

Energy Distribution of Electrons from Ionizing Collisions of Heavy Particles

H. W. BERRY

Syracuse University, Syracuse, New York

(Received October 17, 1960)

The energy distribution of electrons produced in ionizing collisions of ions and fast atoms with atoms of the parent gas at rest has been measured for A and He. The energy of the incident particles ranged from 0.30 to 3.0 kev. Electrons released at 90° with the incident ion or atom beam were analyzed in an electrostatic energy selector which consisted of 90° segments of coaxial cylinders. The use of a fine wire grid in the region in which the ionization occurred allowed a positive differentiation to be made between the ionization electrons and the secondary electrons from any metal surfaces. The electron distributions for argon ions and atoms in argon are similar and show a rapid decrease in the yield as the energy increases from zero to about 4 ev followed by a plateau and a flat maximum near 12 ev. The distributions for He also show the initial rapid decrease continuing to a small maximum which for the ion is at about 31 ev and for the neutral atom at about 16 ev. These distributions are almost collision energy independent, though with an increase in the over-all yield with increasing energy.

INTRODUCTION

SLOW, inelastic collisions of ions and atoms are believed to involve the formation of a temporary molecule in which energy of relative motion is transferred to that of excitation and ionization. The general theoretical method of treating the problem was developed by Stueckelberg and others¹; but it has not been applied to any particular atomic species though some general results have been obtained by Bates and Massey.² In this approach a transition may occur, if the initial and final potential energy curves of the quasi-molecule cross or nearly cross at some internuclear separation. As Bates and Massey point out, however, the lack of knowledge of the potential curves and of the prevalence of curve crossing makes it difficult to estimate even order-of-magnitude effects. For the case of ionization, the final state is a continuum corresponding to the energy given to the ejected electron. It might therefore be expected that the energy spectrum of the ionization electrons would be characteristic of the atomic species and the quasi-molecule involved in the collision.

Few experimental investigations of the electron energy distributions in ionizing collisions have been made. In one such measurement,³ for collisions of protons in H₂, A, N₂, and Kr, the distributions show the expected decreasing yield at first but with a slow increase above a hundred or so electron volts. Also, Moe and Petsch⁴ recently measured the energy spectrum of electrons from ionizing collisions of K⁺ in A, Ne, and Kr. In these, they found maxima in the distributions that seemed to be characteristic of the particles involved.

In the investigation reported here, the electron energy distribution has been measured for ionizing col-

lisions of helium ions and neutral atoms with helium gas and argon ions and neutral atoms in argon gas, in which the energy of the incident particle ranged from several hundred to several thousand electron volts. In the following sections, the apparatus and method of measurement are described, with finally a discussion of the results obtained.

EXPERIMENTAL METHOD

The experiment consisted of (1) producing a beam of the desired ion specie and energy, (2) neutralizing, if desired, the ions of the beam by charge exchange with atoms of the parent gas, (3) allowing this beam to traverse a region containing the target gas where the inelastic collisions will occur, and finally (4) measuring the energy distribution of the electrons produced in the inelastic collisions. A schematic arrangement of the apparatus for this is shown in Fig. 1. The vacuum envelope was composed of sections of four-inch Pyrex glass pipe, with the apparatus supported from the brass plates mounted at the ends of these sections. The system was evacuated by a fractionating, three-stage oil diffusion pump. Pressures less than 5×10^{-6} mm of Hg were customary before the admission of gases into the source and ionization regions.

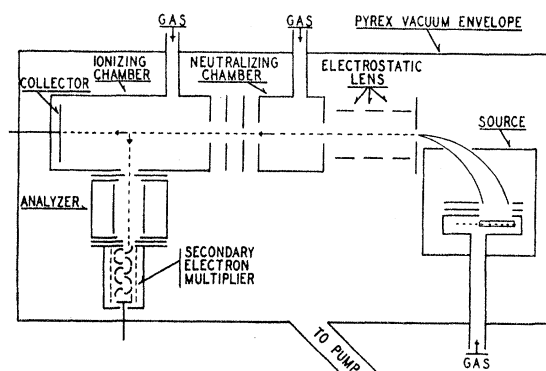


FIG. 1. Schematic diagram of the apparatus.

* This research was supported in whole or in part by the U. S. Air Force, monitored by the Aeronautical Research Laboratories, Wright Air Development Division.

¹ E. C. Stueckelberg, *Helv. Phys. Acta* **5**, 370 (1932).

² C. Zener, *Proc. Roy. Soc. (London)* **A137**, 696 (1932).

³ D. R. Bates and H. S. W. Massey, *Phil. Mag.* **45**, 111 (1954).

⁴ E. Blauth, *Z. Physik* **147**, 228 (1957).

⁵ D. Moe, and E. Petsch, *Phys. Rev.* **110**, 1358 (1958).

The ion source used in this study was adapted from that designed by Beckman.⁵ It is mounted between the poles of an electromagnet so that the magnetic field is oriented normal to the plane of Fig. 1. This field collimated the electrons producing the initial ionization, provided a low-resolution mass spectrograph, and effected an increase in the ion beam density by a novel focussing action. Gas introduced into the source was ionized by the bombardment of electrons emitted from filaments above and below the ionization chamber. The electrons traversed the region in a direction parallel to the magnetic field and thus were prevented by the field from readily going to the walls. Consequently, many traversals through the chamber should be made with a resulting increase in the number of ions produced. Pole shoes mounted in the vacuum system were notched where the filaments were mounted and served also as reflectors to reverse the electron motion. It was found adequate to maintain the pole shoes at the potential of the negative end of the filaments. The source ionization chamber was made of copper with a tantalum insert to form the ion exit slit. The length of this slit was $1\frac{3}{4}$ in. with the electron entrance slits on the top and bottom slightly longer. Two tantalum accelerating electrodes were mounted as shown in front of the source exit slit with 0.150-in. separation between the source chamber and the first grid as well as between the grids. The second grid served to focus the beam in the vertical direction (along the magnetic field) and seemed to do this best when operated at a potential near that of the copper ionization chamber. The first grid was most effective when near the total accelerating voltage. An electric field free brass box completed the source and was operated at the potential desired for the total acceleration.

To produce the "focussing" action on the beam, the pole shoes were tapered to decrease the magnetic field strength in such a way that ideally all trajectories are tangent to one line on emergence of the beam from the field. To do this the field must decrease to zero at this line so that ions from the right side of the source undergo a 90° turn over a longer trajectory than those from the left side. Several sets of pole shoes with different cross sections were tried and for each the magnetic field was measured in the azimuthal plane with a small search coil. Graphical construction of the trajectories showed in all cases that the field did not drop off fast enough to insure proper focussing. The excess of field had the effect of overbending the outermost rays or, if this was compensated for by adjusting the accelerating potential difference, then the innermost trajectories were not turned enough.

Following the source the ion beams passed through a three element electrostatic lens and from there into a neutralizing chamber where the beam may be partially neutralized. On removing the remaining ions, a neutral

beam of the same energy as the ion beam resulted. The lens consisted of three elements with the first and third elements at the same potential. The second and third elements were split parallel to the axis of the system so that in one case a small potential difference between the halves would move the beam vertically and in the other case—horizontally. This has decided advantages in that the exact angle of emergence of the ion beam from the source is not known and further it removes the necessity for very careful alignment. The sources of potential for the lens and the source were so arranged that an additional acceleration could be introduced between the source exit slit and the first element of the lens to change the beam energy without adjusting the source magnetic field.

The neutralizing chamber consisted simply of an enclosed region in which a gas may be confined and of several electrodes for applying a field sufficient to remove any unneutralized ions from the beam. While the neutral atom beam was not measured, it is estimated that this was the order of $\frac{1}{5}$ to $\frac{1}{10}$ of the incident ion flux.

The chamber in which the ionizing collisions occurred was a cylinder $\frac{7}{8}$ -inch inside diameter and $2\frac{1}{4}$ inches long. One side was milled off and fastened to a flat plate which contained the exit slit for the electrons. The slit, $\frac{1}{8}$ inch by $\frac{1}{2}$ inch, was beveled on the inside to conform to the cylindrical symmetry. Concentric with the cylinder was a fine wire grid with the wires mounted parallel to the axis of the cylinder. Twenty-four 0.001-inch tungsten wires were evenly spaced around a circle of $\frac{1}{2}$ -inch diameter. The grid was supported by lavite rings mounted within the ionization cylinder. These rings placed at the ends of the cylinder also served as the mountings for the entrance and exit apertures. The entrance aperture was $\frac{3}{32}$ inch in diameter with a $\frac{5}{16}$ -inch aperture following; the latter was held at ten or so volts negative with respect to the entrance aperture to suppress electron emission generated by ion bombardment of the aperture edges. The grid was maintained at the same potential as this second entrance aperture. At the opposite end of the cylinder, there was a Faraday cage for the beam current measurement with an aperture preceding to again suppress electron emission.

Electrons ejected at about 90 degrees to the beam, which emerge from the cylindrical chamber entered an energy analyzer which is indicated only schematically in Fig. 1. The analyzer electrodes are 90-degree sections of concentric cylinders and attain the same focussing action as the 127-degree electrostatic analyzer by placing the exit and entrance image foci 0.35 times the mean radius from the ends of the deflecting electrodes.⁶ A 2-mm entrance slit restricted the angular spread of the entering electrons while a 1.1 slit at the exit focus was used for all the measurements. For this analyzer, as with the 180-degree magnetic type, the exit slit width, in terms of the energy range passed, is proportional to

⁵ L. Beckman, *Arkiv Fysik* 8, 451 (1954).

⁶ M. G. Ingraham, *Advances in Electronics*, edited by L. Marton (Academic Press, Inc., New York, 1948), Vol. I, p. 219.

the mean particle energy. The correction for this tends to magnify the experimental errors excessively at low energies. By use of an accelerating potential difference between the source of the electrons and the analyzer, the effect of this correction can be much reduced. Further, since it is desired to maintain the grid negative relative the cylinder, this potential difference can be part of that applied between the grid and analyzer. For most of the data taken, about 90% of the total potential difference through which the electrons were accelerated was applied between the grid and cylinder. This could be readily varied, and data were taken with other divisions with no effect on the observed distribution.

A test of the analyzer resolution was made by inserting a small, V-shaped filament of 0.002-inch tungsten wire in the center of the ionization cylinder. Electrons emitted from this were then accelerated with the same potential arrangement used in the distribution measurements. For a total drop of 21.5 volts, the distribution peak at half height was 1.0 volts. This is some 50% larger than the calculated width in which no cognizance is taken of the source size, angular spread of the electron beam, or source energy spread.

To insure proper operation of the analyzer and prevent distortion of the energy distribution, the residual magnetic field in the analyzer region was annulled with three sets of Helmholtz coils with the axis of each pair mutually perpendicular to the others. Since the leakage field from the source magnet tended to cancel the earth's field in the region of the analyzer, the residual field was only a few tenths of a gauss. This could be readily annulled by use of a sensitive dip needle and compass with an uncertainty of about 0.005 gauss.

It was more feasible to measure the energy distribution of the electrons with a constant potential difference for the acceleration of the electrons and so a varying deflection potential difference on the analyzer, rather than the reverse. This method allows the velocity component perpendicular to the plane of symmetry of the analyzer to vary as the transit time of the electrons in the analyzer changes. The range of variation is not large, however. With the analyzer dimensions used, the energy range associated with the perpendicular component of the velocity would be 0.4 to 1.0 ev, while the self-energy of the electrons varied from zero to 30 ev.

The electron current passed by the analyzer was amplified by a secondary electron multiplier then fed into a micromicroammeter and finally to a chart recorder. The ten-stage multiplier was a commercial unit with an original gain of the order of 10^6 ; but on exposure to air this gain dropped to about 5000. Background noise limited the measurements to currents greater than 2×10^{-15} ampere. The analyzer deflection potential difference was applied through a synchronous motor driven potentiometer with the distribution swept out in about two minutes. Examination of the sweep voltage

on the recording chart showed a linearity of better than 5%.

MEASUREMENTS

A major difficulty in these measurements arises from the electrons ejected from the surfaces of the ionization cylinder by ion and atom bombardment. For gas covered surfaces this can be as large as one electron per incident atom or ion with a kinetic energy of about one kev.⁷ Consequently this can easily mask the much smaller yield of electrons from the ionizing collisions with the gas molecules. For ions with particularly large ionization energies such as He^+ or A^{++} , the secondary emission coefficient can be large even for little or no kinetic energy.⁸ In general, the energy range of these secondary electrons is about that expected for the ionization electrons. Consequently, it becomes necessary to suppress the emission or in some way differentiate it from the ionization yield.

This is the prime function of the grid placed in the ionization cylinder. With the grid some twenty volts more negative than the cylinder, most of the electrons ejected from the cylinder would be turned back. Those produced, however, in the neighborhood of the slit would be turned into the slit so as to enter the analyzer, but would originate at a potential much different from that of the grid and the region within and so would be observed as a separate group at a lower total energy as shown in Fig. 2 as the peak on the left. Measurements^{7,8} of the energy distribution of electrons ejected from both clean and gas covered surfaces by both atoms and ions indicated that few electrons would have energies in excess of 15 ev. This seems to be borne out by the measurements here, for the contribution from the cylinder walls seems to have levelled off before the onset of the peak of the ionization electrons.

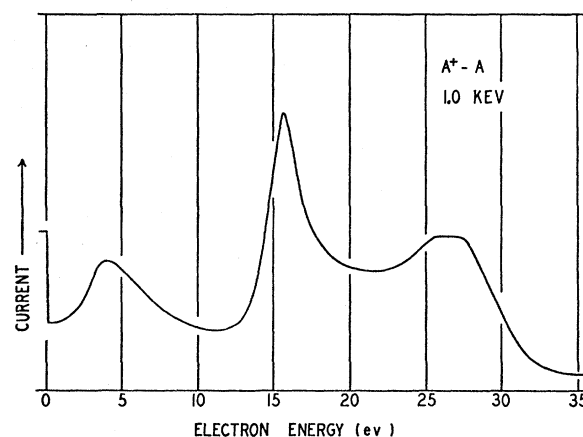


FIG. 2. Sample data record for 1.0-kev A^+ in A. The analyzer deflection voltage increases from left to right. The small peak on the left represents secondary electrons from the cylinder walls.

⁷ H. W. Berry, J. Appl. Phys. **29**, 1219 (1958).

⁸ H. D. Hagstrum, Phys. Rev. **96**, 325 (1954).

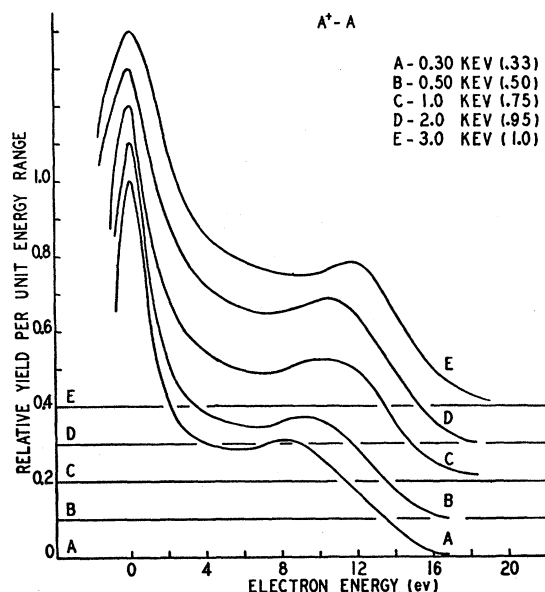


FIG. 3. Electron energy distributions for ionizing collisions of A^+ in A. The numbers appearing after the beam particle energies represent the relative heights of the zero-energy peak.

There still exists though a possible source of trouble. The grid was designed to present as small an obstacle as possible to the ions and atoms scattered from the beam. Yet, since it is the most negative electrode within the cylinder, it will still attract any slow positive ions produced in a charge exchange collision. Secondary electrons originating on the grid wires would generally be indistinguishable from the ionization electrons. However, the space within the grid does not have the potential of the grid. An electrolytic tank analysis of the potential distribution within the cylinder showed that the space inside the grid was almost uniform in potential with a value 20% lower than the potential of the grid relative to the cylinder. Over an area of cross section four times larger in diameter than the beam, the potential varied less than 2%. The value of the potential within the grid may be altered by changing the voltage between the grid and the cylinder, while still leaving the total potential difference constant. In this way, the potential of the point of origin of the ionization electrons can be shifted while that of the secondary electrons from the grid remains fixed. When this was done it was found that the peaks on the data associated with the ionization electrons shifted by the expected amount while those associated with electrons from the grid wires remained fixed.

If the zero-energy peak position of the ionization electrons is plotted against the potential difference between the ionization cylinder and the grid, the result is a straight line which, when extrapolated to zero potential difference, appears near the expected position for electrons of zero self-energy. Actually this was true only for ionization by incident fast neutral atoms; for

ions an additional peak shift was superimposed on that above which moved the peaks to lower apparent energy. Such an effect as this could arise from a positive space charge accumulating within the grid. Since it was not present for ionization by neutral atoms, the space charge very likely arises from the slow positive ions produced in charge-exchange collisions. Because of the small size of the grid wires it seems possible that an equilibrium concentration much larger than the space charge of the beam alone would be reached, which is largely beam current independent, and which would give the observed change in the potential of the region. This effect amounted to almost two volts for argon ions in argon and about one-half a volt for helium ions in helium.

As described above, when the peak position for the neutral atom ionization is extrapolated to zero potential difference between the grid and the cylinder, the position corresponded within one half a volt to the total potential difference between the grid and the entrance slit of the analyzer. It would seem reasonable that this difference could be accounted for by a contact potential difference. Positions on the recorder chart were reproducible to better than $\frac{1}{10}$ inch so that absolute values of the energy are in doubt by about 0.5 eV. Because all the distributions showed the same rapid rise with increasing voltage, it seemed reasonable to take the peak as the zero of the electron self-energy (Figs. 3 through 6). Hence all data were adjusted to match at this peak. In some cases the yield at higher collision energies produced a broader peak, and some justification would exist for considering the edge of the rapid rise as more

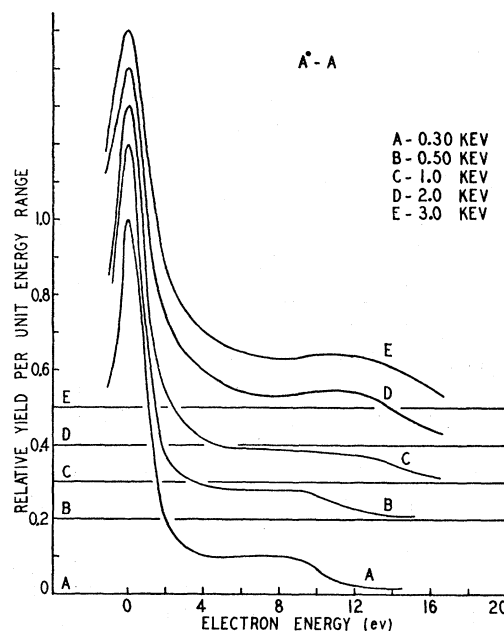


FIG. 4. Electron energy distributions for ionizing collisions of A neutral atoms in A. The beam energy is indicated by the same letter as used on the curve.

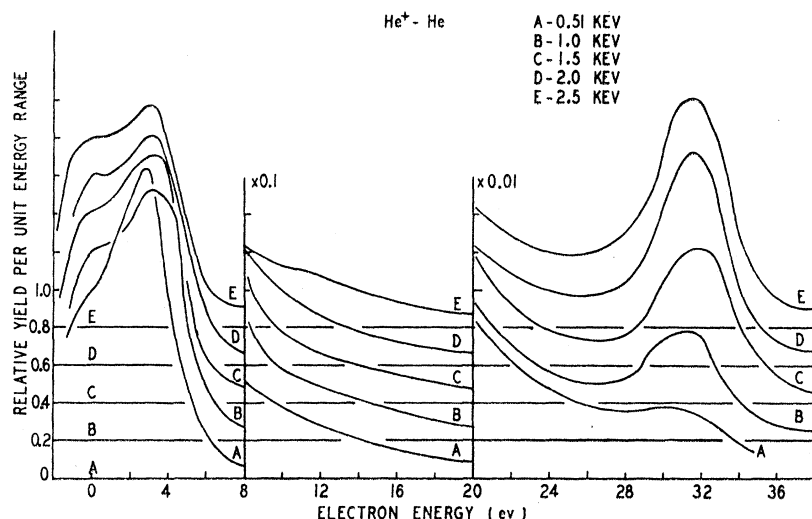


FIG. 5. Electron energy distributions for ionizing collisions of He^+ in He. The incident particle energy is indicated by the same letter as used on the curve. Sections of each curve are magnified by 10 and 100 successively.

appropriate for the zero. It should be noted also that the slit width at the point of zero self-energy is almost one ev.

There is a possibility that the passage through the grid would affect the low-energy part of the distribution since the region in which the ionization electrons originate is more positive than the grid. This did not seem to be the case since the peak height and width remained unchanged as the potential difference between the grid and the ionization cylinder was changed. Further evidence for a lack of effect in this was found in the electrolytic tank study of an enlarged model of the system.

RESULTS

Figures 3, 4, 5, and 6 show the energy distributions observed for argon and helium ions and neutral atoms in the parent gas. The ordinate is proportional to the number of electrons per electron volt represented as a fraction of the zero energy peak height. The data have been corrected for the variation in slit width with analyzer energy by dividing the observed current by the total energy of the electrons selected. The curves are also displaced upward in the figure to reduce the confusion from overlapping. The base line for each curve is indicated by the same letter as used on the curve, as are the incident particle energies.

The electron energy spectrum for ionization by argon ions in argon (Fig. 3) shows, for all collision energies, a rapid decrease in yield as the electron energy increases from zero, with this followed by a plateau terminating in a small peak and finally an almost exponential decrease. While the spectrum is nearly collision energy independent, the peak coming after the plateau appears at about 8.5 ev for the lowest beam energy used (0.30 kev) and shifts upward to about 12 ev at 3.0-kev beam energy. Many of the data records showed evidence of a double peak at 12 ev but attempts to resolve these with narrower analyzer slits failed. Similar distributions

(Fig. 4) were found for the ionization of argon by argon neutral atoms. Again the spectrum shows an extended flat yield from about 4 to 12 ev for the higher collision energies. The yield in the neighborhood of the plateau is about one-half that for argon ions, and also the peak at the end of the plateau is less pronounced but again shows an upward shift with collision energy.

A similarly interesting spectrum was found for the helium ion and neutral atom ionizing collisions in helium. These are shown in Figs. 5 and 6. The helium ion electron distributions are partly masked at low energies by the electron emission from the grid. This was so identified by the shift of the ionization spectrum with changing grid-ionization cylinder potential difference described above. The data have been plotted like that above except that the shoulder on the rising part of the distribution was taken as the usual zero peak. This shoulder becomes relatively more prominent as the collision energy increases. Such would be expected since the cross section for ionization increases with the relative velocity of collision, while the charge-exchange cross section decreases. The large maximum in the region of 4 ev should therefore be disregarded. One might expect the ionization electron distribution to show again a rapid drop as observed with the neutral helium atom collisions. An interesting part of these distributions is the maximum observed in the neighborhood of 31 ev. Again this can be identified as the ionization electron contribution by the shift of the spectrum with variation of the grid-ionization cylinder potential difference. This peak position is essentially collision energy independent although the relative yield increases sharply with energy.

The neutral helium collisions, however, produce a somewhat different spectrum. The high-energy group is much less pronounced and decreases into a mere suggestion of a peak as the collision energy increases. Also, the peak or plateau end is in the neighborhood of 16 ev. The yield observed for the 0.51-kev collisions was

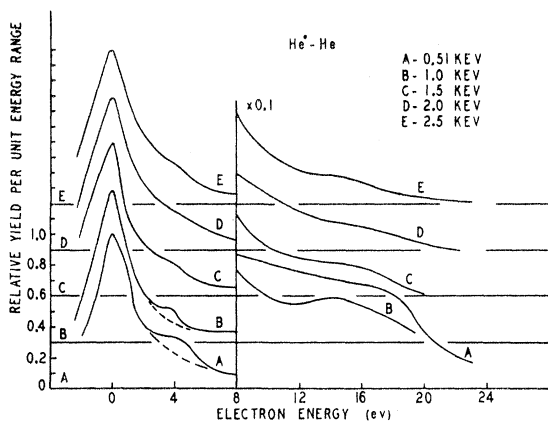


FIG. 6. Electron energy distributions for ionizing collisions of He neutral atoms in He. From 8 ev on, the curves have been magnified by a factor of 10.

very small and seemed to be distorted more than the rest by secondary electrons. Probably, this accounts for the apparently greater relative yield and higher position of the plateau end. For these data, the observed current in this region was only a few times the background noise. The slight rise at 4 ev for the lower energy distributions has the behavior of electrons from the grid wires. The dashed curves in Fig. 6 represent the estimated ionization electron spectra.

It is interesting to note that the high-energy electron groups in He^+-He and A^+-A occur at values (31 and 12 ev, respectively) which seem related to the atomic energy states. These energies are nearly the differences of the first and second ionization potentials. It might indicate a collision in which the incident ion is nearly ionized again but through an Auger transition two ionized atoms and a free electron are produced. A similar relationship can be found for the peaks or plateau ends of the neutral atom induced ionization but involving the excitation of the colliding particles with the subsequent production of an ion and a free electron.

It should be noted that the ionization by ions will include a contribution by those neutral atoms produced through charge exchange in the ionization cylinder. Whenever gas was introduced into the cylinder preparatory to taking data, the ion beam dropped by one-third to one-half of its initial value. Very likely, this was largely the result of partial neutralization of the beam by charge exchange. One might estimate, therefore, that about twenty percent is neutral as the beam passes the exit slit of the ionization cylinder.

ACKNOWLEDGMENT

The assistance of Mr. Lawrence Juhlin in the construction of the apparatus and analysis of the data is very much appreciated.