IONIZATION OF GASES BY FAST HYDROGEN ATOMS AND BY PROTONS

E. S. SOLOV'EV, R. N. IL'IN, V. A. OPARIN and N. V. FEDORENKO

Leningrad Physico-technical Institute, Academy of Sciences, U.S.S.R.

Submitted to JETP editor July 21, 1961

J. Exptl. Theoret. Phys. (U.S.S.R.) **42**, 659-668 (March, 1962)

Ionization of H_2, N_2, He, Ne, Ar, and Kr gases by 10—180 keV fast hydrogen atoms and protons was studied. The ionization cross section, "stripping" cross section for fast hydrogen atoms, and cross section for formation of slow ions with various e/m ratios were measured. The cross sections for "stripping" of hydrogen atoms and for ionization of atoms and molecules by protons are compared with theoretical data obtained using the Born approximation. Factors that influence the ratios of various ionization cross sections and the positions of the maxima in the energy dependences of the cross sections are discussed.

INTRODUCTION

THE present paper reports the results of systematic measurements of cross sections for "stripping" of fast hydrogen atoms and cross sections for ionization of several gases by hydrogen atoms and by 10—180 keV protons, as well as cross sections of formation of slow ions with various e/m ratios. Ionization processes occurring on collision of protons and fast hydrogen atoms with various gas atoms and molecules have been studied by many workers.[1-15] Nevertheless further systematic work under identical and well-defined experimental conditions is useful for verification of earlier results and for filling in some important gaps. Among such gaps is the ionization of inert gases, nitrogen, and molecular hydrogen by fast hydrogen atoms; the available data on ionization of H_2 by H atoms cover only the energy range below 60 keV.[5,6]

As in earlier work,[1-4] we used the well-known capacitor method, supplemented by mass analysis of slow ions.

The 10—180 keV energy range is of special importance because in it the cross sections for various ionization processes reach their maximum values.[1-4] Information on cross sections in a wide range of energies is also of interest for plasma work. The present paper contains, therefore, some published data on "stripping" of atoms[11,12] and ionization by protons,[7-9] which, together with our results, cover the whole keV range (1 keV to 1 MeV). Theoretical curves are also given for the few cases where experimental data are available and cross sections can be calculated using the Born approximation.[16-18]

1. EXPERIMENTAL TECHNIQUE

The apparatus has been described earlier.[19-21] It is shown schematically in Fig. 1. A beam of fast atoms was obtained by charge exchange from a monoenergetic proton beam in a gas-filled chamber B. After leaving B the atomic beam was passed through the field of a capacitor C where charged particles were removed. The beam then entered a scattering chamber S. The chamber S was filled with the gas under investigation at a pressure of $(2-4) \times 10^{-4}$ mm Hg, which was sufficiently low to obtain single collisions. The pressure outside B and S was kept low (at a level of 5×10^{-6} mm Hg) by differential pumping. In the scattering chamber S there was a capacitor M which collected the slow ions and electrons formed in the gas. The saturation currents of these particles gave the total cross section for formation of free electrons, σ_-, and the total cross section for

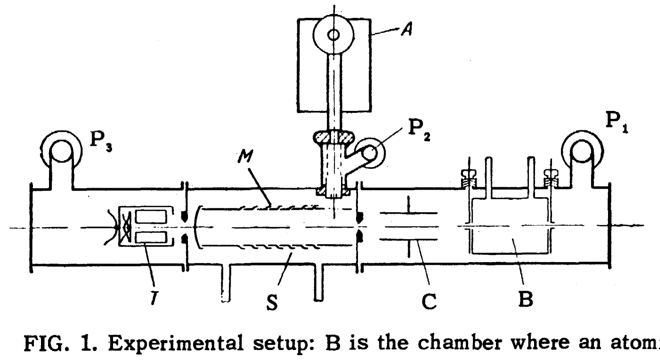

FIG. 1. Experimental setup: B is the chamber where an atomic beam is formed by charge exchange; C is the capacitor where charged particles are removed from the atomic beam; S is the scattering chamber; M is the measuring capacitor; T is the receiver of fast particles; A is the analyzer of slow ions; P_1, P_2, P_3, are pumps.

formation of slow ions, σ_+. Stray effects were avoided by suitable construction of the measuring capacitor M and by control tests described earlier.[19,20,22] Cross sections for formation of slow ions were measured with an analyzer A.

The intensity of the fast atomic beam was deduced from its heating effect by means of a special receiver T using thermistors. The atomic beam represented a current of 5×10^{-7}—2×10^{-8} A. The current flowing to the measuring electrodes of the capacitor M amounted to 10^{-10}—10^{-8} A. The measured cross sections σ_+ and σ_- are given by:

$$\sigma_- = \sigma_i + \sigma_l, \quad (1)$$

$$\sigma_+ = \sigma_i + \sigma_c. \quad (2)$$

The measured "stripping" cross section σ_l represents several inelastic processes leading to "stripping" of fast atoms, and the cross section σ_i represents several processes leading to the ionization of molecules. In the case of fast atoms σ_c represents the cross section for electron capture in the process $H \rightarrow H^-$. Fogel' et al[13] have shown that $\sigma_c \ll \sigma_i$, and therefore $\sigma_+ \approx \sigma_i$ and $\sigma_l = \sigma_- - \sigma_+$. In the case of fast protons $\sigma_l = 0$ and therefore $\sigma_- = \sigma_i$ and $\sigma_c = \sigma_+ - \sigma_-$.

Random errors in measurement of cross sections did not exceed ±15%, except in the case of σ_{H^+} and $\sigma_{N^{2+}}$ (Fig. 5) for which errors reached ±30%.

2. RESULTS

The results are shown in Figs. 2—7. All cross sections are given in cm^2 per gas molecule.

A. Cross Sections for Ionization of Gases by Protons, $\sigma_i(H^+)$. Figure 2a gives the energy dependence of the cross sections for the ionization of molecular hydrogen and nitrogen by protons, and Fig. 2b gives the data for proton ionization of inert gases.

Similar measurements were carried out earlier by Gilbody and Hasted[9] (molecular hydrogen and inert gases; protons of 0.4—40 keV energy), Fogel' et al[10] (molecular hydrogen, 12—37 keV), Keene[14] (helium and molecular hydrogen, 2—35 keV), Schwirzke[6] (molecular hydrogen, 9—60 keV), Hooper et al[7,8] (molecular hydrogen and nitrogen, inert gases; 150—1100 keV), and by members of our laboratory at energies of 5—180 keV (molecular hydrogen,[1] air,[3] inert gases[4]). All these results do not differ greatly from those given here. All the published curves lie slightly below or above our curves and the differences are within the experimental error, except for the cross sections obtained by Keene[14] for molecular hydrogen and helium, which were considerably lower than those of all the other workers.

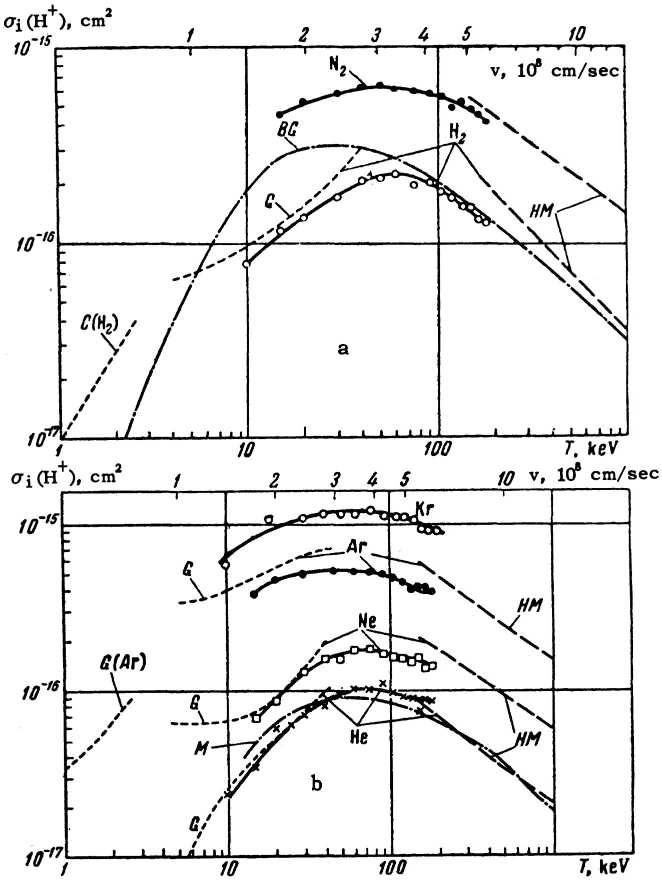

FIG. 2. Cross sections of ionization by protons: a) molecular hydrogen and nitrogen; b) inert gases. The continuous curves represent our results; HM are the data of Hooper et al.;[7,8] G are the data of Gilbody and Hasted;[9] BG is the theoretical curve of Bates and Griffing[16] for ionization of molecular hydrogen by protons; M is the calculated curve of Mapleton[18] for ionization of helium by protons.

Figure 2 gives curves obtained by Gilbody and Halsted[9] and by Hooper et al.[7,8] These curves give information on cross sections outside the energy range studied by us.

B. Cross Sections for Ionization of Gases by Fast Hydrogen Atoms, $\sigma_i(H)$. Figure 3a gives our energy dependences for cross sections of ionization of molecular hydrogen and nitrogen by fast hydrogen atoms. Figure 3b gives the corresponding data for inert gases. Published data are available only for ionization of molecular hydrogen; they were obtained by Schwirzke[6] (hydrogen atom energies of 9—60 keV) and by Curran and Donahue[5] (4—36 keV). Their curves are shown in Fig. 3a and agree with our data within the experimental error. The fine structure found by Curran and

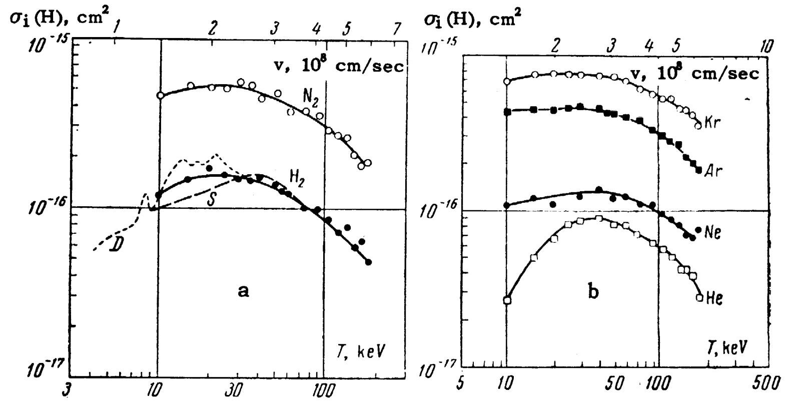

FIG. 3. Cross sections for the ionization by fast hydrogen atoms of: a) molecular hydrogen and nitrogen; b) inert gases. Continuous curves are our data; S represents Schwirzke's results; [6] D gives Curran and Donahue's results. [5]

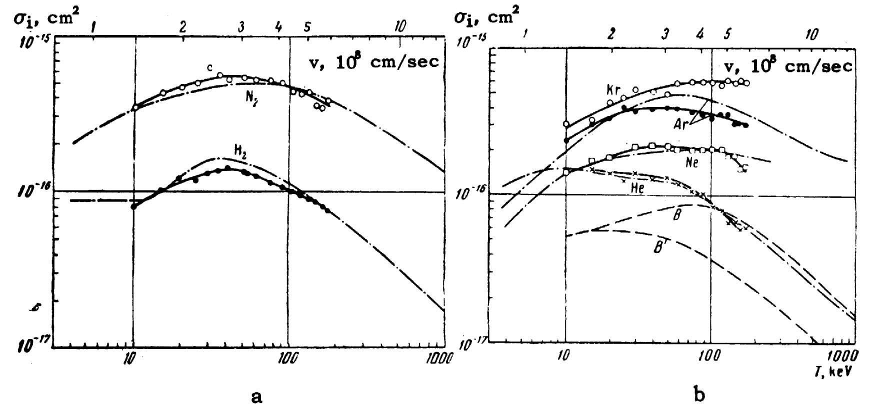

FIG. 4, Cross sections for "stripping" of fast hydrogen atoms in: a) molecular hydrogen and nitrogen; b) inert gases. Continuous curves are out data; chain curves represent the results of Stier, Barnett and Reynolds; [11,12] B are the results of Bates and Williams's calculation [17] for helium atoms with allowance for excitation; B′ is the Bates and Williams calculation for helium atoms which neglects excitation.

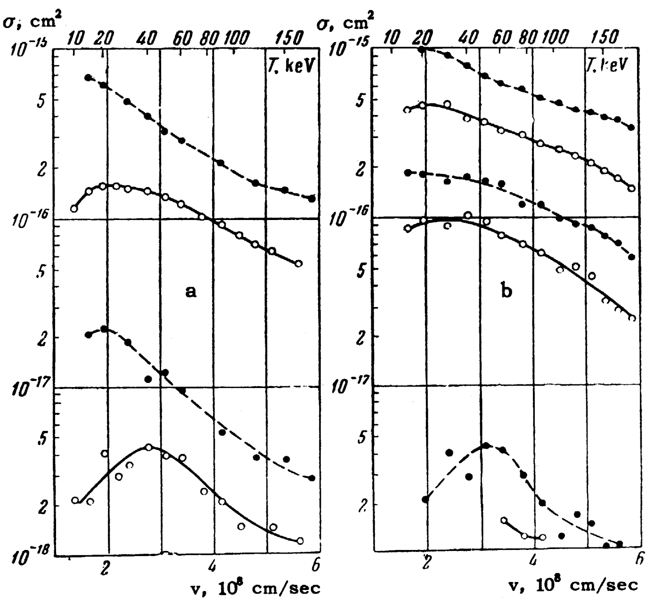

FIG. 5. Cross sections for formation of slow ions in: a) molecular hydrogen (the upper pair of curves represents formation of H_2^+, the lower pair – formation of H^+); b) molecular nitrogen (the upper pair of curves represents formation of N_2^+, the middle pair – formation of N^+, and the lower pair – formation of N^{2+}). The continuous curves give cross sections for incident hydrogen atoms, dashed curves represent the effect of protons.

Donahue and shown as a series of maxima on the $\sigma_i(H)$ curve was not observed because our experimental errors were greater than the heights of these maxima.

C. Cross Sections of "Stripping" of Fast Hydrogen Atoms, σ_l. Figure 4a shows energy dependences of the cross sections for "stripping" of fast hydrogen atoms in molecular hydrogen and nitrogen. Figure 4b gives corresponding data for "stripping" in inert gases. The cross sections of "stripping" of fast hydrogen atoms have also been measured by Curran and Donahue [5] (molecular hydrogen; hydrogen atom energies of 4–36 keV), Fogel' et al [13] (molecular hydrogen and nitrogen, inert gases; 5–40 keV), Schwirzke [6] (molecular hydrogen; 10–60 keV), Stier, Barnett and Reynolds [11,12] (molecular hydrogen and nitrogen, inert gases; 4–1000 keV). The curves obtained by Stier, Barnett and Reynolds are given in Fig. 4. For molecular hydrogen bombarded with 10–180 keV hydrogen atoms all the published data agree satisfactorily among themselves and with our results. For molecular nitrogen and inert gases the data of Stier and Barnett [11] are also in satisfactory agreement with our cross section. Fogel' et al [13] have reported "stripping" cross sections in nitrogen and inert gases which were 20–30% smaller than our results and those of Stier and Barnett. [11] For krypton the cross sections obtained by Fogel' are nearly 50% lower than our cross sections and those of Stier and Barnett.

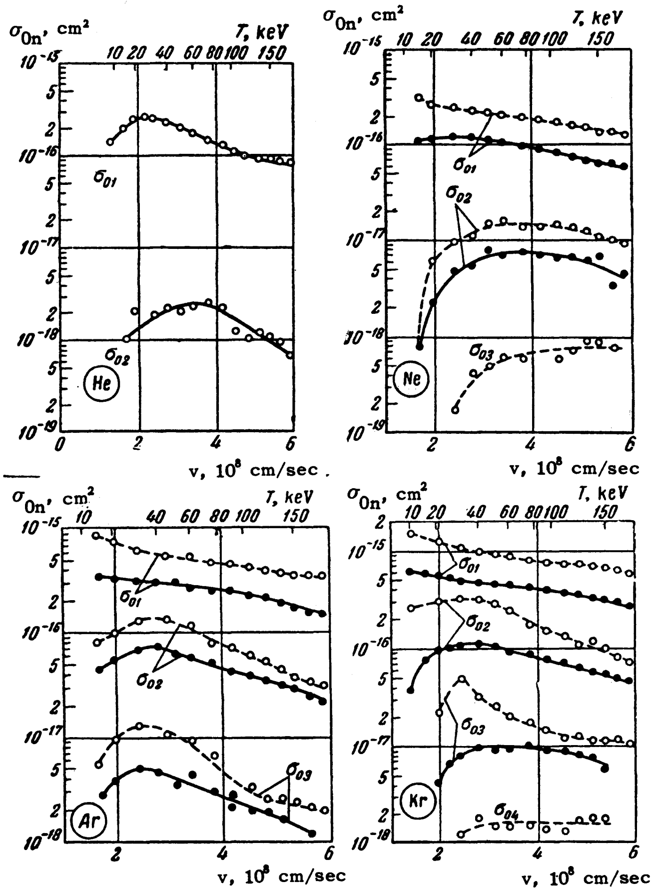

FIG. 6. Cross sections for the formation of slow ions in inert gases. Continuous curves represent formation by hydrogen atoms; dashed curves give the effect of protons.

These differences exceed the experimental error and they may be due to relatively strong scattering of protons produced by "stripping" of hydrogen atoms.

D. Cross Sections for Formation of Slow Ions with Various Charges. Figure 5 gives our cross sections for the formation of slow H_2^+ and H^+ ions in molecular hydrogen and of N_2^+, N^+, N^{2+} ions in molecular nitrogen. Figure 6 gives cross sections for the formation of ions in helium (He^+, H^{2+}), in neon (Ne^+, Ne^{2+}, Ne^{3+}), in argon (Ar^+, Ar^{2+}, Ar^{3+}) and in krypton (Kr^+, Kr^{2+}, Kr^{3+}, Kr^{4+}).

The cross sections for the formation of slow ions by protons are not due to mere ionization: each should be regarded as the sum of ionization and electron-capture cross sections; for molecular ions they are the sum of dissociative ionization and dissociative charge-exchange cross sections. In collisions of atoms with atoms there is practically no electron capture and the cross sections represent pure ionization. This complex nature of formation cross sections explains why they are somewhat larger for protons than for fast atoms when $v \leq v_0$, where $v_0 = e^2/\hbar = 2.2 \times 10^8$ cm/sec is the velocity of an electron in a Bohr atom.

An inspection of Fig. 5a indicates that in molecular hydrogen the number of slow protons does not exceed 2–4% of the total number of slow ions. In molecular nitrogen the proportion of atomic ions is somewhat higher. In helium the doubly-charged slow ions account for no more than 1.5% of the singly-charged ions (Fig. 6). In inert gases the cross sections for formation of slow ions with n missing electrons are approximately an order of magnitude greater than the cross section for formation of ions with $(n+1)$ missing electrons; this is true both for proton-atom and atom-atom collisions.

3. DISCUSSION

A direct comparison of experimental data with theory is possible only for a limited number of cases. For example, Bates and Griffing [16] calculated the cross section for the ionization of hydrogen atoms by protons using the Born approximation. They also reported an approximate method of calculating the ionization cross section for hydrogen molecules. The theoretical curve of Bates and Griffing is shown in Fig. 2a; it agrees satisfactorily with experimental results at energies above 60 keV. Mapleton [18] calculated the cross section for ionization of helium by protons, again using the Born method. Figure 2b indicates that the Mapleton curve agrees with our experimental results and, at higher energies, with those of Hooper et al.[8]

Bates and Williams [17] calculated the cross section for "stripping" of hydrogen atoms in helium using the Born approximation. They considered two cases; first, when a helium atom remains in its ground state after a collision, and second, when a helium atom suffers a transition to an excited state. Both theoretical curves are given in Fig. 4b. The curve which allows for excitation gives cross sections whose absolute values are close to experimental ones above 100 keV. At lower energies both these theoretical curves lie considerably below the experimental one and the discrepancy increases with energy decrease.

The relationship between the cross sections for ionization by protons and hydrogen atoms is of interest. Figure 7 shows typical ionization curves for helium. Calculations of ionization cross sections based on the Born approximation usually give larger values for protons than for fast hydrogen atoms.[16] An analysis of our experimental results confirms this relationship for all gases, but only at high velocities when $v > v_0$. At low velocities

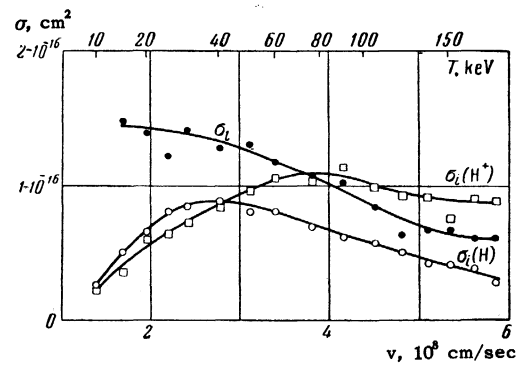

FIG. 7. Cross sections for the ionization of helium by hydrogen atoms, $\sigma_i(H)$, and by protons, $\sigma_i(H^+)$, and the cross section for "stripping" of hydrogen atoms, σ_l.

($v < v_0$) the cross section for the ionization by fast hydrogen atoms was always slightly higher than the cross section for protons; this indicates that the Born approximation is not valid at low velocities.

Another general characteristic of our results is the increase of cross sections of given processes with increase of the atomic number Z of the target atom; this can be easily seen in Figs. 2b, 3b, 4b, 6.

Let us consider the maxima on the "stripping" curves (σ_l) and the curves representing formation of singly charged inert-gas ions (σ_{01}) in the case of ionization by hydrogen atoms. The cross sections σ_l and σ_{01} represent purely a loss of one electron by a fast incident atom or an atom of gas. Figures 4b and 6 indicate that the σ_l and σ_{01} curves usually have a maximum at $v \geq v_0$. However, the cross section for "stripping" of fast atoms in helium and the cross section for the formation of singly charged argon and krypton ions increase continuously with incident atom velocity; their maxima may lie at $v < 1.5 \times 10^8$ cm/sec (for σ_l in helium the maximum lies at $v \approx 1.3 \times 10^8$ cm/sec[11]).

Until recently it has been assumed that for atomic particles with ionization potential close to that of the hydrogen atom, the ionization cross sections possess a maximum at $v \geq v_0$.[23] Maxima discovered at lower velocities can be accounted for qualitatively as follows. Ionization of atomic particles occurs by spatial intersection of atomic shells, as established by experiments on scattering accompanied by "stripping".[23] At velocities $v < v_0$ the colliding pair can be regarded as a quasi-molecule. The ionization potential of such a quasi-molecule is smaller for a collision of hydrogen atoms and inert gases than for the atoms considered in[23]. In the latter case we can assume that the quasi-molecular ionization potential approaches the ionization potential of the appropriate alkali atom. A strong reduction of the ionization potential may displace the ionization cross section maximum of the quasi-molecule into the region where $v < v_0$.

The same reasoning can be applied to explain the position of the maximum on the "stripping" curve of a fast hydrogen atom or the curve representing ionization of a gas. A quasi-molecule consisting of a proton and an inert-gas atom may be slightly more difficult to ionize than a quasi-molecule consisting of an atom of hydrogen and an inert-gas atom. Therefore, at velocities $v < v_0$ we have $\sigma(H)/\sigma(H^+) > 1$.

The probability of ionization of one or the other atomic particle after dissociation of the quasi-molecule depends on two factors. First is the energy factor, i.e., the binding of an electron in a given atom. The second is the statistical factor, i.e., the ratio of statistical weights of possible charged states. For example the statistical weight of the $Ar^+ + H$ state is much greater than that of the $Ar + H^+$ state. The effect of these two factors may produce competition between ionization processes which affects the position of a maximum of ionization cross sections for the two colliding particles.

Figures 4b and 6 show that the maximum on the curves representing formation of singly charged inert-gas ions on bombardment with fast hydrogen ions is displaced with increase of Z toward lower velocities while the maximum of the "stripping" curve is shifted toward higher velocities. In the case of "stripping" of hydrogen atoms this indicates that the ionization potential of these atoms is not the only parameter which affects the position of the maximum on the $\sigma_l(v)$ curve.

The position of the maximum on the curve showing the ionization cross section of a gas by hydrogen atoms, $\sigma_i(H)$, or by protons, $\sigma_i(H^+)$, presents a fairly complex problem because each of these cross sections is the sum of several partial cross sections representing losses of different numbers of electrons. Our results indicate that maxima on these curves lie at $v \approx (1-1.5)v_0$.

Let us consider the curves representing the cross sections of formation of slow argon and krypton ions by fast hydrogen atoms (Fig. 6). These cross sections represent pure ionization. For argon the maxima on the curves representing loss of two and three electrons, $\sigma_{02}(v)$ and $\sigma_{03}(v)$, occur at approximately the same velocity $v_{max} \approx v_0$. An analogous situation is found for ionization of krypton. Similar maxima for ionization by electron impact are always widely separated along the relative velocity axis. This difference between ionization by atoms and by electrons can be accounted for qualitatively. In electron-atom collisions loss of an electron by the atoms is the result of an interaction between the incident electron and the atomic electrons. In this interaction the transferred momentum is large compared with the initial momentum of atomic electrons and to remove each subsequent electron a considerably increase of the incident velocity is required. In atom-atom collisions the atomic "core" (nucleus) interacts with electrons of the second particle with hardly any change in its intrinsic momentum. The position of a maximum on the formation cross section curve is now governed by the ratio of the internal velocity of electrons of the second particle and the velocity of relative motion. Since inert-gas atoms lose mainly the outer p-electrons which all have similar velocities, the maxima on the σ_{0n} curves therefore occur at approximately the same velocities of relative motion; this is true when only a small number of electrons is lost on ionization.

The authors thank V. M. Dukel'skiĭ and O. B. Firsov for discussing the results and giving valuable advice.

[1] Afrosimov, Il'in, and Fedorenko, JETP **34**, 1398 (1958), Soviet Phys. JETP **7**, 968 (1958).

[2] Afrosimov, Il'in, and Fedorenko, ZhTF **28**, 2266 (1958), Soviet Phys. Tech. Phys. **3**, 2080 (1959).

[3] Il'in, Afrosimov, and Fedorenko, JETP **36**, 41 (1959), Soviet Phys. JETP **9**, 29 (1959).

[4] Fedorenko, Afrosimov, Il'in, and Solov'ev, Proc. IVth Intern. Conf. on Ionization Phenomena in Gases, held in Uppsala in 1959; Amsterdam, 1960, **1**, p. 47.

[5] R. Curran and T. M. Donahue, Phys. Rev. **118**, 1233 (1960).

[6] F. Schwirzke, Z. Physik **157**, 510 (1960).

[7] Hooper, McDaniel, Martin, and Harmer, Phys. Rev. **121**, 1123 (1961).

[8] Hooper, McDaniel, Martin, and Harmer, Abstr. Second Intern. Conf. on Electronic and Atomic Collisions, held at Boulder, USA in 1961; pp. 61-80.

[9] H. B. Gilbody and J. B. Hasted, Proc. Roy. Soc. (London) A240, 382 (1957).

[10] Fogel', Krupnik, and Safronov, JETP **28**, 589 (1955), Soviet Phys. JETP **1**, 475 (1955).

[11] P. M. Stier and C. F. Barnett, Phys. Rev. **103**, 896 (1956).

[12] C. F. Barnett and H. K. Reynolds, Phys. Rev. **109**, 355 (1958).

[13] Fogel', Ankudinov, Pilipenko, and Topolya, JETP **34**, 579 (1958), Soviet Phys. JETP **7**, 400 (1959).

[14] J. P. Keene, Phil. Mag. **40**, 369 (1949).

[15] J. H. Montague, Phys. Rev. **81**, 1026 (1951).

[16] D. R. Bates and G. Griffing, Proc. Phys. Soc. (London) **A66**, 961 (1953).

[17] D. R. Bates and A. Williams, Proc. Phys. Soc. (London) **A70**, 306 (1957).

[18] R. A. Mapleton, Phys. Rev. **109**, 1166 (1958).

[19] Fedorenko, Afrosimov, and Kaminker, ZhTF **26**, 1929 (1956), Soviet Phys. Tech. Phys. **1**, 1861 (1957).

[20] N. V. Fedorenko and V. V. Afrosimov, ZhTF **26**, 1941 (1956), Soviet Phys. Tech. Phys. **1**, 1872 (1957).

[21] Afrosimov, Il'in, Oparin, Solov'ev, and Fedorenko, JETP **41**, 1048 (1961), Soviet Phys. JETP **14**, 747 (1962).

[22] N. V. Fedorenko, ZhTF **24**, 2113 (1954).

[23] N. V. Fedorenko, UFN **68**, 481 (1959), Soviet Phys. Uspekhi **2** (**68**), 526 (1959).

Translated by A. Tybulewicz
107