

crystals, and which one can calculate using the self-consistent field method for atoms, molecules, and solids, show a good consistency with each other. Such comparison as we have made here, between the one-electron energies of the atoms and of the other systems, could well be made in the course of future calculations of molecular wave functions or energy bands. In particular, it is interesting to observe the great increase in the width of the bands as we go through the first two

periods, adding more electrons and reducing interatomic distances. A particularly interesting thing will be to find just how the $3d$ level drops below the valence band, in the elements to the neighborhood of Ge. Interesting deductions regarding molecular binding are of course suggested by the drop of the average value of the one-electron energy in a band, below the atomic levels, but we shall not try to go further into the implications of the results in the present paper.

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Differential Cross-Section Measurements for Large-Angle Collisions of Helium, Neon, and Argon Ions with Argon Atoms at Energies to 100 keV*

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The differential cross section for the scattering of positive charge from collisions of noble gas ions with the target gas argon has been measured. The ion beam is passed through a chamber containing the target gas at a pressure of a few microns of mercury, which is low enough to insure that single collisions will predominate. The particles scattered out of the beam are collected and the total positive charge is measured. The collector incorporates collimating holes to select only those particles at a chosen scattering angle with a resolution of about two degrees. The apparatus allows the angle to be varied continuously up to thirty-eight degrees. Data are presented for collisions of singly ionized helium, neon, and argon ions with argon atoms at 25, 50, and 100 keV. The cross sections observed differ from those expected from the Rutherford scattering law and these differences are interpreted in terms of electron screening, ionization, and charge exchange.

1. INTRODUCTION

THE large-angle collisions studied here are those in which ions of helium, neon, and argon with an energy from 25 000 to 100 000 electron volts collide with stationary argon atoms. These collisions can be expected to follow the Rutherford scattering law in the limit of sufficiently high energies, but at the lower energies there will be a modification due to the screening effect of the electrons.

In this energy range the vast majority of the collisions will be small-angle and, in fact, these small-angle collisions account for nearly all of the total cross section. There have been numerous studies of the total cross sections for elastic scattering, charge exchange, and ionization in this energy range.¹⁻³

Although the few large-angle collisions which do occur account for a nearly negligible portion of the total cross section, they are interesting, for these are collisions in which the atoms come very close together.

Their study gives information regarding the potential energy function between two atoms at very close distances on an atomic (not nuclear) scale. Under some conditions in these experiments, the actual distance of closest approach is less than the radius of the innermost classical electron orbits of either of the colliding atoms. This means that the electron configuration during the collision will be complicated.

In this energy range, the relative velocity of the ion and atom in the collision is of the same order of magnitude as the classical orbital velocity of the atomic electrons. This is the condition under which ionization and charge exchange cross sections are thought to be a maximum.¹

The scattered particle current into a given solid-angle segment depends on the forces between the atoms during the collision, but the scattered positive charge measured in the experiments described here depends, in addition, on the extent to which the scattered particle was ionized or neutralized during the collision. Specifically, we are measuring in these experiments the differential cross section for scattering of positive charge.

Heydenburg, Hafstad, and Tuve⁴ have performed differential cross section measurements for collisions of protons with protons at 600 to 900 keV. Their apparatus is similar in some respects to that to be described here.

⁴ Heydenburg, Hafstad, and Tuve, *Phys. Rev.* **56**, 1078 (1939).

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¹ H. S. W. Massey and E. H. S. Burhop, *Electronic and Ionic Impact Phenomena* (Oxford University Press, London, 1952). Chapter VIII contains an excellent discussion and bibliography of this field.

² Niels Bohr, *Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd.* **18**, 8 (1948).

³ S. K. Allison and S. D. Warshaw, *Revs. Modern Phys.* **25**, 779 (1953).

Ramsauer and Kollath⁵ have made measurements at large angles of the scattering of protons at energies up to 120 electron volts from several gas targets, and Rouse⁶ has made measurements of the large angle scattering of potassium ions from several gases at energies to 360 electron volts. However, there have been, to our knowledge, no previous measurements of large-angle single scattering of ions in the particular energy range of 25 to 100 kev.

2. THEORY OF THE MEASUREMENT

A study of differential cross sections for single collisions requires a suitable thin gas target. This can be realized with an apparatus as shown in Fig. 1. The ion beam enters the chamber, which contains the target gas, through the small defining hole *a*. At various points along the beam path, there are a few ions scattered out of the beam. If two holes, *b* and *c*, are aligned as shown, a small fraction of the scattered ions can pass through them and enter a charge collector. The observed current to this collector is proportional to the differential cross section for scattering of positive charge at the angle in question. Lines drawn from the edges of holes *b* and *c* intersect the ion beam and define an effective target volume.

The target gas pressure should be low enough so that the mean free path for elastic scattering, charge exchange, or ionization is at least several centimeters. This is more than adequate to guarantee that multiple large angle collisions within the small target volume are extremely rare, and therefore the scattered beam will be the result of single collisions. This mean free path is also long enough to insure that most of the ions in the beam make no collisions of any sort within the scattering chamber, and that most of those few ions scattered at large angles from the target volume make no further collisions before leaving the chamber through hole *b*. The test of these statements lies in a measurement of the scattered current at a given angle as a function of the target gas pressure. When this current is found to be proportional to the pressure, the pressure is low

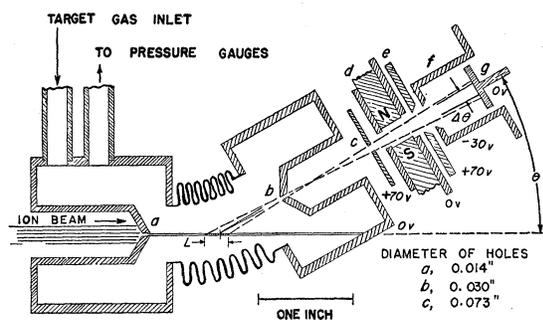


FIG. 1. The scattering apparatus.

⁵ von C. Ramsauer and R. Kollath, *Ann. Physik* **16**, 570 (1933).

⁶ A. G. Rouse, *Phys. Rev.* **52**, 1238 (1937).

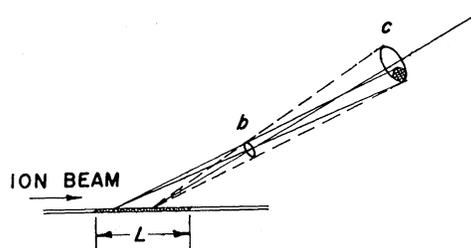


FIG. 2. The solid angle subtended by the charge collector as seen from an arbitrary point in the effective target volume.

enough to insure the aforementioned conditions. In practice, the target gas pressures used in these experiments were of the order of a micron of mercury.

There can be no foils across holes *a* and *b* at the ion energies used in these experiments. The ratio of the target gas pressure to the pressure in the accelerator is about 200 to 1, and this pressure difference is easily maintained across the two small holes by the accelerator's vacuum system.

The ratio of the scattered current *I* to the incident ion current *N* at the angle θ is

$$I/N = n\sigma(\theta)L\Delta\Omega, \quad (1)$$

where *n* is the number of target particles per unit volume, $\sigma(\theta)$ is the differential cross section for the process in question averaged over the effective solid angle of acceptance $\Delta\Omega$, and *L* is the length of the target volume measured in the direction of the ion beam.

The value of $L\Delta\Omega$ depends on the location and size of the defining holes. In the apparatus of Fig. 1, holes *b* and *c* have diameters s_1 and s_2 which are proportional to their respective distances y_1 and y_2 from the center of the target volume. This defines a cone, as shown in Fig. 2, whose interior angle is

$$\Delta\theta = s_1/y_1 = s_2/y_2. \quad (2)$$

Introducing a new length y_0 defined by

$$y_0 = y_1 y_2 / (y_2 - y_1), \quad (3)$$

the distance *L* is found from the geometry of Fig. 1 to be

$$L = 2y_0(\Delta\theta) \csc\theta. \quad (4)$$

An integration must now be performed along length *L* to find the average solid angle. At the center of the region the solid angle is evidently $\pi(\Delta\theta)^2/4$, and it is zero at each end. At intermediate points the solid angle intersects an area shaped something like the small shaded area in Fig. 2. This integration results in an effective solid angle

$$\Delta\Omega = (\Delta\theta)^2/3. \quad (5)$$

Combining all the aforementioned equations and solving for $\sigma(\theta)$, there results

$$\sigma(\theta) = (3I/N)/[2ny_0(\Delta\theta)^3 \csc\theta]. \quad (6)$$

A convenient dimensionless way to plot the measured cross sections is to show $\sigma(\theta)/b^2$ as a function of θ . Here b is a characteristic length defined by

$$b = Z_1 Z_2 e^2 (1 + \gamma) / U. \quad (7)$$

In this formula Z_1 and Z_2 are the atomic numbers of the incident particle and the target particle respectively, e is the electronic charge, γ is the ratio of the mass of the incident particle to that of the target particle, and U is the kinetic energy of the incident particle in the laboratory coordinate system. In classical Rutherford scattering theory, b is called the collision diameter and is the distance of closest approach in a "head on" collision.

3. DESCRIPTION OF THE APPARATUS

The University of Connecticut heavy-ion accelerator⁷ used in this experiment is of the Cockcroft-Walton type. It produces a focused monoenergetic beam of positive ions at energies continuously variable from 25 to 250 kev. The radio-frequency ion source⁸ furnishes 10 to 100 microamperes of singly charged helium, neon, or argon atoms. The ion beam is analyzed by a deflection magnet so that the charge-to-mass ratio of the ions striking the gas target is known. Three sets of electrostatic deflection plates together with the analyzing magnet may be used to line up the beam properly with the axis of the scattering chamber.

The scattering chamber is itself inside a large vacuum box. As shown in Fig. 1, it consists of two parts bridged by a flexible bellows. The first part is rigidly fixed. It contains the hole a , which defines the incoming ion beam, and the connections to the target gas handling system. The flexible bellows allows the second part to rotate through any angle of scattering up to 38° about the center of the effective target volume. This part rotates on a shaft which goes through the vacuum wall by means of an "O"-ring seal. The second part supports the charge collection system and the electrometer.

In the charge collection system, holes b and c define the scattered beam and determine the angular resolution. All other holes are oversize and do not limit the scattered current. The ion current is collected on electrode g which is connected to the electrometer input. Electrode f is maintained 30 volts negative as this was found to be a sufficient potential to prevent secondary electrons from leaving the collector.

At certain angular settings some difficulty was experienced with a background electron current which partially masked the positive current to the collector. This was eliminated by two magnetic fields, the one from a small permanent magnet of 500 gauss behind hole d , and the other from a large Helmholtz coil which immersed the entire scattering apparatus and electrometer in a uniform 10-gauss field. These magnetic

fields and potentials have no appreciable effect on the scattered ions, which have a good fraction of the incident energy at these angles. With these extraneous electron currents removed, the observed scattered currents were always positive and were negligibly small when the target gas was removed.

The electrometer circuit is mounted in the vacuum just behind the collector electrode. Various combinations of input resistors and galvanometer shunts give this a useful range of 10^{-6} to 10^{-16} ampere. On the most sensitive current scale the uncertainty was 5×10^{-17} ampere.

The target gas handling system is shown in Fig. 3. The gas leaks from a low-pressure reservoir, passes through a liquid air trap, and enters the scattering chamber. An oil diffusion pump competes with the leak and this shortens the time required to reach equilibrium pressure. A separate closed line leads from the scattering chamber to the pressure measuring gauges. There is no flow of gas in this line and the pressures, which were measured by the McLeod gauge, are therefore the pressures at the scattering chamber.

4. DATA

The value of I/N was found by dividing the current observed at a given angle θ by the main beam current. The value of N was obtained by setting the collector to zero angle and reading the current. The current I at the angle θ would then be measured several times and averaged. Then the apparatus would be returned to zero angle and the beam current N recorded again. If more than a 10 percent change had occurred in N ,

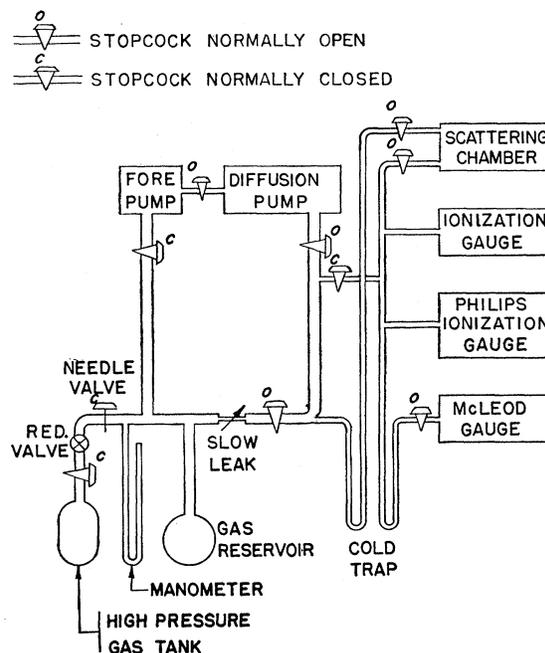


FIG. 3. The target gas handling system.

⁷ Everhart, Carbone, and Stone, Technical Report No. 1 to Office of Ordnance Research, January 15, 1954 (unpublished).

⁸ Moak, Reese, and Good, *Nucleonics* **9**, 18 (1951).

the point would be repeated. This process was followed for each point or pair of points. The values of N were in the range of 10^{-7} amperes and I was between 10^{-10} and 10^{-16} ampere for most of the points.

The measurements were made on each side of zero angle up to 38° on one side and to 20° on the other. The values of I/N were plotted against θ , and the zero for θ was taken as the angle about which the curves measured on either side of the center were symmetrical. The angular resolution is approximately $\Delta\theta$ which was 0.04 radian, or 2.3° . The lengths y_1 and y_2 were 1.90 and 4.67 cm respectively, so that the geometrical factor $y_0(\Delta\theta)^2$, calculated using Eq. (3), was 2.06×10^{-4} cm.

A measurement of pressure and of temperature allowed the target gas concentration n to be calculated. The pressures were in the range of 0.5 to 3 microns of mercury, measured to an accuracy of 5 percent. The target gas was analyzed with a mass spectrograph and found to be better than 99.9 percent pure argon.

The values of the cross sections, calculated from the data by using Eqs. (6) and (7), are plotted in Fig. 4 for collisions of singly ionized helium ions with stationary argon atoms at energies of 25, 50, and 100 kev. Figures 5 and 6 are the same except that the incident particles are singly ionized neon ions and argon ions respectively. The cross sections calculated from the data in this way should be independent of target gas pressure if this pressure is low enough. The extent to which this is true is seen on the figures. Each set of points represents data taken at two different pressures as given in the caption.

The differential cross section plotted for each point is the average cross section over the two degree angular resolution. The over-all accuracy assigned to the cross sections is ± 30 percent. The curves are so steep that

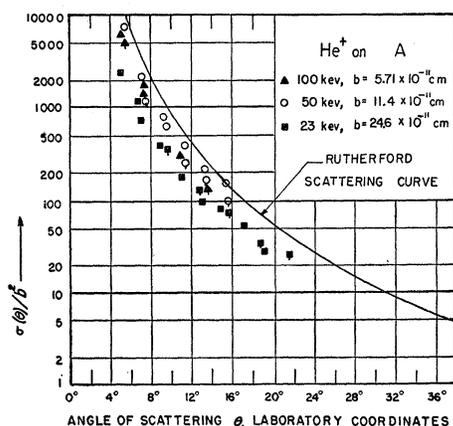


FIG. 4. Differential cross-section measurements for scattering of positive charge from single collisions of He^+ ions with argon atoms. The ratio of this cross section to the square of the collision diameter is plotted. Each set of points represents data taken at two pressures, the points for the higher pressure being indicated by a small stem on the symbol. In microns of mercury, the pressures were 2.65 and 0.89 at 100 kev, 2.06 and 0.69 at 50 kev, and 2.65 and 0.89 at 25 kev.

it would be difficult to assign a smaller uncertainty unless the angular resolution were made smaller.

5. DISCUSSION

If the scattering were due only to the Coulomb repulsion between the two colliding nuclei, and if there were no change in charge during the collision, all measured curves of $\sigma(\theta)/b^2$ would lie on top of one another, regardless of the energy. In the center-of-mass coordinate system the theoretical value for this is given by the Rutherford scattering formula $\sigma_0(\theta_0)/b^2 = \frac{1}{16} \times \csc^4(\theta_0/2)$, but this must be transformed to the laboratory coordinate system⁹ by using the appropriate value of the mass ratio γ . These Rutherford curves have been plotted for comparison on each of the figures.

In Figs. 4, 5, and 6, the curves taken at lower energies generally lie below those taken at higher energies. This effect is most pronounced in the case of argon ions incident on argon atoms and least pronounced in the case of helium ions incident on argon. This departure from Rutherford scattering is interpreted as being due to the electron screening, which should become important when the characteristic length b becomes comparable with atomic dimensions.² As seen by inspection of Eq. (7), this length becomes larger at the lower energies and higher atomic numbers. Thus for 100-kev helium ions incident on argon, the calculated value of b is 0.00571×10^{-8} cm, which is much smaller than the dimensions of either atom. At the opposite extreme, for 25-kev argon ions incident on argon, the value of b is 0.373×10^{-8} cm, a size which is comparable with the dimensions of these atoms.

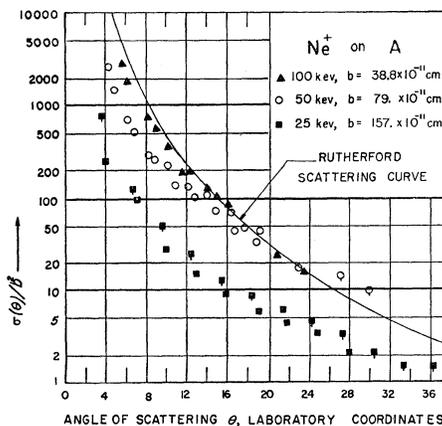


FIG. 5. Differential cross-section measurements for scattering of positive charge from single collisions of Ne^+ ions with argon atoms. The ratio of this cross section to the square of the collision diameter is plotted. Each set of points represents data taken at two pressures, the points for the higher pressure being indicated by a small stem on the symbol. In microns of mercury, the pressures were 2.15 and 0.53 at 100 kev, 2.65 and 0.53 at 50 kev, and 2.10 and 0.61 at 25 kev.

⁹ See, for example, L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1949), Chap. V, Sec. 18.

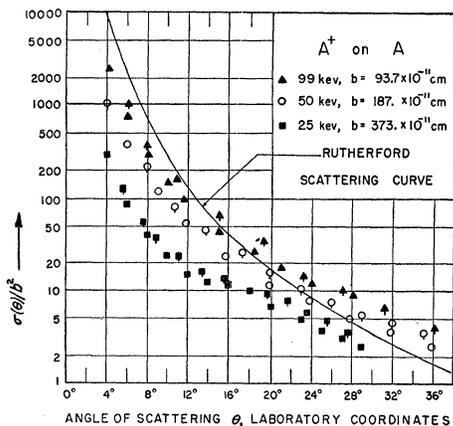


FIG. 6. Differential cross-section measurements for scattering of positive charge from single collisions of A^+ ions with argon atoms. The ratio of this cross section to the square of the collision diameter is plotted. Each set of points represents data taken at two pressures, the points for the higher pressure being indicated by a small stem on the symbol. In microns of mercury, the pressures were 2.94 and 0.67 at 99 keV, 2.64 and 0.69 at 50 keV, and 2.60 and 0.69 at 25 keV.

The points in Fig. 4 for the 100-keV helium ions incident on argon lie below the Rutherford curve by a factor of about two. As we have seen above, this cannot be accounted for by electron screening in this case. Although this could be explained by an unsuspected systematic error of a factor of two in our measurements, it is more likely to be accounted for by charge exchange during the collision process. An effect of this magnitude would be observed if about half of the scattered helium atoms were neutral.¹⁰

There is the criterion, mentioned in Sec. 1, that ionization and charge exchange are most likely when the velocity of the incident ion is comparable with the classical orbital electron velocities.¹ An elementary calculation shows that this is fulfilled in the energy range of these experiments. In Fig. 5, the measured cross section for 50-keV neon ions scattered from argon at large angles is higher than one would expect from the form of the other curves. Possibly there is a maximum in the ionization probability at that energy. The cross section shown in Fig. 6 for scattering charge from the collisions of argon atoms at 100 keV is higher than the Rutherford curve at large angles. This indicates that the average scattered particle is more than singly charged for these particular collisions.

¹⁰ E. Snitzer [Phys. Rev. 89, 1237 (1953)] has indeed observed that a 100-keV helium beam in argon is 53 percent neutral after multiple collisions at zero angle. This may be compared with our data which indicates that a similar ratio may hold for single collisions at large angles.

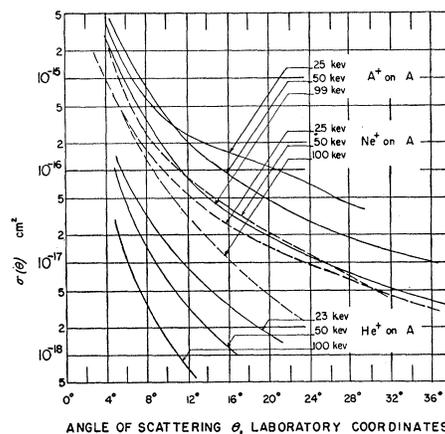


FIG. 7. Differential cross sections in square centimeters for scattering of positive charge from large-angle collisions.

The scattering apparatus described here would also measure the current due to the recoil target particle if this particle were ionized by the collision. Most of the recoil particles are scattered with low energies at 90° , but there are a few scattered at smaller angles with considerable energy.¹ For Rutherford scattering, the ratio of the cross section for the recoil target particle to that for the scattered incident particle¹¹ is less than 0.03 for angles less than 30° . This ratio increases until it is unity at about 45° , and as the 90° angle is approached, practically all the current is due to the recoil particles. In our apparatus the maximum angle is 38° , at which angle a small fraction of the current may be due to the recoil particles.

The curves of $\sigma(\theta)/b^2$ do not show easily how the absolute cross section depends on the energy or the kind of ion since the value of b is different for each curve. To show this, the differential cross sections in square centimeters have been plotted in Fig. 7. The scatter of the individual points is not shown, since this was given in the preceding figures. The curves shown are smooth lines drawn through the data. The cross sections for the helium collisions vary about as the inverse square of the energy and as the inverse fourth power of the angle, as would be expected since these data follow the Rutherford law approximately. The other curves are not as regular, although it is seen that generally the cross sections are largest for the heavier ions at the lower energies. The curve for 50-keV neon seems to lie too high and this exception to the rule might be explained, as above, in terms of the dependence of ionization probability on energy.

¹¹ Everhart, Stone, and Carbone, Technical Report No. 2 to Office of Ordnance Research, April 20, 1954 (unpublished). This is readily calculated from the Rutherford formulas for the scattered and recoil cross sections. See reference 9 also.