EXPERIMENTAL STUDY OF ELECTRON LOSS BY MULTIPLY CHARGED IONS IN GASES

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Measurements are reported of cross sections for collisions with loss of one electron by ions of light elements ($Z = 2-18$) in helium, nitrogen, argon, and krypton, for ion velocities $v = (2.6-12) \times 10^8$ cm/sec. The cross section per electron of the ion's outer shell was usually determined by the binding energy of the lost electron. The velocity dependence of the cross section was in agreement with Massey's adiabatic hypothesis. The dependence of the cross section on the atomic number of the gaseous medium indicated a strong influence of the polarization of gas atoms. It was found that the regions of applicability of theoretical calculations for high and low ion velocities nearly overlapped.

1. INTRODUCTION

ELECTRON loss on passage of ions through matter has been studied most extensively for ions of hydrogen and helium.[1] Electron loss by heavier ions has been studied mainly at ion velocities $v < 10^8$ cm/sec. Questions connected with the passage of fast multiply charged ions through matter require information on cross sections of electron loss at ion velocities close to the orbital electron velocity. This region of ion velocities is difficult to treat theoretically and high-velocity data are available only for electron loss by ions of lithium,[2] nitrogen[3-5] and oxygen.[6]

The present paper is a report of an experimental study of cross sections, $\sigma_{i,i+1}$, for collisions with loss of one electron by ions of light elements ($Z = 2-18$) and by Kr ions in helium, nitrogen, argon, and krypton. Ion velocities ranged from 2.6×10^8 to $\sim 12 \times 10^8$ cm/sec. The initial ionic charge i varied from 1 to 6. Cross sections were measured by mass spectrometry using the apparatus described in an earlier paper.[7]

Values are also given of the sum $\Sigma_i' \sigma_{0i}$ of the cross sections for the loss of one and more electrons by neutral atoms, where the first subscript of σ denotes the initial ion charge and the second subscript denotes the final charge. These sums were deduced from experimentally determined equilibrium charge distributions[8] and electron capture cross sections.[7,9] The following formula was used in the calculations

$$\sum_i' \sigma_{0i} = \sum_k' \Phi_k \sigma_{k0} / \Phi_0,$$

where Φ_k is the relative number of ions of charge k in the equilibrium charge distribution and σ_{k0} is the cross section for capture of k electrons by ions with initial charge k. Since cross sections representing loss of two or more electrons are usually considerably smaller than the cross section for loss of one electron, the sum $\Sigma_i' \sigma_{0i}$ is close in value to the cross section σ_{01} for one-electron loss.

2. MEASUREMENT RESULTS

The cross sections $\sigma_{i,i+1}$ for loss of one electron per atom of the gaseous medium are shown in Figs. 1—4. The experimental errors in these cross sections do not usually exceed ±15%.

The measured cross sections represent electron loss accompanied by scattering of ions through angles $\theta \lesssim 0.005$ radian.[7,9] The contribution to the cross section due to ions scattered through larger angles should not exceed $\frac{1}{2}\sigma_S$, where σ_S is the total cross section for scattering through angles $\theta \gtrsim 0.005$ radian.[7] The values of $\frac{1}{2}\sigma_S$ for singly charged nitrogen ions are given in Fig. 1. The values of $\frac{1}{2}\sigma_S$ for other ions are similar.[7] The graphs show that in helium and nitrogen the quantity $\frac{1}{2}\sigma_S$ is always smaller than the experimental error of $\sigma_{i,i+1}$. For multiply charged ions in argon and krypton the quantity $\frac{1}{2}\sigma_S$ may be greater than the experimental error of $\sigma_{i,i+1}$. Since σ_S decreases and $\sigma_{i,i+1}$ increases with increase of the ion velocity, the ratio $\sigma_S/\sigma_{i,i+1}$ is small at high ion velocities.

The cross sections reported here can be com-

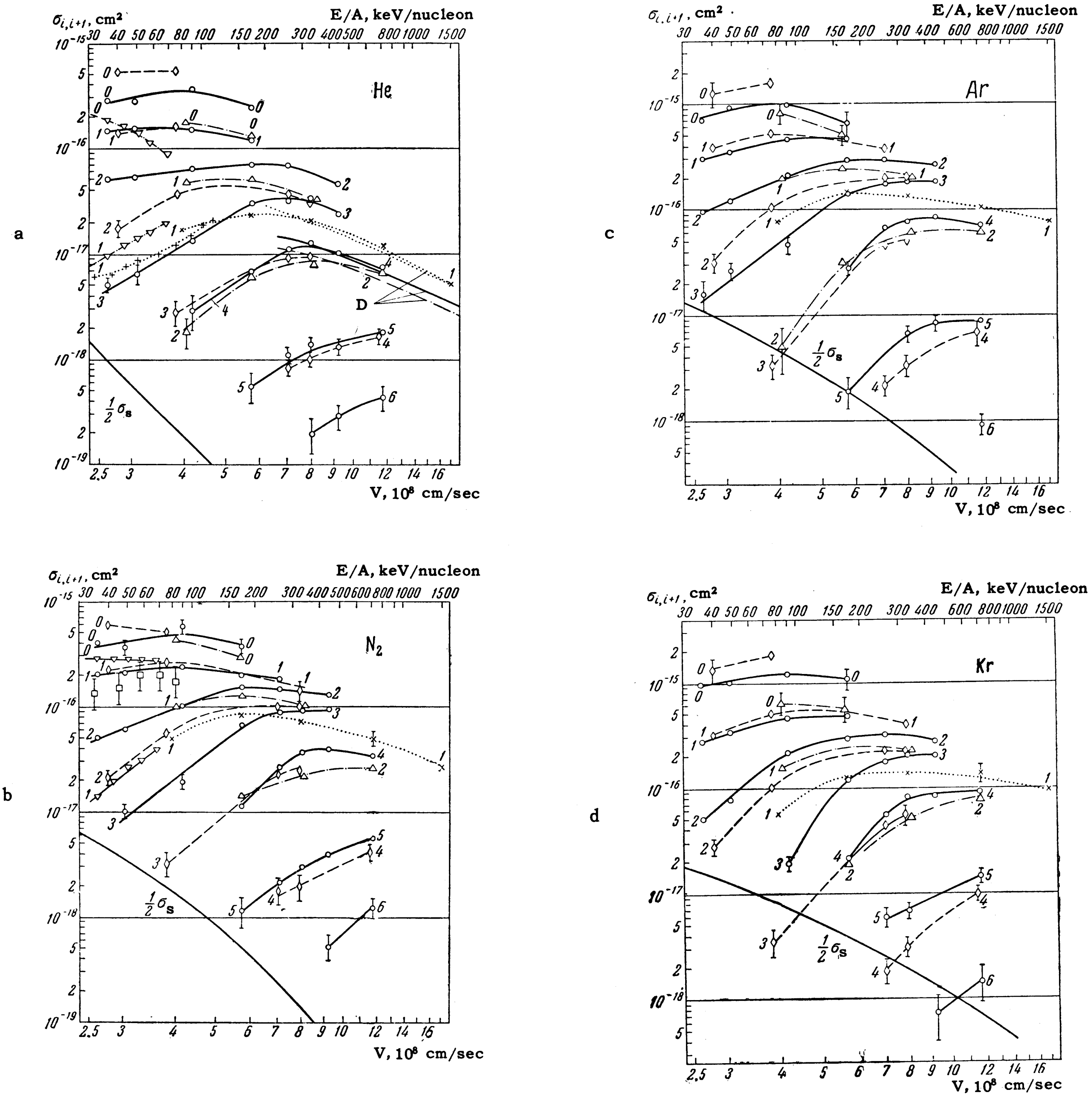

FIG. 1. Cross sections for loss of one electron, $\sigma_{i,\,i+1}$, as a function of the ion velocity, v, and the ion energy per nucleon, E/A, in helium (a), nitrogen (b), argon (c) and krypton (d). × denotes He ions, Δ – Li ions, ◇ – B ions, and ○ – N ions. The values of i are marked near the curves. The error is shown only when it exceeded 15%. The sign + denotes σ_{12} values for He ions, taken from Allison's review;[1] ∇ denotes values of σ_{01} and σ_{12} for Li ions, taken from the work of Allison et al.;[2] □ denotes σ_{12} for N ions, taken from the work of Korsunskii;[3] D denotes theoretical curves for He^{+}, Li^{2+} and N^{4+} ions.

pared with other experimental results only in the case of He, Li, and N ions. The values of σ_{12} for He ions in helium and nitrogen at $v \approx 4 \times 10^8$ cm/sec agree, within the experimental error, with the values of σ_{12} in helium and air reported by Allison.[1] At $v = 11.7 \times 10^8$ cm/sec the cross section σ_{12} of He ions in nitrogen agrees with σ_{12} in air obtained by Jacobsen.[10] The values of σ_{12} for He in air taken from Rutherford's work and quoted by Allison[1] are 30–40% lower than those obtained by Jacobsen and the present authors.

For Li ions the values of σ_{01} and σ_{12} in helium and nitrogen at $v \approx 4 \times 10^8$ cm/sec are about twice as large as those obtained by Allison.[2] The general nature of the dependence of σ_{01} on the ion velocity, deduced from the present work and that of Allison, indicates that near 4×10^8 cm/sec Allison's values are too low. Allison determined cross sec-

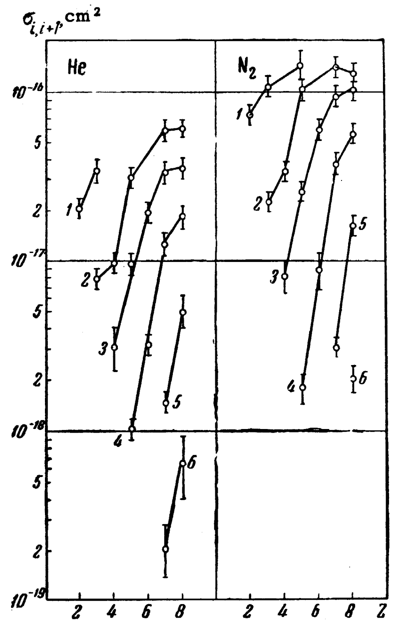

FIG. 2. Dependence of $\sigma_{i,\,i+1}$ on the atomic number Z for $v = 8 \times 10^8$ cm/sec in helium and nitrogen. The initial ionic charge i is marked near the curves.

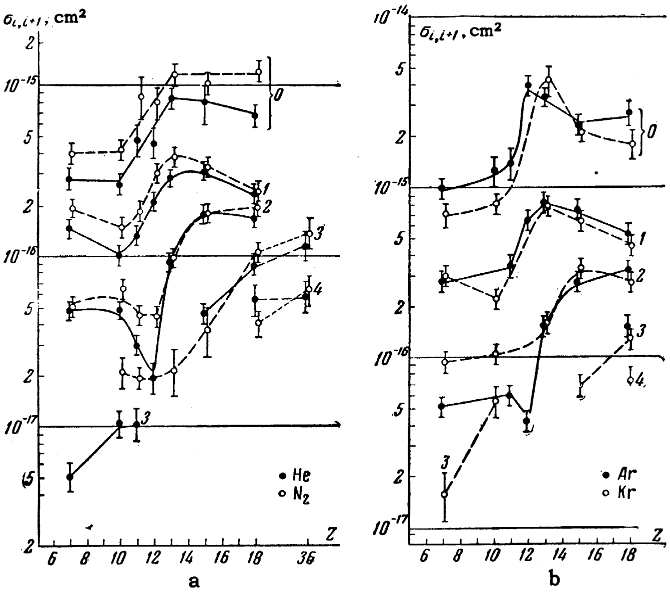

FIG. 3. Dependence of $\sigma_{i,\,i+1}$ on Z for $v = 2.6 \times 10^8$ cm/sec in helium, nitrogen, argon, and krypton. The initial ionic charge i is marked near the curves.

tions by attenuation of a beam in a magnetic field and the measured cross sections applied only to 5–10% of the beam particles. Under these conditions considerable errors could be caused by the difficulty of allowing for reverse processes. The difference between the two sets of σ_{12} values may be due to some of the Li^+ ions in the present work being in an excited state.

The values of σ_{12} for N ions in nitrogen obtained by Korsunskii et al [3] were 25% lower than those reported here, but this difference is within

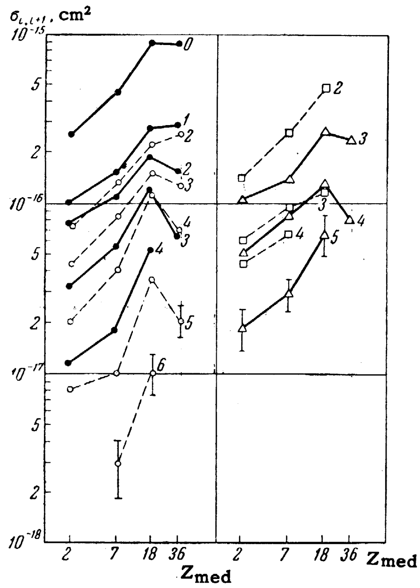

FIG. 4. Dependence of $\sigma_{i,\,i+1}$ on the atomic number of atoms in the gaseous medium, Z_{med}, for Ne ions ($v = 4.1 \times 10^8$ cm/sec, denoted by black dots, and $v = 5.6 \times 10^8$ cm/sec, denoted by open circles), P ions (denoted by □) and Ar ions ($v = 4.1 \times 10^8$ cm/sec, denoted by Δ).

the experimental error because Korsunskii et al found these cross sections with an accuracy of ±30%. The cross sections for electron loss by N ions in nitrogen and argon obtained earlier [4] using somewhat different experimental geometry were, within the experimental error, identical with those reported in the present work.

Figure 1 shows that the general nature of the dependence of cross sections on the ion velocity is the same in all four gases. For ions with i = 1 or 2, and also for N ions with i = 3 and 4, the cross sections have maxima in their dependences on the ion velocity. The velocity v_m at which the cross section becomes greatest increases with increase of the ionic charge and obeys $v_m \approx \gamma u$, where $u = (2I/\mu)^{1/2}$, I being the binding energy of the lost electron, μ the electron mass, and γ a coefficient that depends only on the medium (Fig. 5). With increase of the atomic number of the medium, Z_{med}, the coefficient γ increases from ≈1.3 in helium to ≈2 in krypton.

The formula for v_m is valid also for neutral atoms of nitrogen, helium, and lithium and for negative hydrogen ions, for which experimental data are available [1,2] (Fig. 5). When $v < v_m$ the dependence of $\sigma_{i,i+1}$ on v is strongest for ions with large values of i (or I). For example, the cross section for N^{4+} ions fell to half its maximum value when the ion velocity was reduced by a factor of 1.5, while the cross section for N^+ ions fell to half its maximum value only when v was reduced by a factor of 3.

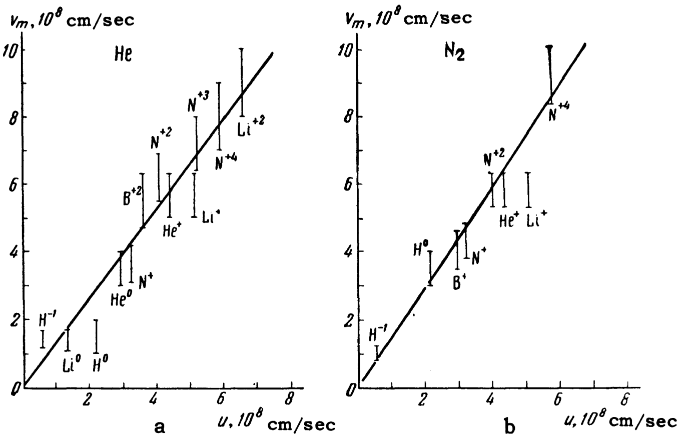

FIG. 5. Relationship between the velocities u and v_m for various ions in helium and nitrogen.

Figures 2 and 3 show that for a given value of i the cross sections $\sigma_{i,i+1}$ rise in general with increase of Z. A rapid rise of $\sigma_{i,i+1}$ was found for ions with a small number of electrons in the outermost shell. In the case of ions with the outermost shell almost completely filled the cross section for electron loss was not greatly affected by an increase in Z.

For ions of a given element the cross section decreased as $\sigma_{i,i+1} \sim \exp(-mi)$ with increase of the ionic charge i. When $v \approx 3 \times 10^8$ cm/sec, $m \approx 1$ for $Z = 10$ and 18 and $m \approx 1.5$ for $Z = 3$ and 12. With increase of the ionic velocity the value of m fell somewhat. In the case of ions with one electron outside the filled shell the values of $\sigma_{i,i+1}$ were usually 5–8 times greater than in the case of ions which did not have this electron. Consequently it can be assumed that electrons are lost mainly from the outermost shells of the ions. The dependence of $\sigma_{i,i+1}$ (per electron in the outer shell) on i and Z reduces essentially to a dependence on the electron binding energy I (Fig. 6). An increase in I caused a decrease in $\sigma_{i,i+1}/q$ (q is the number of electrons in the outermost shell). The strongest dependence of $\sigma_{i,i+1}/q$ on I ($\sigma_{i,i+1}/q \sim I^{-2.5}$) was observed for $I > \frac{1}{2}\mu v^2$. The quantity $\sigma_{i,i+1}/q$ did not depend strongly either on the number of electrons or on the shell number; somewhat larger values of $\sigma_{i,i+1}/q$ were found in the case of loss of M electrons. Increase of $\sigma_{i,i+1}/q$ was observed also for $v < u$ in ions with $i = Z - 2$ formed in a celluloid film at a distance of 1.5 m from a charge-exchange chamber (see Fig. 1 in [7]). No such increase was found in the case of Be^{2+} ions which were not passed through this film and were formed at a considerably larger distance from the chamber.

For the same values of v/u the quantities $\sigma_{i,i+1}/q$ were approximately proportional to $I^{-\alpha}$,

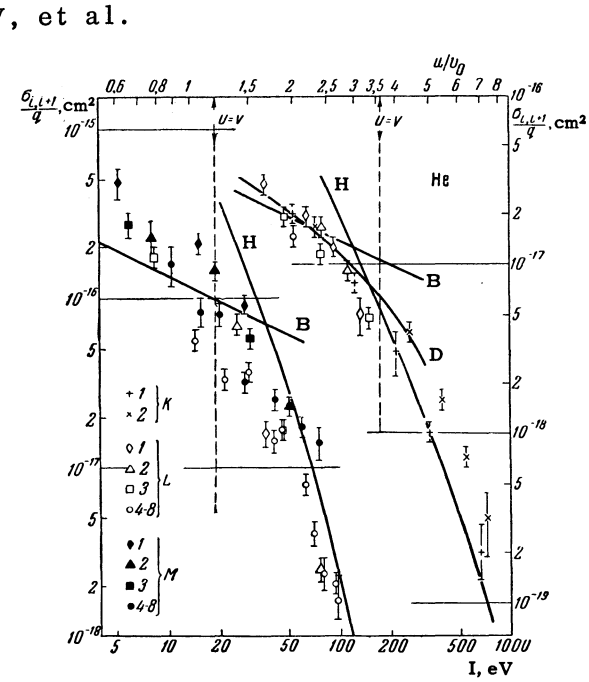

FIG. 6. Dependence of $\sigma_{i,i+1}/q$ on I and on u/v_0 in helium for $v = 2.6 \times 10^8$ cm/sec (left-hand part of the figure) and $v = 8 \times 10^8$ cm/sec (right-hand part). The values of q for K, L and M electrons are given in the figure. H, B and D denote, respectively, the theoretical curves of Henneberg, Bohr, and the present authors.

where α is close to unity and depends weakly on v/u: $\alpha \approx 1.3$ when $v/u \approx 0.5$ and $\alpha \approx 1$ when $v/u \approx 1.5$. Values of $\sigma_{i,i+1}/q$ reported for atoms of helium,[1] lithium,[2] and boron and carbon,[11] at $v/u = 0.55$ lie on the same general curve $\sigma_{i,i+1}/q = f(I)$ as obtained for ions with large charges (Fig. 7).

The cross sections for electron loss in helium, nitrogen, and argon rose with increasing atomic number of the medium, Z_{med}. When $v \approx 5 \times 10^8$ cm/sec the quantity $d(\log \sigma_{i,i+1})/d(\log Z_{med})$ was of the order of unity for He and Li ions and about 0.5 for heavier ions. With increase of the ionic velocity the dependence of $\sigma_{i,i+1}$ on Z_{med} became stronger: the ratio of the cross sections in nitrogen to those in helium varied as $v^{0.8}$ while the ratio of cross sections in argon to those in helium was proportional to $v^{1.1}$. At low ionic velocities the cross sections of the majority of multicharged ions were smaller in krypton than in argon and the relative difference of the cross sections in these two gases rose with increase of the ionic charge i. The ratio $(\sigma_{i,i+1})_{Kr}/(\sigma_{i,i+1})_{Ar}$ was roughly proportional to $v^{1.5}$ so that at $v > 10^9$ cm/sec the cross sections in krypton became greater than or comparable with those in argon. When $v \approx 8 \times 10^7$ cm/sec the cross sections decreased in heavier gases also for neutral atoms.[11]

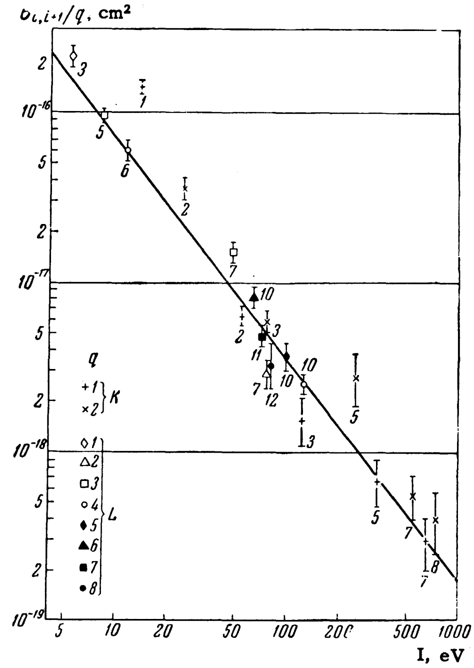

FIG. 7. Dependence of $\sigma_{i,i+1}/q$ on I when $v/u = 0.55$ in helium. The numbers next to the experimental points give the values of Z. The values of q for K and L electrons are given in the figure. The cross sections $\sigma_{i,i+1}$ for H, He and He^+ are taken from Allison's review,[2] those for Li and Li^+ from the work of Allison et al,[2] and those for B and C from the work of Fogel' et al.[11] The sloping straight line represents a dependence of $I^{-1.3}$ type.

3. DISCUSSION

Approximate calculations of $\sigma_{i,i+1}$ for multiply charged ions were carried out by Gluckstern[6] for the range of ionic velocities close to the orbital velocities of the lost electrons. These calculations were performed for ions of oxygen, neon, phosphorus, and argon in argon gas with $v > 6.5 \times 10^8$ cm/sec. Extrapolation of the calculated cross sections to lower ion velocities gives results which are in good agreement with the experimental data obtained by the present authors. Moreover the general nature of the dependences of the calculated cross sections on v and I is close to the empirical relationships.

The simplest case suitable for theoretical study is the loss of one electron when $v \gg u$ and when the electron-ion interaction can be neglected in collisions with atoms of the gaseous medium. Bohr's formula[12] corresponds to this case for electron loss in media consisting of light atoms; this formula gives for helium ions in helium at $v \doteq (8-17) \times 10^8$ cm/sec values of σ_{12} which are 1.5 times greater than the experimental values. Allowance for the screening of the Coulomb field of the helium atoms by the atomic electrons shows that for ions with $u < v$ the theoretical values of $\sigma_{i,i+1}/q$ in helium agree with experiment when $v \geq 8 \times 10^8$ cm/sec. Details of calculations which allow for the screening effect will be published in a separate paper; the results are shown in Fig. 1a and in Fig. 6 where they are denoted by D. Figure 6 indicates that it is permissible to neglect the interaction of the electron that is being lost with the ion up to ion velocities $v = u$.

Electron loss in media consisting of heavy atoms can be dealt with using Bohr's approximate formula,[12] generalized to the case of electron loss by any ion:

$$\sigma_{i,i+1} \approx \pi a_0^2 q Z_c^{2/3} v_0^2 / vu,$$

where $a_0 = 0.53 \times 10^{-8}$ cm, $v_0 = 2.19 \times 10^8$ cm/sec. This formula was obtained using classical mechanics and it should apply to ions with $u \gg Z_{med}^{1/3} v_0$, i.e., for $I \gg 50$ eV in nitrogen, and for $I \gg 150$ eV in krypton. The experimental dependence of $\sigma_{i,i+1}/q$ on I was close to that given by the above formula even at much smaller values of I: when $u \leq 1.5\,v$ the theoretical and experimental cross sections differed by a factor of less than 2. A similar correspondence between theory and experiment was also found for helium (Fig. 6).

In the region $v \ll v_0$ many cases of electron loss by neutral atoms in heavy gases can be treated by the approximate method of Firsov.[13] The results reported in the present work show that even for $v = 1.2\,v_0$ the experimental values of $\Sigma_i^{\prime}\sigma_{0i}$ for atoms with $Z \geq 5$ in nitrogen, argon, and krypton differ only by a factor of 2.5 or less from the quantities σ_{01} reported by Firsov.

Henneberg[14] calculated cross sections for K-electron knocked out by α particles from atoms with $Z \gg 2$; these calculations were carried out in the Born approximation for $v < u$. Henneberg's results can be used to deal with K-electron loss in helium. Although we are dealing with helium atoms and not helium nuclei, the cross sections for loss of the last K electron, $\sigma_{Z-1,Z}$, calculated by Henneberg for B^{4+} and N^{6+} ions for $(v/u) < 1$, agree with experiment (Fig. 6). For the same values of v/u the experimental values of $\sigma_{Z-1,Z}$ for He^+ and Li^{2+} ions are 2–3 times smaller than the theoretical ones, owing to violation of the condition $Z \gg 2$.

The experimental results reported here indicate that Henneberg's calculations can be used also to deal with loss of L and M electrons because, in most cases, the values of $\sigma_{i,i+1}/q$ depend weakly on the electron shell structure. An interesting exception is the case of electron loss by ions with charges $i = Z - 2$, for which the values of $\frac{1}{2}\sigma_{Z-2,Z-1}$ are 2—3 times greater than the values of $\sigma_{Z-1,Z}$ for the same values of I in the region $v < u$. This increase of cross section is due to one of two electrons being in the metastable state in the L shell after passage through a solid target and consequent capture of the electrons by the excited states of some of the ions. Since the binding energy of this metastable electron is considerably smaller than that of the K electron, the loss cross section is greater. The experimental values of $\sigma_{Z-2,Z-1}$ represent the case when there is one metastable electron in the L shell in 10—20% of the ions.

Henneberg's calculations can also be used to describe results for heavier gases because in such gases the dependence of cross sections on I is similar to that in helium. According to Henneberg, the cross sections should be proportional to $Z^2_{\rm med}$ but the experimental dependence of the cross sections on $Z_{\rm med}$ is much weaker. Consequently we should use an effective value of $Z_{\rm med}$, denoted by $Z^*_{\rm med}$, which depends on v. For example, for $v = 8 \times 10^8$ cm/sec the values of $Z^*_{\rm med}$ for nitrogen, argon, and krypton are 2.7, 4.1, and 3.7, respectively.

The smaller cross sections and the smaller values of $Z^*_{\rm med}$ in krypton, as compared with argon, indicate that deformation of atoms in the medium affects strongly the cross section magnitudes. In an undeformed krypton atom the Coulomb field is greater than in an argon atom at all distances from the nucleus. The field in krypton can be reduced only by additional screening of the nucleus when the atom is polarized. Since the atomic polarizability rises with increase of $Z_{\rm med}$, this additional screening is stronger in media consisting of heavier atoms. The polarization should rise with increase of i and fall with decrease of v, because at ion velocities larger than the orbital electron velocity there is insufficient time for the atoms of the gas medium to be deformed during collisions. The experimental results support these ideas: reduction of electron-loss cross section due to atomic polarization caused a fall of the mean charge of ions which passed through krypton.[8]

Comparison of the experimental results with theoretical calculations for very high or low ionic velocities shows that the regions of the applicability of these calculations practically overlap. It follows that the theoretical calculations, with appropriate corrections, are valid in the region $v \approx u$ which is the most difficult to treat theoretically.

Krasner[15] attempted to reduce the dependence of $\sigma_{i,i+1}/q$ on v/u for various ions in a given medium to one common curve. Later Fogel' et al[16] showed that the values of $\sigma_{i,i+1}/q$ do not depend on v/u alone. The results reported in the present work indicate that, in the region v/u = 0.5—1.5, the values of $\sigma_{i,i+1}/q$ taken at the same ratios v/u are approximately inversely proportional to I, and the electron-loss cross sections can be approximated by

$$\sigma_{i,i+1} \approx qI^{-1}f(v/u),$$

where f(v/u) is a function characteristic of the given medium. This relationship gives the best agreement with experiment for values of $\sigma_{i,i+1}$ near their maxima (Fig. 8). When the ratio v/u is reduced, the values of $\sigma_{i,i+1}I/q$ for various ions begin to differ because the dependence of $\sigma_{i,i+1}/q$ on I is stronger in the region of small v/u.

The general dependence of $\sigma_{i,i+1}$ on v and the expression for electron-loss cross sections near their maxima are in agreement with the adiabatic hypothesis of Massey.[17] According to this hypothesis the cross section should be a function of the adiabatic parameter $|\Delta E|a/\hbar v$ and should be greatest at $|\Delta E|a/\hbar v \approx 1$, where ΔE is the difference between the electron binding energies in the initial and final states, and a is the distance over which interaction takes place. In our case the adiabatic parameter is the ratio u/v, which

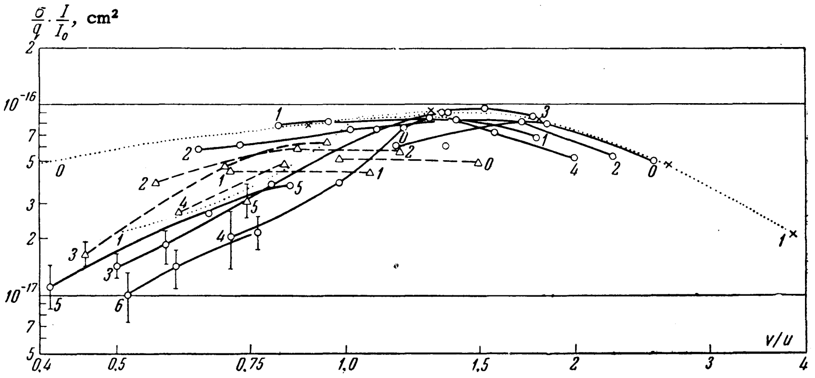

FIG. 8. Dependence of $\sigma_{i,i+1}I/qI_0$ on v/u for He (×), N (○) and Ne (△) ions in helium (I_0 = 13.5 eV). The values of i are marked by the curves. The values of σ_{01} for helium atoms were taken from Allison's review.[1]

can be expressed in a form suitable for the adiabatic treatment by assuming that $|\Delta E| = I = \mu u^2/2$ and $a = 2\hbar/\mu u = 2r/n$, where $r = n\hbar/\mu u$ is the mean radius of the electron-losing shell and n is the principal quantum number.

In electron capture by singly charged ions and by neutral atoms a depends only on the initial charge of the ion or atom,[11] but in the case of electron loss by positive multiply charged ions we find that a depends on I and is of the order of ionic dimensions. For example, when electrons are removed from the L-shell we have $n = 2$ and $a = r$. In the case of K-electron loss with the same value of r the value of a is twice as large. This difference between K- and L-electrons is due to the fact that the distance $a = 2\hbar/\mu u$ is governed not by the ion dimensions but by the momentum $p_e = \mu u$ which is necessary to remove an electron from an ion.

Drukarev[18] pointed out that the adiabatic parameter can be represented in the form $pa/\hbar$ or p/p_0, where $p = |\Delta E|/v$ is the change of momentum of an ion in the case of forward inelastic scattering, and $p_0 = \hbar/a$ is the change of ion momentum corresponding to the greatest probability of the process considered. Using the expression for a given above, we find that $p_0 = 2p_e$, i.e., p_0 is directly proportional to the momentum p_e transferred to the electron.

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