Loss of Two Electrons in Single Collisions Between Negative Hydrogen Ions and Molecules of Gases

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(Submitted to JETP editor, October 22, 1956)

J. Exptl. Theoret. Phys. USSR, **32**, 453-462 (March, 1957)

The effective cross sections for the loss of two electrons in single collisions between negative hydrogen ions and He, Ne, A, Kr and Xe atoms and H_2, N_2, and O_2 molecules were measured. The measurements were carried out in a mass spectrometer in the energy range from 5 to 40 kev.

INTRODUCTION

THE measurements of effective cross sections for the loss of several electrons ("stripping") in single collisions between ions and gas molecules have been made recently in connection with the use of this phenomenon for the production of ions with multiple charges. Some of the studies dealing with this phenomenon investigate the loss of electrons by singly charged positive ions[1-5]. There are several investigations[6-12] devoted to the measurement of effective cross sections for the loss of a single electron in collisions of negative ions with gas molecules. There is only one paper[13] which cites the data on the loss of two electrons by ions Cl^-, Br^-, I^-, Na^-, Sb^-, Bi^-, Sb_2^-, in collisions with He and A atoms and with H_2 and N_2 molecules.

This paper deals with the measurement of effective cross sections σ_{-11}* for the loss of two electrons by the negative hydrogen ions with energies of 5 to 40 kev in single collisions with He, Ne, A, Kr, Xe atoms and with H_2, N_2, and O_2 molecules. The measurement of this cross section for the negative hydrogen ions is of considerable interest because these cross sections can be compared with electron loss cross sections for other particles with the same electron shell structure (He, Li^+). Furthermore, the knowledge of the cross section σ_{-11} enables one to compare it with the single electron loss cross section σ_{-10} as well as with the cross section σ_{-11} for the double electron capture by a proton.

DESCRIPTION OF THE EXPERIMENTAL SETUP AND THE MEASUREMENT METHODOLOGY

To obtain negative hydrogen ions, we used a previously developed method[14] for the transformation of positive hydrogen ions into negative ions by their passage through an ultrasonic stream of mercury vapor ("mercury vapor target").

The schematic diagram of the layout for the measurements described below is shown in Fig. 1. A detailed description of the apparatus for producing and shaping the beam of negative hydrogen atoms is given in Ref. 14.

By means of the magnetic analyzer *1*, the ion beam is separated from the protons and the neutral hydrogen atoms and is directed into the collision chamber *2*. The beam enters the collision chamber through a diaphragm 4 mm in diameter attached to the wall of a duct with an inner diameter of 5 mm and a length of 50 mm. The beam leaves the collision chamber through a duct having the same length and diameter as the inlet duct. The effective length of the collision chamber is 10 cm. The currents of the beam which entered and left the collision chamber were measured by means of Faraday cylinders 4 and 5, magnetically controlled. The current of the H_1^- ion beam entering the collision chamber was in the 10^{-10} to 5×10^{-9} A. range. The highest value of the current was obtained for the conversion $H_1^+ \rightarrow H_1^-$ with energies from 20 to 30 kev. To obtain H_1^- ions with energies of 5 kev, it was necessary to use the conversion $H_2^+ \rightarrow H_1^-$, because the ion gun focused poorly the primary beam of positive ions for this energy. The H_2^+ ion concentration was low in the primary beam of the high frequency ion source and, as a result, the H_1^- ion current passing through the collision chamber in this case did not exceed 10^{-10} A.

The separation of the beam leaving the collision chamber into neutral, positive, and negative components was achieved by means of the electric field in the parallel plate condenser *6* having plates 80 mm long at a distance of 28 mm from each other. The measurement of the currents of the positive and

*σ_{ik} is the effective cross section of the process in which a particle with a charge *ie* is converted into a particle with a charge *ke*.

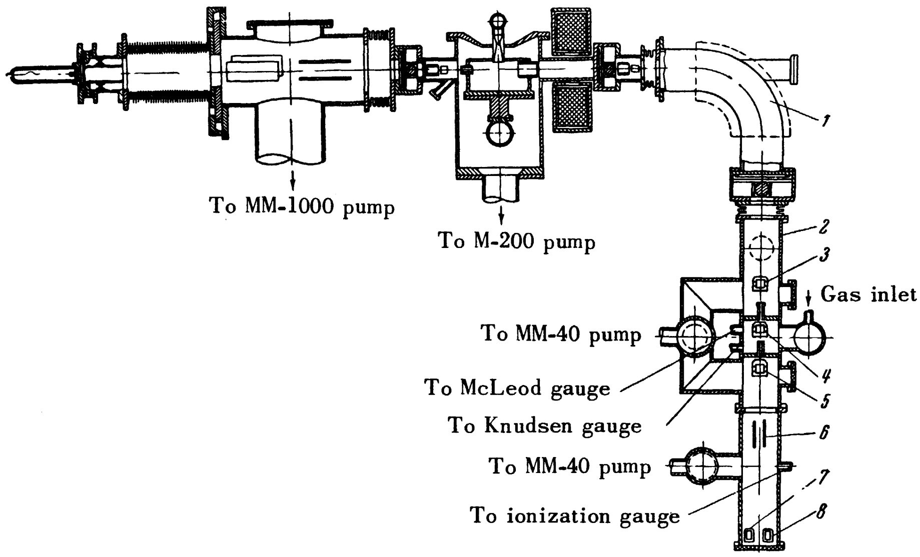

FIG. 1.

negative beam components was accomplished by means of two string electrometers connected to the Faraday cylinders *7* and *8*. The fibers of both electrometers were projected on two screens located one above the other so that the measurement of the beam component currents could be made simultaneously. This method eliminated the error due to random intensity fluctuations of the primary H_1^- ion beam. The pressure of the gas admitted into the collision chamber was measured with a Knudsen gauge, calibrated against a McLeod gauge. The calibration of the Knudsen gauge was made for all the gases in the investigation and was repeated several times during the measurements.

The collision and analyzer chambers were evacuated by separate MM-40 oil diffusion pumps. To freeze out the condensed vapors, liquid air traps were inserted into the collision chamber and into the space on either side of it. Besides, one more trap was located above the throat of the MM-40 pump which evacuated the collision chamber. The residual gas pressure in the collision chamber was 10^{-5} mm Hg.

The mass spectroscopic method fully described in Refs. 1, 15, 16 was applied to the determination of the effective cross sections σ_{-11}.

From the solutions of the differential equations [see Eq. (3) in Ref. 16), determining the beam composition produced as a result of the negative hydrogen ions passing through the gas in the collision chamber, (n = the number of gas atoms in 1 cm^3, L = effective length of the collision chamber), the formula is

$$I^+/I^- = \sigma_{-11}nL + \frac{1}{2}[\sigma_{-10}\sigma_{01} + \sigma_{-11}(\sigma_{-10} + \sigma_{-11} - \sigma_{10} - \sigma_{1-1})](nL)^2, \quad (1)$$

where I^+ and I^- — the currents of the protons and negative hydrogen ions in the beam emerging from the collision chamber. The dependence of I^+/I^- on the gas pressure in the collision chamber can be written in the following form:

$$I^+/I^- = \varkappa p + \Omega p^2, \quad (2)$$

where

$$\varkappa = \sigma_{-11}L/kT, \quad \Omega = \frac{1}{2}[\sigma_{-10}\sigma_{01} + \sigma_{-11}(\sigma_{-10} + \sigma_{-11} - \sigma_{10} - \sigma_{1-1})](L/kT)^2. \quad (3)$$

During the investigation of the relationship between I^+/I^- and pressure we observed two cases: in the first case the graph of the I^+/I^- function is linear up to pressures on the order of $2 + 3 \times 10^{-4}$ mm Hg. and it is only above these pressures that a marked deviation from linearity is observed; in the second case, beginning with pressures close to p_ϕ (p_ϕ — the residual gas pressure in the collision chamber) one observes a parabolic relationship between I^+/I^- and p. Evidently, in the first case with pressures p 10^{-4} mm Hg the assumption $(\Omega/\kappa)p \ll 1$ is satisfied and formula (2) can be written in the form:

$$I^+ / I^- = \varkappa p. \qquad (4)$$

In the second case even with the smallest pressures of the admitted gas the above condition is not satisfied.

As an illustration, Fig. 2 shows the graph of I^+/I^- as a function of pressure for H_1^- ions with 30 kev energy passing through hydrogen; fig. 3 — the corresponding graph for H_1^- ions with 20.6 kev energy passing through argon. In plotting these graphs the loss of two electrons by H_1^- ions during collisions with the residual gas molecules in the collision chamber was taken into account. This was accomplished by plotting the values of $p - p_\phi$ and $(I^+/I^-) - (I^+/I^-)_\phi$ abscissas and ordinates respectively, where $(I^+/I^-)_\phi$ -is the value of the (I^+/I^-) ratio at the residual gas pressure.

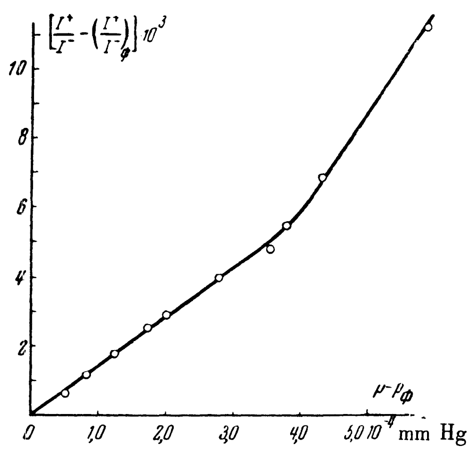

FIG. 2.

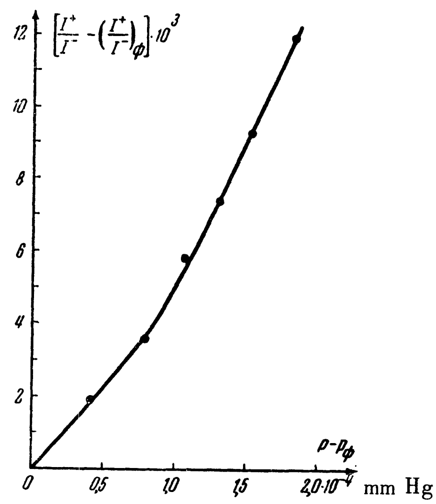

FIG. 3.

In the case where a linear portion was observed in the I^+/I^- graph, the cross section σ_{-11} was determined from this linear portion by the method of least squares. It is to be noted that if one determines σ_{-11} from all the points of the $I^+/I^- = f(p)$ curve which fit a parabola, then the calculated value of σ_{-11} does not differ significantly from the value calculated from the linear part of the graph.

If the linear portion was not present in the graph $I^+/I^- = f(p)$ over the range of measured pressures, then the cross section σ_{-11} was determined by the method of least squares according to formula (2). For the rapid selection of points satisfying this formula it is expedient to represent it in the form

$$(I^+/I^-)/p = \varkappa + \Omega\, p. \qquad (5)$$

As can be seen from formula (5), the value $(I^+/I^-)/p$ is a linear function of p in the range of pressures for which formula (2) holds. The cross section σ_{-11} is determined from formula (5) by the method of least squares.

The method used by us for the measurement of cross sections σ_{-11} has inherent systematic errors associated with the effect of pressure and composition of the residual gas in the apparatus on the value of the measured cross section,* with the unequal scattering of negative hydrogen ions and protons in the collision chamber, and with the unequal weakening of the negative hydrogen ion and proton beams during their passage from the collision chamber to the Faraday cylinders of the analyzer. As for the error associated with the presence

*This error was detected and discussed by us in Ref. 17.

of residual gas in the path of the beam, we were able to show by investigating dependence of the measured value of σ_{-11} on the value of $(I^+/I^-)_\phi$ that for our conditions this error was small and was within the limits of random experimental errors. To clarify the effect of unequal scattering of ions and protons in a hydrogen-filled collision chamber, cross sections σ_{-11} were measured with a diaphragm 2 mm in diameter attached to the end of the exit duct, and without the diaphragm. The diaphragm decreased considerably the solid angle for the particles leaving the collision chamber. It turned out that the cross section values σ_{-11} measured with and without a diaphragm in the exit duct were the same within the limits of experimental errors. Thus, the effect under consideration, for the case investigated by us, can not materially alter the measurement results. The correction for the unequal weakening of the negative and positive beams in the analyzer chamber for the cases when it can be calculated* does not exceed several tenths of a percent and is consequently unimportant. The random error of the individual cross section σ_{-11} measurement was ±15%. To ascertain the most probable error, some of the cross sections were measured 5-7 times; this error was ±7%.

The energy of the negative hydrogen ions was determined by the summation of the potential differential differences in the ion source and the accelerator tube. These potential differences were measured with electrostatic voltmeters, calibrated with a resistance voltmeter which was used in Refs. 15, 16. Such a method for determining the H_1^- ion energies could lead to an error if the protons lost an appreciable part of their energy in the mercury vapor stream while passing through it. Using the magnetic analyzer of our setup, we determined the proton energy loss during the passage through a beam of mercury vapor 2-3 × 10^{15} *atoms/cm²* thick, giving the optimum conversion $H_1^+ \to H_1^-$. It turned out that this energy loss was quite small and, as a consequence, the energy determination of the negative hydrogen ions by the above mentioned method is completely justified. The error associated with the measurement of the negative hydrogen ion energy was on the order of ±3%.

During the course of the present investigation, we obtained some information about the reliability of the steam-jet target. The steam-jet target worked more than 700 hours and during that time the boiler had to be taken apart only once in order to replace the nickel-constantan thermocouple, in which the constantan wire was corroded by the mercury vapor.

*To calculate this correction, it is necessary to know the cross section σ_{10} and σ_{-10} for the gas admitted to the collision chamber.

DATA AND DISCUSSION

The effective cross sections were measured for the loss of two electrons in collisions of H_1^- ions with energies of 5 to 40 kev with He, Ne, A, Kr, and Xe atoms, and with H_2, N_2 and O_2 molecules. The collision chamber was filled with palladium-filtered hydrogen, spectroscopically pure helium, neon, krypton, and xenon, oxygen with 0.9% impurities, argon with 0.3% impurities, and nitrogen with 0.3% impurities.

There are shown in Figs. 4 and 5 the curves of the relationship between the effective cross section σ_{-11} and the energy of the H_1^- ions for atomic and molecular gases. The value of σ_{-11} for each energy was obtained by averaging two measurements. As can be seen from Figs. 4 and 5, the effective cross section σ_{-11} for H_1^- ions in the investigated energy range grows with energy in neon, argon, xenon, nitrogen, and oxygen; in krypton the increase in σ_{-11} takes place up to an energy of 20.6 kev and thereafter remains constant to the end of the interval; in hydrogen, the cross section σ_{-11} remains constant over the entire interval; in helium, a maximum is indicated in the neighborhood of 10 kev, after which the cross section decreases monotonically with energy. The cross section σ_{-11} value varies from 3.3×10^{-17} cm² (in Kr, energy of 5.1 kev) to 1.8×10^{-16} cm² (in Xe, energy of 39.7 kev). One observes the dependence of the cross section σ_{-11} on the gas particle on colliding with which the H_1^- ion loses two electrons. In the case of molecular gases, the cross section σ_{-11} in oxygen and nitrogen is considerably greater than in hydrogen over the entire range of explored energies. In atomic gases, the cross section σ_{-11} is approximately the same at the beginning of the interval for all gases, but at the end of the interval the value is considerably greater for the heavy gases A, Kr, and Xe than for the light gases He and Ne.

As mentioned in the Introduction, there were no data in the literature on cross sections σ_{-11} for H_1^- ions. A comparison is only possible for the cross section data σ_{-11} of a number of heavy ions obtained by Dukel'skii and Fedorenko.[13] It is most expedient to compare the cross sections for the same ion velocities. Unfortunately, the data of Ref. 13 and ours differ widely with respect to the velocity intervals. The data on the cross sections σ_{-11} for Na^- ions came closest to the interval of velocities investigated by us. The comparison of cross sections σ_{-11} for H_1^- and Na^- ions is interesting also because, according to the most recent data[18], the electron affinity of for the sodium atom is equal to 0.84 ev, which is very close to that for hydrogen atoms (0.75 ev).

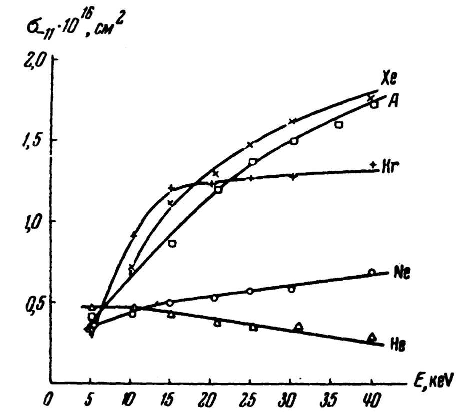

FIG. 4

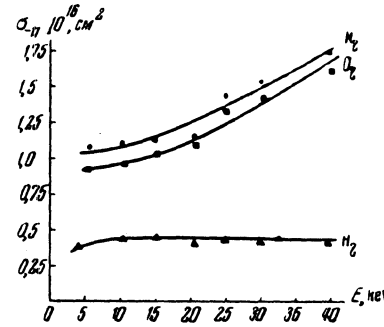

FIG. 5.

The curves of the relationship $\sigma_{-11} = f(v)$ for H^- and Na^- ions are shown in Fig. 6. By extrapolating the graph $\sigma_{-11} = f(v)$ for the H_1^- ion into the region of velocities where the corresponding curve of the Na^- ion is located, the cross section of the process $H_1^- \to H_1^+$ should decrease. It follows therefrom that in this region of velocities the cross section of the process $Na^- \to Na^+$ should be larger than the cross section of the $H_1^- \to H_1^+$ process. It was established in Ref. 17 that the capture cross section of two electrons by a positive ion increases with a decrease in the binding energy of the electrons in the particle losing them. One can assume that the same relationship holds for the stripping of two electrons in collision of negative ions with gas molecules. The fact that the cross section σ_{-11} is larger for the process $Na^- \to Na^+$ than for the process $H_1^- \to H_1^+$ confirms the stated assumption, since the binding energy of the stripped electrons in the Na^- ion is equal to the sum of the electron affinity and the first ionization energies of the Na atom, and is smaller than the corresponding quantity for the H_1^- ion. However, it is necessary to note that the paper gives data which contradict the assertion that the cross section σ_{-11} increases with a decrease in the binding energy of the stripped electrons. Evidently, an important role is played by the actual electron shell structure of the negative ion which is losing two electrons.

An opportunity presents itself to compare the cross sections for the loss of two electrons by H_1^- ions with the cross section σ_{-10} for the loss of a single electron by the same ions. The cross sections σ_{-10} for H_1^- ions in He, Ne, A, Kr, and Xe gases were measured by Stedeford and Hasted[10]. The corresponding measurements in hydrogen were made by Whittier[12]. The curves showing the relationship between cross sections σ_{-11} (solid curves) and σ_{-10} (dotted curves) and H_1^- ion energies are compared in Fig. 7. As can be seen from this figure, the effective cross section for the loss of two electrons in the investigated energy range is considerably smaller than the effective cross section for the loss of a single electron. The ratio σ_{-11}/σ_{10} for H_2, A, Kr, and Xe decreases with a decrease in energy, remains approximately constant over the entire energy range for Ne, and goes through a minimum for He at 15 kev. From this data given below, one can see the limits within which the ratio σ_{-11}/σ_{10} changes for various gases in passing from higher to lower energies of H_1^- ions.

The values cited in the Table show that in evaluating the intensity reduction of the H_1^- ion beam, it is sufficient (because of inelastic collisions with gas molecules) to account for the loss of a single electron only. Using these data, one can evaluate the errors which arise as a result of not taking into account the process of double electron loss by negative ions during the measurement of the effective cross section of a single electron loss by the method

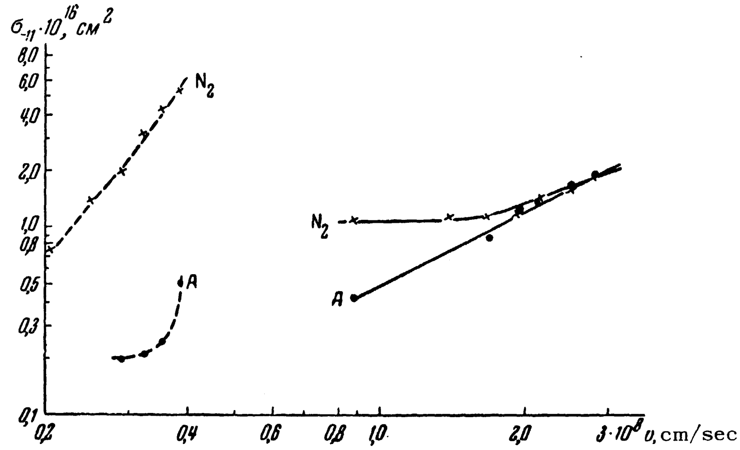

FIG. 6. Solid lines – H_1^- ions; dotted – Na^- ions.

of collecting slow ions and electrons produced in the gas by the passage through it of a beam of fast particles.*

Gas	σ_{-11}/σ_{10}
H_2	0.12-0.06
He	0.07-0.09
Ne	0.08
A	0.07-0.03
Kr	0.04-0.016
Xe	0.04-.02

By the method of collecting slow particles, one actually measures the cross section $\sigma = \sigma_{-10} + 2\sigma_{-11}$, from which the true cross section of a single electron loss $\sigma_{-10} = \sigma[1 - 2(\sigma_{-11}/\sigma_{-10})]$. The ratio $\sigma_{-11}/\sigma_{-10}$ was given above. As can be seen from the given data, the systematic error in Ref. 10 associated with the presence of the double electron loss is not large and in the worst case (He) does not exceed 20%. However, one must keep in mind that with the increase in ionic energies, because of the increase in the ratio $\sigma_{-11}/\sigma_{-10}$ with energy, this error may become quite considerable, and the use of the method of collecting slow particles for measuring cross sections σ_{-10} becomes inadmissible.

In spite of the large differences in the cross-section values of σ_{-11} and σ_{-10}, the nature of their dependence on the H_1^- ion energies is the same in the interval under study. This is particularly noticeable for the curves $\sigma = f(\varepsilon)$ in He and Ne. It is interesting to note that for single and double electron capture cross sections in hydrogen and helium (see Figs. 3 and 4 in Ref. 16), the curves $\sigma = f(\varepsilon)$ are different in approximately the same energy range.

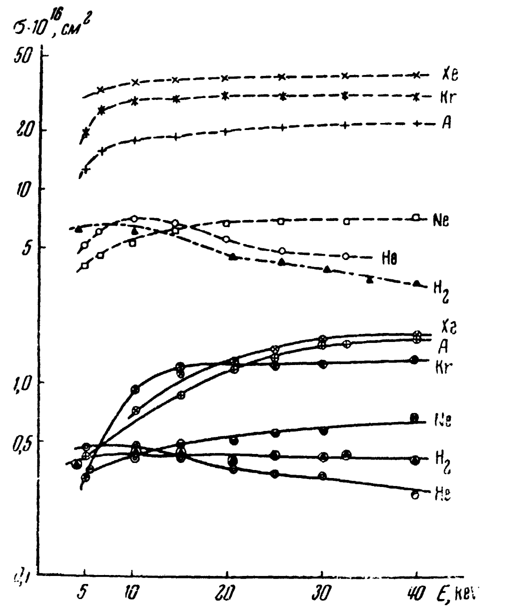

FIG. 7.

The experimental data for single [7,10] and double electron [16,17] capture cross sections are in definite agreement with the adiabatic hypothesis of Massey[19]. The sequence of maxima on the graphs of cross sections versus ion energy follows from the well-known Massey condition [see Eq. (8) in Ref. 16] with the assumption that the collision parameter differs little for different ion-molecule pairs, but that its value for a single electron exchange is considerably larger than for a double electron exchange. Assuming also that for double electron loss processes the collision parameter is approximately the same for different

*It was by this very method that Stedeford and Hasted[10] measured the cross section σ_{-10} in inert gases.

pairs, and having in mind that the resonance defect (the sum of the ionization potential and electron affinity of the hydrogen atom) is the same for all collision processes between H_1^- ions and atoms of inert gases, it is to be expected that the cross section maximum on the energy relationship graphs should be observed for all these processes at the same energy. As can be seen in Fig. 4, the cross section σ_{-11} maximum for helium is at an energy of about 10 kev, whereas for the other inert gases it is at an energy greater than 40 kev. In some contradiction with the adiabatic hypothesis is also the fact that for small energies the cross section σ_{-11} is quite large. Hasted [7,9] also points out the discrepancy between his data on the single electron loss cross sections and the adiabatic hypothesis. The paper by Bates and Massey [20] gives some reasons for the inapplicability of the adiabatic hypothesis to the processes for the single electron loss. A sounder judgment as to the existence of an adiabatic region in two electron loss processes will be possible after the cross section σ_{-11} measurements have been made with smaller energies than those in the present investigation.

It is possible to compare the electron loss cross sections for helium and other "helium-like" particles (H^- and Li^+). As has been already mentioned, such a comparision is interesting because of the similarity in the electronic shell structure of these particles.

Krasner [21] measured the sums of effective cross sections $\sigma_{01} + \sigma_{02}$ for the loss of one and two electrons by atoms of helium with energies from 100 to 450 kev in collisions with helium atoms and molecules of hydrogen and air. Using the data of our work and those of Refs. 10, 12, the sum $\sigma_{-10} + \sigma_{-11}$ of effective cross sections for the loss of one and two electrons in hydrogen and helium can be calculated for negative hydrogen ions. For Li^+ the sum of cross sections in air can be calculated from the data of Ref. 2.

In Fig. 8 are shown the curves of the relationship between the $\sigma_{01} + \sigma_{02}$ cross sections and velocities for helium atoms and the corresponding graphs of the cross section sums $\sigma_{-10} + \sigma_{-11}$ for negative hydrogen ions and $\sigma_{12} + \sigma_{16}$ for Li^+ ions. As is seen from the drawing, for the same velocities, the electron loss cross sections by particles of the isoelectronic sequence H_1^-, He, Li^+ increase with a decrease in the binding energy of the lost electrons. A fact that calls attention to itself is that electron loss cross sections by these particles in the same velocity interval have a different dependence on velocity namely, the electron loss cross sections by H_1^- ions decrease; for helium atoms these cross sections increase slowly, and for Li^+ ions-increase rapidly with an increase of the particle velocities. One might suppose that the comparison of the electron loss cross sections by different particles would be accomplished most expediently for the same values of the parameter $\gamma_i = v_i/v$, where v_i – the orbital velocity of the stripped electron in the moving particle, calculated

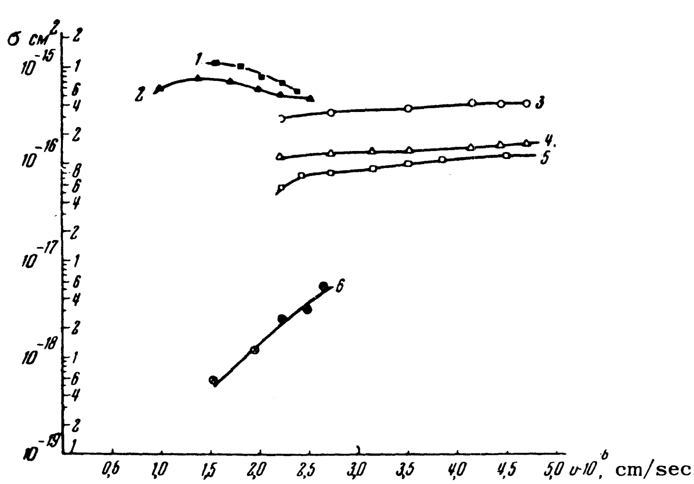

FIG. 8. 1 – $(\sigma_{-10} + \sigma_{-11})$ H^- in H_2; 2 – $(\sigma_{-10} + \sigma_{-11})$ H^- in He; 3 – $(\sigma_{01} + \sigma_{02})$ He^0 in air; 4 – $(\sigma_{01} + \sigma_{02})$ He^0 in He; 5 – $(\sigma_{01} + \sigma_{02})$ He^0 in H_2; 6 – $(\sigma_{12} + \sigma_{13})$ L_7^+ in air

from the formula ($v_i = 2V_i/m)^{1/2}$ (V_i – binding energy of the electron in the particle), and v – particle velocity.

Krasner[21] attempted to represent his results as well as those of Montague[22] and Kanner[23] as a function of the parameter γ_i. For the case of a moving helium atom the value of γ_i refers to either of the two 1s electrons. For this case, as can be shown easily, the electron binding energy is equal to the half-sum of the first and second ionization potentials of helium. In constructing the graph of the effective electron loss cross sections for helium and hydrogen as a function of the parameter γ_i, Krasner decreased the effective electron loss cross sections for helium atoms by a factor of two, because of the fact that an atom of helium has two 1s electrons. The points of the function $\sigma_{01} = f(\gamma)$ thus obtained for hydrogen and helium atoms moving in air and hydrogen fell on two smooth curves, from which Krasner drew the conclusion that the effective electron loss cross section, calculated for the equivalent removable electron, is a function of the parameter γ_i only. However, this conclusion has no general significance because, for other particles having only 1s electrons, the effective electrons loss cross section values do not lie on the curves plotted by Krasner; this is illustrated in Fig. 9 where, alongside the curves for the effective electron loss cross sections for hydrogen and helium atoms σ_{01} and $\frac{1}{2}\sigma_{01}$, the curves for $\frac{1}{2}\sigma_{-10}$ and $\frac{1}{2}\sigma_{12}$ are shown, corresponding to the cross sections from Refs. 2, 12 for H_1^- and Li^+ ions. In an article which appeared recently[24] it is stated that the data on the cross sections σ_{12} for He^+ ions do not fit the Krasner curve. Thus, it is impossible as yet to display a universal parameter which would determine the electron loss cross sections even for particles with the same electronic shell structure.

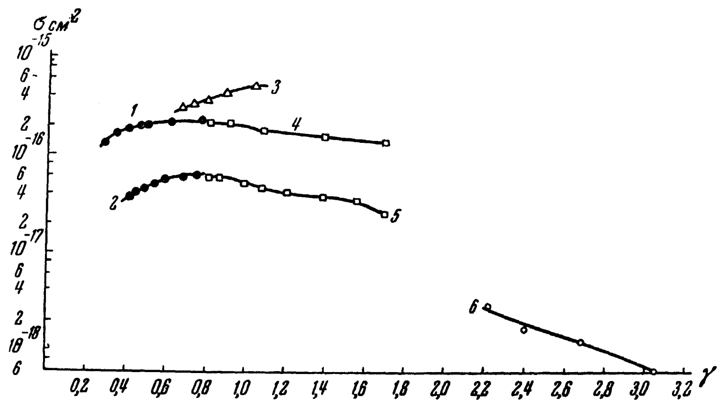

FIG. 9. 1 – $\sigma_{01}H^0$ in air; 2 – $\sigma_{01}H^0$ in H_2; 3 – $\frac{1}{2}\sigma_{-10}H^-$ in H_2; 4 – $\frac{1}{2}\sigma_{01}H^0$ in air; 5 – $\frac{1}{2}\sigma_{01}He^0$ in H_2; 6 – $\frac{1}{2}\sigma_{12}Li_7^+$ in air.

It is of some interest to compare the cross sections for the loss of two electrons by H_1^- ions measured in this investigation and the cross sections for the capture of two electrons by protons measured in Ref. 16. This comparison shows that the cross section σ_{1-1} is smaller than the cross section σ_{-11} for the entire investigated range of energies and for all investigated gases. For the gases He, Ne, A, N_2 and O_2 the ratio $\sigma_{1-1}/\sigma_{-11}$ does not exceed 7% and changes little with the energy of ions; for hydrogen the maximum value of this ratio is 30%.

The dependence of the ratio $\sigma_{1-1}/\sigma_{-11}$ on the parameter γ for Ne and H_2 is shown in Fig. 10 (the binding energy of either of the two electrons in the negative hydrogen ion was set equal to half the sum of the

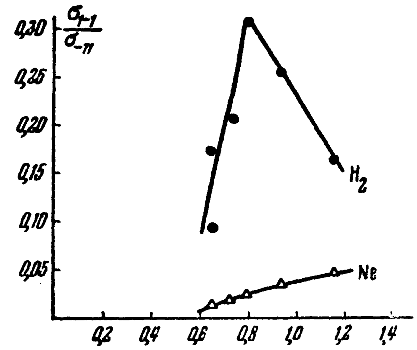

FIG. 10.

ionization potential and electron affinity of the hydrogen atom). The experimental data for the single electron exchange processes show (see Ref. 25) that the ratio σ_{10}/σ_{01} for hydrogen particles and the ratio σ_{21}/σ_{12} for helium particles is equal to one for $\gamma \approx 1$, *i.e.*, the cross sections for the capture and loss of a single electron are equal for a particle velocity equal to the orbital velocity of the electron. As is seen from Fig. 10, a similar situation does not take place for two-electron exchange processes.

D. V. Pilipenko participated in the investigation.

In conclusion the authors consider it a pleasant duty to express their thanks to prof. A. K. Val'ter for his constant interest in the investigation.

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Translated by H. Kruglak
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