\log I_p'$) *vs.* x graph, and hence σ, assume a zero value when V_2 was set for energy losses greater than 33.1 percent. The procedure for taking the readings eliminated most of the normal causes for such an apparent discrepancy in the results. Thus a possible divergence of the beam was doubly eliminated, first, because with a divergent beam a graph of

TABLE II. Values of σ_u, the neutralization cross section in units of 10^{-16} cm^2 per molecule, for A^+ ions in He.

Energy of ion	1000	800	600	400	200 ev
σ_u	3.41	3.46	4.02	4.32	4.93

($\log I_p - \log I_p'$) against x would not have been a straight line, which it was, and second, because the procedure of making the correction for foreign gas also eliminated geometric errors. Multiple collisions were eliminated as a possible cause of the apparent error because the ($\log I_p - \log I_p'$) *versus* x curves would not have been straight lines if multiple collisions had occurred to a measurable extent. Foreign gases were adequately handled by the correction procedure used. Neither foreign gases nor foreign ions would be likely to cause an effect like the one observed, namely, a constant correction independent of the retarding voltage. Finally, it is repeated that no inelastic collisions in the gas can account for the observed effect.

Only one possible explanation for the observed weakening of the beam as a function of path length but not of retarding voltage seems to remain. The effect will appear if A^+ ions are removed from the beam by charge exchange with the helium atoms, the so-called Kallman-Rosen "Umladung" process. Accepting this interpretation, the results of the present experiment provide an excellent quantitative measure of the cross section for such charge exchange. Thus the σ calculated above is composite: It should be written $\sigma = \sigma_u + \sigma_r$, where σ_u, the cross section for exchange neutralization, is the value of σ for the 33.1 percent loss criterion, and σ_r, the retardation cross section, is the difference remaining when σ_u is subtracted from the remaining table entries.

The σ_u values are listed in Table II. These values may be compared with those of 63×10^{-16} cm^2 for N_2^+ in nitrogen and 28×10^{-16} cm^2 for A^+ in argon.

The occurrence of charge exchange between an ion of argon and an atom with a very different ionization potential, like helium, has been the subject of some debate. Such measurements as existed indicated that the cross section must certainly be very small. In collisions between ions and atoms of different elements, two possibilities arise: The ionization potential of the projectile may exceed that of the target atom, or vice versa. Quantum mechanical considerations by Morse and Stueckelberg[14] lead to the prediction that there is a certain likelihood of electron transfer in both cases which reaches a maximum when the ionization potentials of the two atoms are the same. Their results also indicate that the probability of transfer of an electron is an even function of the difference of ionization potentials provided the kinetic energy of the ion relative to the atom is large, the same therefore for an A^+ ion meeting a He atom as for a He^+ ion meeting an A atom. The present experiment provides a most promising means for investigating this question further.

TABLE III. Values of σ_r for A^+ ions in He, the cross section for specific percentage loss of energy, in units of 10^{-16} cm^2 per molecule.

Energy of ion	1000	800	600	400	200 ev
Energy loss in percent					
33	0	0	0	0	0
25	0.189	0.287	0.391	0.580	0.725
20	0.586	0.744	0.900	1.45	—
15	1.01	1.26	1.47	2.54	—
10	1.67	2.11	2.51	4.35	—
5	—	3.58	—	—	—

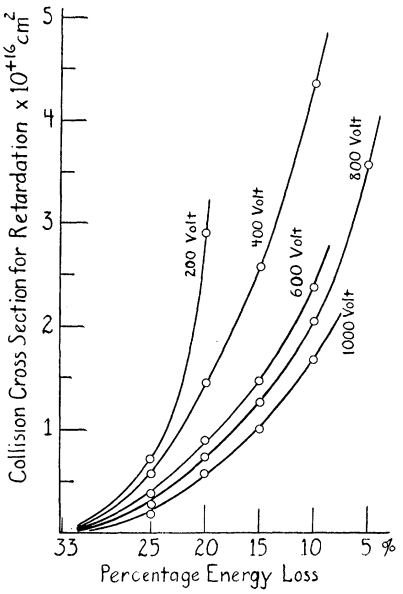

FIG. 4. Collision cross section for energy loss by retardation plotted against percentage energy loss for A^+ ions of various speeds passing through He.

The σ_r values, the cross sections for energy losses at single encounters, are listed in Table III and plotted *versus* percentage energy loss in Fig. 4. The ordinate of any curve gives the

[14] P. Morse and E. Stueckelberg, Ann. d. Physik **9**, 589 (1931).

cross section within which the ion must pass if it is to incur at least the percentage energy loss indicated as abscissa. It is believed that the curves will lend themselves in a very satisfactory manner to analysis of the forces existing between the particles.

The author wishes to acknowledge gratefully the guidance and valuable suggestions of Doctor Robert N. Varney who directed this problem by correspondence from the Naval Proving Ground at Dahlgren, Virginia; also the courtesies extended by Professor A. L. Hughes, Professor G. E. M. Jauncey, and Doctor Joseph Keller of this department.

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Diffusion Rates of Carbon in Iron-Molybdenum and Iron-Tungsten Alloys

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Measurements are reported of the influence of tungsten and molybdenum on the diffusion rate of carbon in face-centered iron at 1000°C. Both of these elements slow down the diffusion of carbon, the influence of tungsten being more than twice that of molybdenum. These results are compared with previous measurements on iron-cobalt alloys and other known data. No connection seems to exist between the variation in lattice parameter of face-centered iron and the variation in diffusion rate of carbon.

IN a previous paper[1] a strong accelerating influence of cobalt on the diffusion rate of carbon in face-centered iron was reported.* In continuation of this study, experiments were made with iron-molybdenum and iron-tungsten alloys.

There are relatively few binary systems in which measurements of this type can be made conveniently. The main condition is the existence of a sufficiently wide range of homogeneous face-centered solid solution. It should be possible to vary the concentration of carbon and of the alloying element, within relatively wide limits, to suit the accuracy of the measurements. Both in tungsten-iron and in molybdenum-iron alloys no single phase region with face-centered lattice exists beyond a few percent of the added element. Carbon, however, increases this range somewhat. Accordingly, experiments were made with the following alloys; the tungsten pair: 1.1 atomic percent (about 3.5 wt. percent) tungsten with 2.6 atomic percent (0.58 wt. percent) carbon and with 0.22 atomic percent (0.05 wt. percent) carbon; the molybdenum pair: 1.1 atomic percent (about 1.9 wt. percent) molybdenum with 3.1 atomic percent (0.7 wt. percent) carbon and without carbon, and finally a pair of alloys with the same molybdenum content but with 4.2 atomic percent (about 0.9 wt. percent) carbon, and without carbon. The first two groups of alloys, the tungsten alloys and the molybdenum alloys with lower carbon content, fall within the region of homogeneous face-centered lattice at 1000°C.[2] The third group, the molybdenum alloys with higher carbon, fall into the region where at 1000°C some of the carbides do not dissolve. These alloys were included in the experiments in order to see to what extent the presence of carbides affects the observed diffusion.

The experimental method was essentially the same as that used in the previous investigation. The welded samples were heated for three days at 1000°C and the concentration of carbon in layers parallel to the interface was determined.

[1] R. Smoluchowski, Phys. Rev. **62**, 539 (1942).

* *Note added in proof:* Mr. Malcolm F. Hawkes and Dr. Robert F. Mehl of the Carnegie Institute of Technology kindly informed me that their experiments do not show this strong accelerating influence of cobalt. This point needs thus further experimental clarification in particular in view of reference 6 quoted in reference 1 of this paper.

[2] S. Takeda, Tech. Rep. Sendai **9**, 483 (1930); **9**, 627 (1930); and **10**, 42 (1931). E. C. Bain, *The Alloying Elements in Steel* (Am. Soc. Metals, Cleveland, Ohio, 1939).

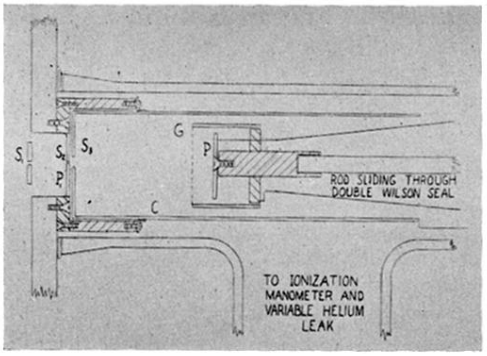

FIG. 2. Details of retardation chamber electrode assembly.