EXPERIMENTAL INVESTIGATION OF THE EFFECTIVE CROSS SECTIONS FOR THE ANNIHILATION AND FORMATION OF FAST NEGATIVE IONS IN ATOMIC COLLISIONS

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Measurements were made of the effective cross sections for the loss—in helium, nitrogen, and argon—of one, two, and three electrons from negative carbon, nitrogen, and oxygen ions formed by charge exchange of positive ions accelerated in a 72-cm cyclotron to a velocity $v = 2.6 \times 10^8$ cm/sec. The cross section for the simultaneous loss of two electrons by negative ions amounts to about 50—70% of the value of the cross section for the loss of one electron. A comparison of the data obtained here with the known cross sections for the loss of electrons by other negative and positive ions shows that the characteristic feature of negative ions, which is weak binding of the outer electron to the ion core, has little effect on the considered characteristics of the interaction with matter of negative ions of velocity $v = 2.6 \times 10^8$ cm/sec.

For carbon and oxygen, data were obtained on the cross sections for the formation of negative ions as a result of the capture of two electrons by positive ions and the capture of one electron by neutral atoms. Moreover, the equilibrium values were obtained for Φ_{-1}, which was the fraction of negative carbon and oxygen ions in a beam which had traversed a sufficiently thick layer of matter. The maximum values of Φ_{-1} were obtained for media in which the cross sections for the formation of negative ions at a given velocity were maximal.

1. INTRODUCTION

An investigation of the process of ionization of fast multiply charged ions by collision with gas atoms has shown that, at a fixed velocity of colliding particles $v \gtrsim v_0 = 2.19 \times 10^8$ cm/sec, the average cross section for the loss of a single electron from the outer shell of an ion, σ_i, is governed by its ionization potential I_i and is independent of the number of electrons in the shell, and that the value of σ_i increases monotonically as I_i decreases from 700 to 10 eV.[1]

Because of insufficient experimental data, it is at present difficult to predict to what degree similar relationships apply to negative ions whose values of I_i are an order of magnitude smaller than those for positive ions. In fact, in the range of velocities $v \gtrsim v_0$, the processes of electron loss have been investigated only for H^- ions[2] and for Li^- ions.[2-3] In the case of heavier ions, similar measurements have been carried out only in the range $v \ll v_0$,[4-6] where these dependences were not observed.

Therefore, in the study described here, we measured the cross sections for the loss of one, two, and three electrons by C^-, N^-, and O^- ions in helium, nitrogen, and argon for an ion velocity $v = 2.6 \times 10^8$ cm/sec (energy ≈35 keV/nucleon), which has been used earlier to determine the cross sections for the loss of electrons by many positive ions.[7] We also obtained data on the values of the cross sections for the formation of negative carbon and oxygen ions, $\sigma_{1,-1}$, by the attachment of two electrons to corresponding singly charged positive ions, as well as the equilibrium values of the fraction of negative ions $\Phi_{-1,\infty}$ in a beam which had passed through a sufficiently thick layer of matter. From the experimental data on the cross sections and on the equilibrium ion charge distribution, we calculated the values of the cross sections $\sigma_{0,-1}$ for the formation of fast negative ions from neutral atoms by the capture of electrons. Until now, only

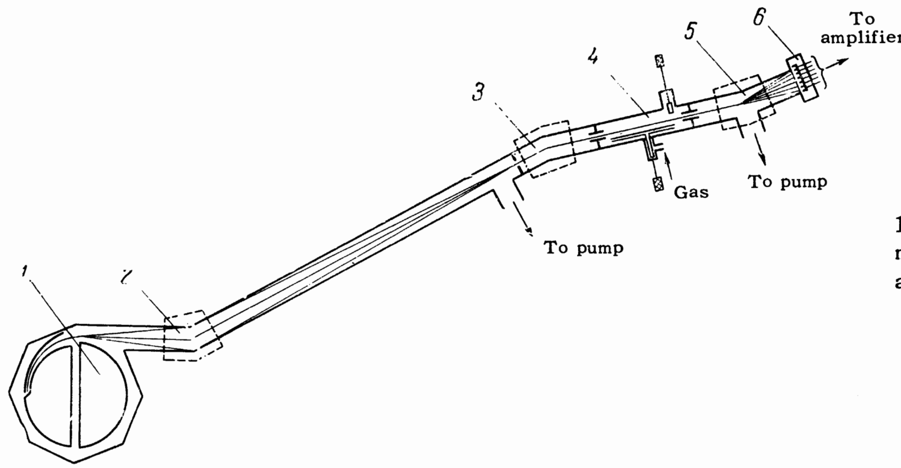

FIG. 1. Schematic diagram of apparatus. 1) Cyclotron; 2) focusing magnet; 3) mass monochromator; 4) collision chamber; 5) analyzer; 6) detectors.

the data on the values of the quantities $\sigma_{0,-1}$, $\sigma_{1,-1}$ and $\Phi_{-1,\infty}$ have been known in the range of velocities $v \lesssim 1 \times 10^8$ cm/sec.[8-11]

2. DESCRIPTION OF EXPERIMENTS

The experimental measurements were carried out using the mass spectrometer shown schematically in Fig. 1 and described in detail earlier.[12] Positive singly charged ions were accelerated in a 72-cm cyclotron using an accelerating electric field of 4.2 Mc frequency in the "third harmonic."[13] The intensities of beams of $^{12}C^+$, $^{14}N^+$, and $^{16}O^+$ ions extracted from the cyclotron were 0.3—0.5 μA. Negative ions were formed by the charge-exchange between positive ions and residual-gas atoms in an ion guide between a focusing magnet 2 and a mass monochromator 3 (equivalent target thickness $\approx 10^{14}$ atoms/cm^2). A beam of negative ions, selected by the mass monochromator, was directed to a collision chamber 4, which was a continuously replenished gas target.

The charge composition of an ion beam which had passed through the collision chamber was determined by a magnetic analyzer 5 and a system of proportional counters 6. During the measurement of the cross sections for the loss of an electron by negative ions, the average density of the current of positive nitrogen ions N^+ in front of the mass monochromator was $\approx 10^{11}$ particles/cm^{2}sec while the density of C^+ and O^+ ions was $\approx 10^{10}$ particles/cm^{2}sec and the density of negative ions in the collision chamber was $\approx 10^4$ particles/cm^{2}sec^{-1} for $^{14}N^-$ and $\approx 10^5$ particles/cm^{2}sec for $^{12}C^-$ and $^{16}O^-$. The nitrogen ions were accelerated working the ion source of the cyclotron under overload conditions, while for the oxygen ions the current did not reach its maximum value.

An investigation of the cross sections for the formation of negative ions, $\sigma_{1,-1}$, and of the equilibrium fractions of negative ions in a beam, $\Phi_{-1,\infty}$, was carried out for $^{12}C^+$ and $^{16}O^+$ ions. In the investigation of the equilibrium distribution in gaseous substances, a narrow gas-filled tube was placed in the path of a beam; in a solid, equilibrium was reached in the target itself, which was a celluloid film whose thickness was ≈ 5 μg/cm^2.

The values of the cross sections for the loss of one electron, $\sigma_{-1,0}$, were measured with an accuracy of about 20%, while the values of the cross sections for the loss and capture of two electrons, $\sigma_{-1,1}$ and $\sigma_{1,-1}$, were determined to within $\approx 30\%$. The error in the measurement of the cross sections for the loss of three electrons could reach 50%.

4. LOSS OF ELECTRONS BY NEGATIVE IONS

Values of the cross sections for the loss of electrons by negative ions, calculated per one atom

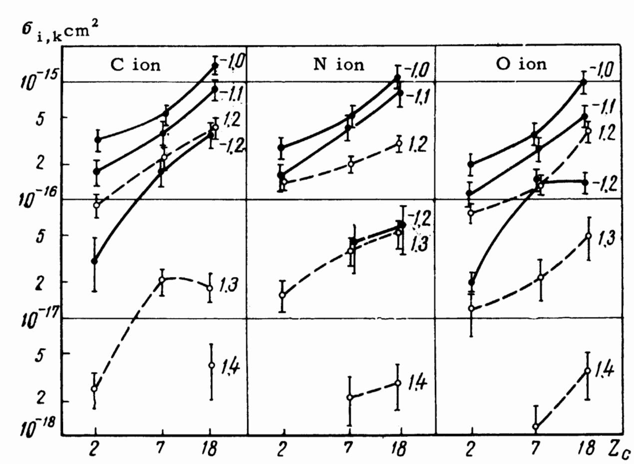

FIG. 2. Dependence of the cross sections for the loss of one, two, and three electrons by negative ions C^-, N^-, and O^- (continuous curves) and positive ions C^+, N^+, and O^+ (dashed curves) on the atomic number of the target Z_C for an ion velocity $v = 2.6 \times 10^8$ cm/sec. The numbers by the curves indicate the initial and final ion charges.

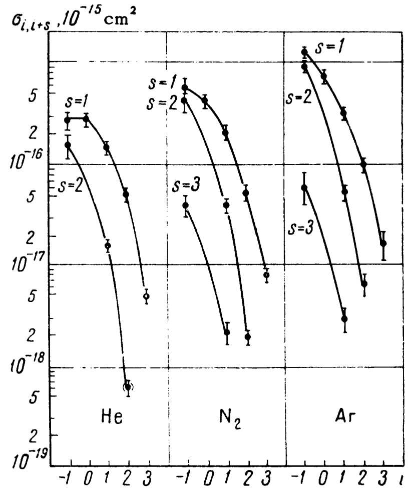

FIG. 3. Dependence of the cross sections for the loss of one (s = 1), two (s = 2), and three (s = 3) electrons by nitrogen ions on the initial ion charge i, in helium, nitrogen, and argon.

of a medium, are given in Figs. 2 and 3. The same figures include, for comparison, the analogous data for positive ions.[7] It is evident from Fig. 2 that when the atomic number of the target, Z_c, is increased from 2 to 18, the cross section for the loss of electrons by negative ions increases as $Z_c^{2/3}$, i.e., approximately at the same rate as the cross section for the loss of an electron by positive ions. The maximum value of the cross section is obtained for carbon ions $^{12}C^-$; this value decreases monotonically when the charge of the ion nucleus increases.

The dependence of the cross section for the electron loss $\sigma_{i,i+s}$ (where s = 1, 2, 3 is the number of lost electrons) on i weakens rapidly when the ion charge i is reduced from 3 to −1 (Fig. 3). In particular, on going over from neutral particles to negative ions, the value of the cross section $\sigma_{i,i+1}$ increases only by a factor 1.2—1.5. Consequently, a considerable decrease in the ionization potential in the transition to negative ions does not give rise to a great change in the cross sections for the electron loss. The curves $\sigma_{i,i+s}(v)$ for s = 1, 2, 3 approach each other when the initial ion charge is reduced. Because of this, the relative probability of the simultaneous loss of two electrons is greater for negative ions than for positive ones. For the ions $^{12}C^-$, $^{14}N^-$, $^{16}O^-$, the ratio $\sigma_{-1,1}/\sigma_{-1,0}$ is a quantity of the order of 0.6—0.7, while for positive ions of the same elements the corresponding ratio is $\sigma_{1,3}/\sigma_{1,2} \approx 0.1$—0.2 (however, for H^- ions the ratio $\sigma_{-1,1}/\sigma_{-1,0}$ is considerably less and equal to 0.07 in helium and 0.2 in nitrogen[2]).

Thus, with the addition of a very weakly bound electron, the relative probability of the loss of a single electron not only does not increase but even decreases slightly. Consequently, we may assume that the expression used in[7] for the average cross section of the loss of one electron from the outer shell in positive ions, $\sigma_i = q^{-1}\Sigma s\sigma_{i,i+s}$ (i is the initial ion charge; q is the effective number of electrons participating in a collision), will be valid also for negative ions, except that the contribution, to $q\sigma_i$, of the cross section for the loss of one electron $\sigma_{i,i+1}$ is less than that in the case of positive ions; for the latter, the value of $\sigma_{1,2}$ represents ≈70% of $q\sigma_i$, while for negative ions the value of $\sigma_{-1,0}$ amounts to only ≈40% of $q\sigma_i$. In the calculation of σ_i, the value of q was selected on the basis of two assumptions. In one case q was assumed to be equal to q_L, i.e., the number of L-electrons in the ions C^-, N^- and O^- which was 5, 6, and 7, respectively. In another case, we took $q = q_L - 2$, i.e., we assumed that the 2s-electrons did not make a great contribution to the cross section for electron loss. The values of σ_i obtained for negative ions in helium (including the data for H^- and Li^- taken from[2,3] are shown in Fig. 4 as a function of I_i. The same figure includes the analogous data for positive ions.[7]

When $v = 2.6 \times 10^8$ cm/sec, the Born approximation for the loss of electrons in helium should give the correct result only for ions with $I_i = (15—30)I_0$, where $I_0 = 13.55$ eV.[14] However, a comparison with experiment shows that this approximation is valid over a wider range of I_i and it gives the correct value, to within 20%, of the cross section $\sigma_{i,i+1}$, right up to the value $I_i \approx 2I_0$. There-

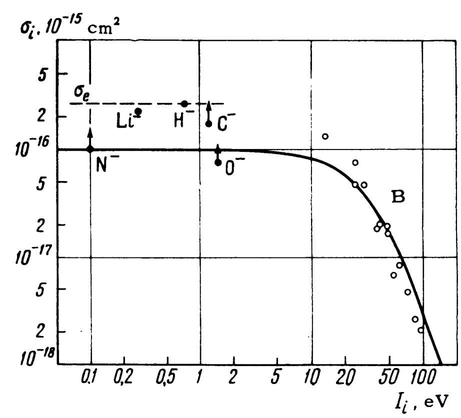

FIG. 4. Cross section for the loss of a single electron σ_i as a function of I_i in helium. The value of σ_i for a negative ion is denoted by a black dot and the [chemical] symbol for the ion (for $q = q_L$), with the addition of an arrow for $q = q_L - 2$ (see text). The values of σ_i in the range 10 eV < I_i < 100 eV for positive ions [7] are denoted by open circles. B (continuous) is a theoretical curve; [14] σ_e (dashed line) is the experimental value of the electron scattering cross section σ_e for E_e = 20 eV. [16]

fore, in addition to experimental points, Fig. 4 includes the theoretical dependence of σ_i on I_i, obtained in the Born approximation.[14] In the $I_i \gtrsim I_0$ range, this curve has been calculated for hydrogen-like ions. However, both theory[14] and experiment[7] show that the results of the calculation can be used also to estimate the cross sections for the loss of an electron from the L-shell. In the $I_i < I_0$ region, the Born approximation becomes identical with the free-collision approximation, which is valid for the loss of electrons from any shell.[14]

The dependence of σ_i on I_i for negative ions is not monotonic. However, in the majority of cases, the experimental points lie above the theoretical curve calculated in the Born approximation and, as expected,[15] they do not exceed the total cross section for the scattering of a free electron of velocity $v = 2.6 \times 10^8$ cm/sec by a helium atom: $\sigma_e = 2.5 \times 10^{-16}$ cm^2.[16] It is known that the correct value of the scattering cross section for electrons of this velocity is obtained only when the polarization of the atom is taken into account. We may expect that, at low values of I_i approaching zero, the correct theoretical value of the electron-loss cross section is again obtained only if the polarization is allowed for. It is possible that the experimentally observed nonmonotonic dependence of σ_i on I_i is due to the influence of the polarization of atoms, the magnitude of which depends on the structures of colliding particles.

Apart from the cross sections for the loss of one electron σ_i, we used the experimental data to calculate the cross sections for the loss of a pair of electrons[7]

$$\sigma_i^{(2)} = \frac{2}{q(q-1)} \sum \frac{s(s-1)}{2} \sigma_{i,\,i+s}.$$

The values of $\sigma_i^{(2)}$ are given in Fig. 5 as a function of $I_i^{(2)} = I_i + I_{i+1}$, which is the sum of the binding energies of the first and second electrons which are being removed. Figure 5 gives also the values of $\sigma_i^{(2)}$ for positive ions[7] (open circles). As is evident from this figure, the values of $\sigma_i^{(2)}$ for negative ions (black dots) lie on the continuation of the dependence of $\sigma_i^{(2)}$ on $I_i^{(2)}$, obtained for positive ions.

It should be mentioned that the values of the ratios

$$\sigma_i^{(2)} / \sigma_i \approx [2/(q-1)]\sigma_{i,\,i+2} / \sigma_{i,\,i+1}$$

which govern the average probability of the loss of a second electron,[7] are, for all negative ions (including H^-), close to the corresponding values for positive ions and they amount to 0.1—0.2 (for the case $q = q_L$). Therefore, the relatively large

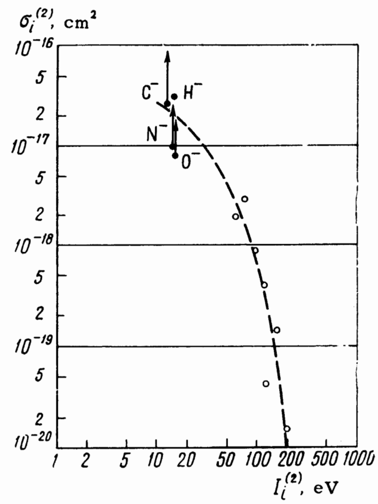

FIG. 5. Cross section for the loss of a pair of electrons $\sigma_i^{(2)}$ as a function of $I_i^{(2)} = I_i + I_{i+1}$ in helium. The notation of experimental points is the same as in Fig. 4.

cross section, $\sigma_{-1,1}$, for the loss of two electrons by C^-, N^- and O^- ions, compared with the cross section for H^-, can be explained by an increase in the number of electrons $(q-1)$ in the corresponding neutral atoms.

We finally conclude that the consideration of the various processes of interaction between negative ions and matter at $v > v_0$ shows that the weak binding of the outer electron to the ion core, characteristic of negative ions, has practically no effect on the absolute and relative values of the characteristics of the interaction of these ions with matter.

4. CROSS SECTIONS FOR THE FORMATION OF NEGATIVE IONS AND THE FRACTION OF THESE IONS IN THE EQUILIBRIUM CHARGE DISTRIBUTION

The experimental data obtained in the present investigation on the values of the cross sections for the loss and capture of electrons, $\sigma_{1,-1}$, $\sigma_{-1,-1+s}$, and $\Phi_{i,\infty}$—the fractions of ions with the charge i in the equilibrium distribution—made it possible to calculate the cross sections for the formation of negative ions $\sigma_{0,-1}$ from neutral atoms. For this purpose, we used the formula

$$\sigma_{0,-1} = \frac{\Phi_{-1,\infty}}{\Phi_{0,\infty}} \sum \sigma_{-1,\,-1+s} - \frac{\Phi_{1,\infty}}{\Phi_{0,\infty}} \sigma_{1,-1}.$$

The obtained values of the cross sections $\sigma_{0,-1}$ and $\sigma_{1,-1}$ are given in Figs. 6 and 7. The same figures include the experimental data on analogous cross sections obtained by Fogel' et al.[8-10] The arrows in these figures indicate the positions of

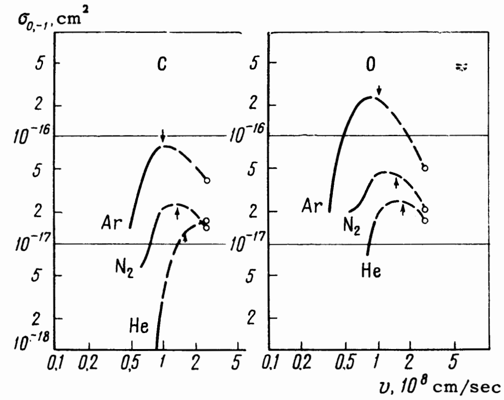

FIG. 6. Cross sections for the capture of a single electron $\sigma_{0,-1}$ by C and O atoms in He, N_2, Ar as a function of v. o— values of $\sigma_{0,-1}$, obtained in the present investigation; the continuous curves are the results of measurements reported in [10]. The arrows indicate the expected positions of the maximum v_{max}.

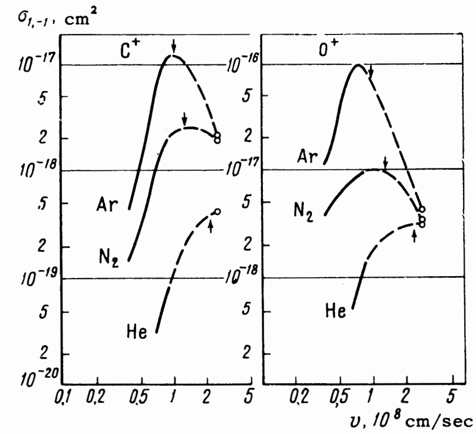

FIG. 7. Cross sections for the capture of two electrons $\sigma_{1,-1}$ by C+ and O+ ions in He, N_2, and Ar as a function of v. o— Values of $\sigma_{1,-1}$ obtained in the present investigation; the continuous lines are the results of measurements reported in [8,9]. The arrows indicate the expected positions of the maximum v_{max}.

the velocities v_{max} at which $\sigma_{0,-1}$ and $\sigma_{1,-1}$ should reach their maximum values. The values of v_{max} were estimated from the well-known relationship $a|\Delta E|/hv_{max} = 1$,[17] in which a = 3 Å for the capture of one electron,[10] and a = 1.5 Å for the capture of two electrons.[9] It is evident from Figs. (6) and (7) that the results of the present investigation do not contradict the data obtained at lower velocities and the conclusions of Fogel' et al. on the positions of the maxima in the dependences of the cross sections on the velocity.

The data obtained on the cross sections for the capture of one and two electrons allow us to estimate the importance of these processes in the formation of $\Phi_{-1,\infty}$ negative ions in the equilibrium charge distribution. Such an estimate shows that for v = 2.6 × 10^8 cm/sec, about a third of the negative ions are formed as a result of the simultaneous capture of two electrons by positive ions.

The results of the measurements of the values of $\Phi_{-1,\infty}$ for carbon and oxygen ions are listed in the table, together with the corresponding data for hydrogen and lithium ions.[2,3] We can see that, in most cases, when I_i increases, the values of $\Phi_{-1,\infty}$ for negative ions increase. The highest values of $\Phi_{-1,\infty}$ are obtained when ions pass through light gases (hydrogen and helium), which is evidently due to the approach to the maximum values of the functions $\sigma_{0,-1}(v)$ and $\sigma_{1,-1}(v)$ at v = 2.6 × 10^8 cm/sec (cf. Figs. 6 and 7).

The maxima in the cross sections for the formation of negative ions in heavier media are reached at lower velocities. Consequently, the maximum of $\Phi_{-1,\infty}$ should also shift. In fact, according to the measurements of Fogel' et al.,[11] the maximum value of $\Phi_{-1,\infty}$ for oxygen ions in argon is obtained for v = 0.8 × 10^8 cm/sec. Therefore, in order to obtain the highest coefficient of transformation of positive into negative ions, we ought to pass a beam of ions through a medium in which the cross section for the formation of negative ions reaches its maximum at a given ion velocity.

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Ion	I_i, eV	H_2	He	N_2	Ar	Celluloid
H^-	0.75	0.014	0.010	0.006	—	0.004
Li^-	0.49	0.00035	0.00035	0.00015	—	—
C^-	1.25	—	0.007	0.003	0.0025	0.004
O^-	1.68	—	0.014	0.005	0.005	0.004

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