SLOW-IONS PRODUCED IN GASES UPON PASSAGE OF FAST ATOM AND ION BEAMS

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The cross section σ_{0k} for production of slow ions with various charges in single collisions of fast Ne^0 atoms and Ne^+, Ne^{2+}, Ne^{3+} ions with Ne, Ar, Kr, Xe atoms and of fast Kr^0, Kr^+, Kr^{2+}, Kr^{3+} atoms and ions with Kr and Xe atoms was measured at accelerating potentials from 3 to 30 kv. The experimental data show that with increasing charge of the fast particle the cross section σ_{0k} increases as a rule. In atom-atom collisions slow ions are produced only by ionization and the cross section σ_{0k} continuously grows with increasing velocity of the fast ionizing particle. The main processes that determine the cross sections σ_{0k} and their energy dependence in ion-atom collisions are charge exchange and ionization with capture.

INTRODUCTION

WHEN fast atomic particles I^{n+} collide with atoms A of a gas, slow ions can be produced by three processes: pure ionization

$$I^{n+} + A \rightarrow I^{n+} + A^{k+} + ke, \tag{1}$$

charge exchange

$$I^{n+} + A \rightarrow I^{m+} + A^{k+} \qquad (m + k = n) \tag{2}$$

and ionization with capture*

$$I^{n+} + A \rightarrow I^{m+} + A^{k+} + (m + k - n)\, e. \tag{3}$$

The cross sections of processes (1)—(3) can be written in general form as σ_{0k}^{nm}, where the superscripts pertain to the initial and final charge states of the fast particle while the subscripts pertain to the charge state of the slow atomic particle. Since the ionization of atoms can also be accompanied by "stripping" of fast particles, the indices n, m, and k can range in principle from zero to the total number of electrons Z_I and Z_A in the shells of the colliding atomic particles, and to describe the processes one must have complete data on the charge states of both particles before and after collision. Data of this kind can be obtained only by the coincidence method.

Considering the limited capabilities of mass-spectrometric methods, which enable us to investigate separately the charge states of only fast or only slow secondary ions, we use a two-index system. The cross sections for the production of fast secondary ions will be designated by superscripts (σ^{nm}), while the cross sections for the formation of slow ions will be denoted by subscripts (σ_{0k}). In both cases, the first index denotes the initial charge state and the second the final state.

In our earlier papers [3,4] we reported measurements of the total cross sections for the formation of slow ions, denoted by σ_+, and the total cross section for the formation of free electrons (or the total ionization cross sections) σ_-, while in [5,7] we measured the charge-exchange cross sections σ^{nm} of differently charged ions. These measurements never determined the charge composition of the slow ions and the dependence of this composition on the charge of the fast atomic particles. The first qualitative data of this kind were obtained for Ar^+ and Ar^{2+} and for the atoms Ar and Kr. [8] The purpose of the present investigation was to determine the cross sections for the formation of slow ions of different charges (σ_{0k}), and to determine their connection with the charge of the fast atomic particle by comparison with our earlier data. [5-7]

The procedure used to measure the cross sections σ_{0k} was similar to that employed earlier, [2] but was modified somewhat. The choice of objects for the investigation was determined by the best conditions under which this procedure can be applied, when the mass of the fast ionizing particle does not exceed the mass of the gas atom. We investigated the following pairs: Ne^{n+}–Ne, Ne^{n+}–Ar, Ne^{n+}–Kr, Ne^{n+}–Xe and Kr^{n+}–Kr, Kr^{n+}–Xe. The charge n varied from 0 to 3, and the accelerated voltage V varied from 3 to 30 kv.

*The possible existence of processes of group (3) was first pointed out in [1,2].

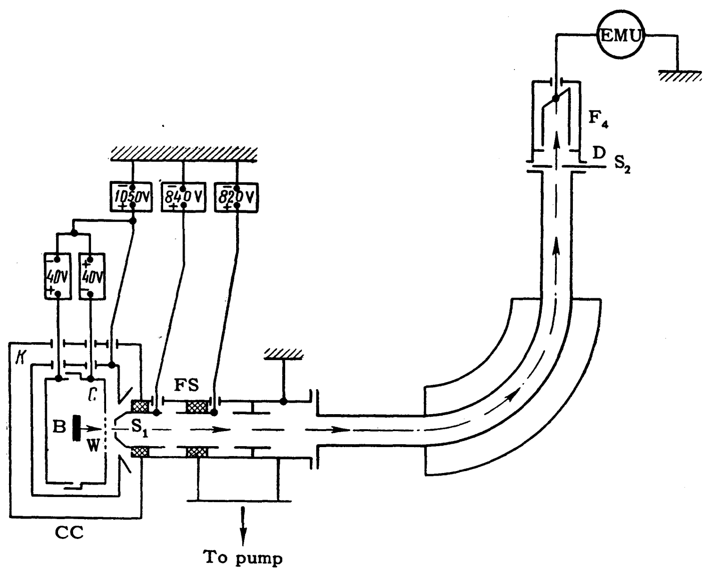

FIG. 1. Analyzer for slow ions.

1. MEASUREMENT PROCEDURE

The cross sections for the formation of slow ions of different charges were measured in the experimental setup described in [3,4], supplemented with a secondary-ion analyzer. A diagram of this analyzer is shown in Fig. 1. The slow ions were produced in a gas broached by a fast atom or ion beam B. They were extracted from this volume by a potential $V_C = \pm 40$ v applied to the plates of a capacitor C in a metallic container K insulated from the ground. To accelerate slow ions, an accelerating voltage $V_p = +1050$ v was applied to the container K and to the electrodes in it.* The ions then proceeded to the analyzer through a window W in the capacitor plates; the window was covered with a loose metal grid.

The potentials V_C and V_p and the potentials on the focusing system FS were determined by control experiments similar to those described in [2].

The analyzer was a sector type magnetic mass spectrometer with a deflection angle 90°, an average trajectory radius 150 mm, and a 10 mm gap between magnetic poles. The analyzer was connected to the grounded outer container of the collision chamber CC. The measuring part of the analyzer was also at ground potential. This was the principal difference between our method of measuring cross sections and the method described in [2], where the analyzer and the ion receiver connected to the input of a vacuum-tube electrometer were at approximately 1000 v relative to ground, so that the sensitivity of the electrometer could not be fully utilized.

The beam of slow ions entered the analyzer through a slit S_1 (1.5×10 mm). At the output of the analyzer was a slit S_2 (2×14 mm), which served to determine exactly the position of the lines in the mass spectrum and to verify the beam focusing conditions. When the cross sections σ_{0k} were measured, the slit S_2 was moved out and the input aperture of receiver F_4 was limited by the aperture of the diaphragm D (25 mm dia). Receiver F_4 was connected to a type ÉMU-2 electrometer with maximum sensitivity 2×10^{-15} amp/div.

The analyzer chamber was evacuated with a TsVL-100 supplementary pump. In the presence of gas in the collision chamber the pressure in the analyzer chamber was less than 1×10^{-5} mm Hg. That the collisions were single was confirmed by the presence of practically linear portion on the slow-ion beam intensity vs collision-chamber gas pressure curve. The pressure of the investigated gas did not exceed 2×10^{-4} mm Hg.

The slow ions were identified by comparing the spectrograms with and without the investigated gas in the collision chamber.

The cross sections for the formation of differently charged slow ions was determined from the formula

$$\sigma_{0k} = \frac{1}{k} \alpha_k \sigma_+, \tag{4}$$

where α_k — relative intensity of the lines of slow ions with charge k and σ_+ — total cross section for the production of slow ions (per unit charge).

*In measurements with fast ions the accelerating voltage V was increased by the same amount.

The cross sections σ_+ for the atom-atom and single-charge ion-atom pairs investigated in the present work were measured by us earlier.[3] For doubly-charged and triply-charged ions, the cross sections σ_+ were measured simultaneously with σ_{0k}. It is obvious that σ_+ is the sum

$$\sigma_+ = \sigma_{01} + 2\sigma_{02} + 3\sigma_{03} + \ldots . \quad (5)$$

In most cases the overall error in the measurement of the cross section σ_{0k}, estimated from the reproducibility of the results, did not exceed 15%. In measurements made with krypton, the lines of the slow Kr^{3+} ions were masked by a large background due to the N_2^+ which has the same ke/M, so that an additional systematic error has crept into the measurement of the cross sections σ_{03} in krypton.

2. MEASUREMENT RESULTS AND DISCUSSION

We investigated the dependence of the cross sections for the production of slow ions of various charges (σ_{0k}) on the kinetic energy T of the fast ionizing particles. The results of the measurements for fast Ne^0, Ne^+, Ne^{2+}, and Ne^{3+} particles are shown in Figs. 2—5 for the Ne, Ar, Kr, and Xe atoms, respectively, while the result for the fast Kr^0, Kr^+, Kr^{2+}, and Kr^{3+} particles are shown in Figs. 6 and 7 for the Kr and Xe atoms respectively.

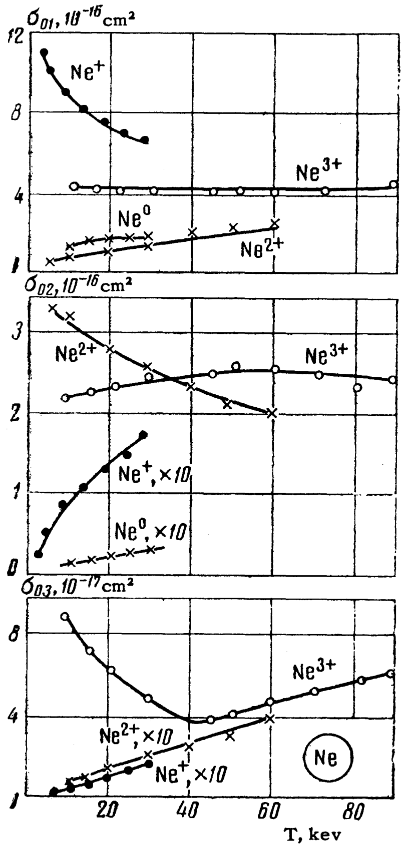

FIG. 2. Dependence of the cross sections for the production of slow singly-charged (σ_{01}), doubly-charged (σ_{02}), and triply-charged (σ_{03}) ions in neon on the energy T of the ionizing particles. The ionizing particles were Ne^0, Ne^+, Ne^{2+}, and Ne^{3+}.

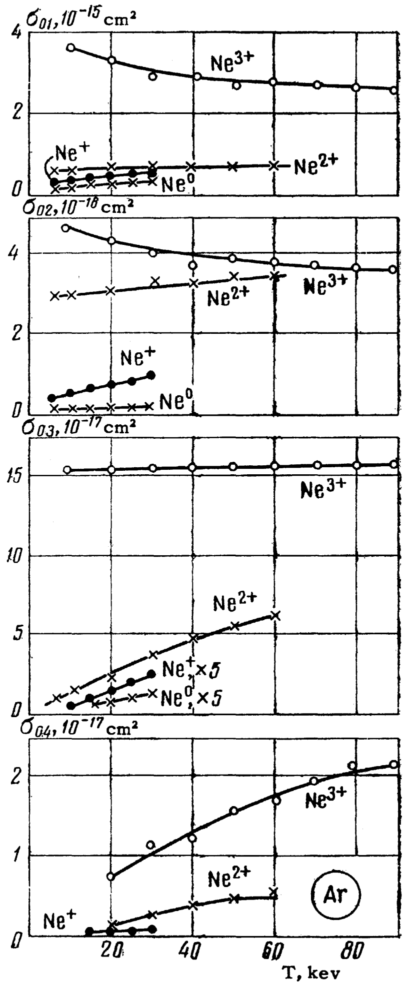

FIG. 3. Cross sections for the production of slow ions in argon.

It can be established, first, that in any given gas the cross sections σ_{0k} for the production of slow ions, increase as a rule with increasing fast-particle charge n, and that for neutral atoms the $\sigma_{0k}(T)$ curves lie lower than all other curves. The course of the $\sigma_{0k}(T)$ curves for different pairs of colliding atomic particles is not the same; in the case of fast neutral atoms the σ_{0k} increase continuously with increasing cross-section energy, while for the ion-atom pairs the σ_{0k} both increase and decrease. In some cases the variation of σ_{0k} was not monotonic.

A. Formation of Singly-Charged Slow Ions

It can be seen from the general schemes (1)—(3) that when a neutral inert-gas atom I^0 collides

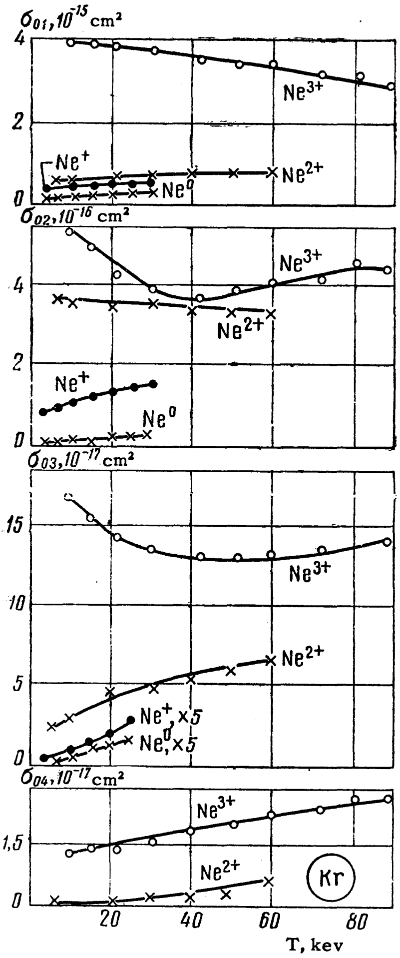

FIG. 4. Cross sections for the production of slow ions in Kr.

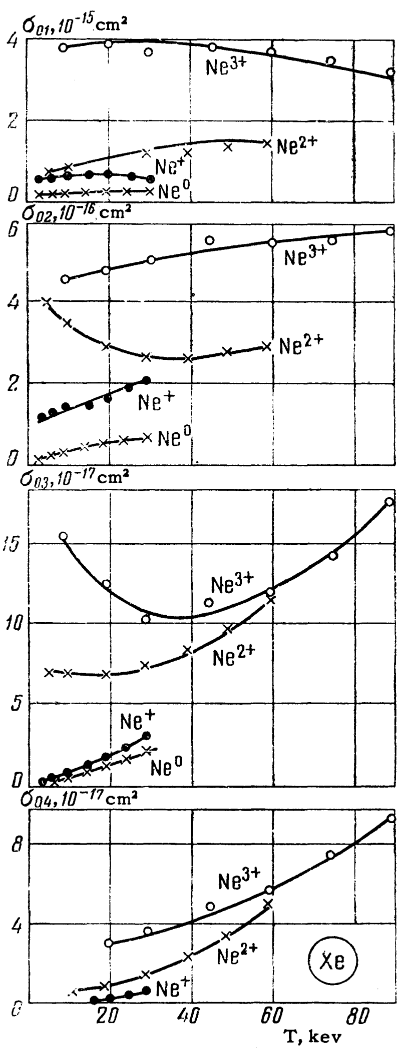

FIG. 5. Cross sections for the production of slow ions in Xe.

with gas atoms A slow singly-charged ions A^+ are produced only via pure ionization

$$I^0 + A \rightarrow I^0 + A^+ + e. \qquad (6)$$

In the energy interval 3—30 kev, simultaneous "stripping" and ionization ($I^0 + A \rightarrow I^+ + A^+ + 2e$) can be neglected since σ_{01}^{01} is small compared with σ_{01}^{00}.

It follows from general considerations that the maximum of the energy dependence of the pure ionization should lie above the Bohr velocity $e^2/\hbar$, i.e., beyond the investigated energy interval in the case of heavy fast particles. [9] In this connection, all the $\sigma_{01}(T)$ curves for fast neutral atoms increase with energy (Figs. 2—7).

In the case of ion-atom collisions, single-charged slow ions can be created both via pure ionization

$$I^{n+} + A \rightarrow I^{n+} + A^+ + e, \qquad (7)$$

and via single-electron charged exchange

$$I^{n+} + A \rightarrow I^{(n-1)+} + A^+. \qquad (8)$$

The form of the curves $\sigma_{01}(T)$ for the like pairs Ne^+–Ne and Kr^+–Kr (Figs. 2 and 6) shows clearly that the predominant role in the formation of singly-charged ions is played by resonant charge exchange, the cross section of which decreases continuously with increasing energy. For other I^+–A pairs, σ_{01} increases with energy because of the increase in the cross section for the non-resonant charge exchange and pure ionization.

As a rule, σ_{01} increases with increasing n, owing to the increase in the relative role of the charge-exchange processes. Only for the like pairs Ne^+–Ne and Kr^+–Kr is σ_{01} greater than for the pairs Ne^{2+}–Ne, Kr^{2+}–Kr and Ne^{3+}–Ne, Kr^{3+}–Kr, since the cross section of the resonant single-electron charge exchange, σ^{10}, is greater than the cross sections for the single-electron charge exchange σ^{21} and σ^{32}. [5-7]

From a comparison of the data of the present paper and the earlier data [5-7] we can also es-

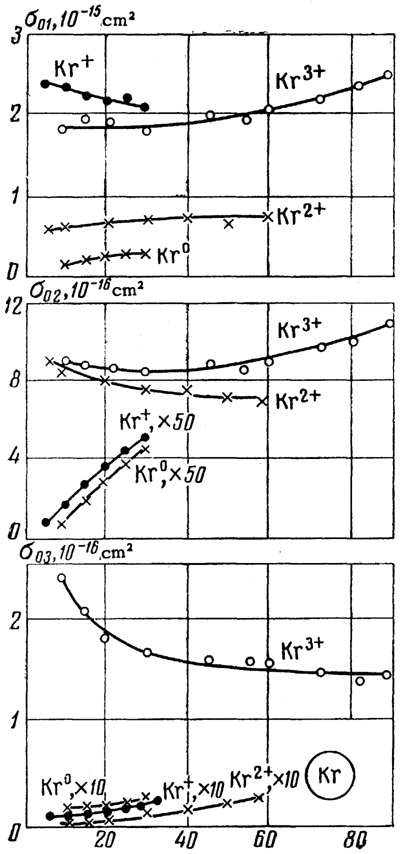

FIG. 6. The same as Fig. 4; the ionizing particles are Kr^0, Kr^+, Kr^{2+}, and Kr^{3+}.

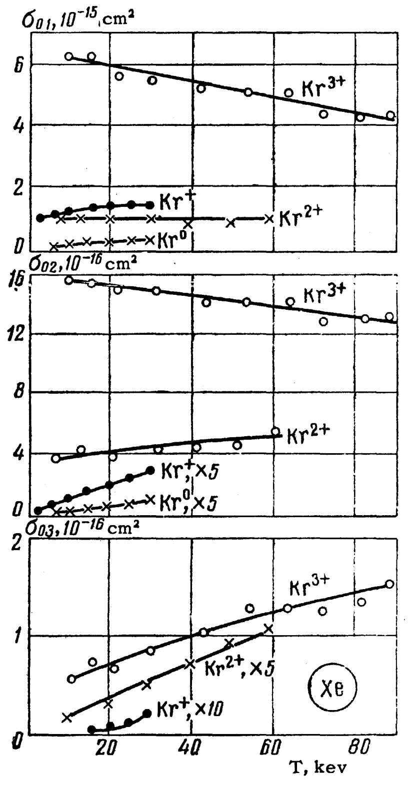

FIG. 7. The same as Fig. 5; the ionizing particles are Kr^0, Kr^+, Kr^{2+}, and Kr^{3+}.

tablish that the $\sigma_{01}(T)$ curves for fast doubly-charged ions are similar to the $\sigma^{21}(T)$ curves for single-electron charge-exchange, while for the Ne^{3+}–Kr pair and the same velocity σ_{01} is very close to the charge-exchange cross section σ^{32}. For the like pairs Ne^{3+}–Ne and Kr^{3+}–Kr, the cross sections σ_{01} and σ^{32} are also close in magnitude and increase simultaneously with increasing energy. We have no data to compare the pairs Ne^{3+}–Ar, Ne^{3+}–Xe, and Kr^{3+}–Xe, but the large values of σ_{01} undoubtedly indicate that the charge-exchange cross sections of the fast triply-charged ions, which have not yet been measured, are large.

B. Doubly-Charged Slow Ions

For the I^0–A pairs, the production of A^{2+} ions, as of all slow ions with larger charge, must be ascribed to pure ionization. For the I^{n+}–A pairs, a considerable contribution to the cross sections σ_{02} is made by ionization with capture, and when $n \geq 2$ the two-electron charge exchange processes also contribute.

An examination of Figs. 2–7 shows that for all the investigated I^0–A pairs σ_{02} continuously increases with increasing energy T and with decreasing energy of double ionization of the gas atom. In the initial region of the energies, the cross sections for the formation of slow ions, $\sigma_{02}(I^0)$ and $\sigma_{01}(I^0)$ differ by one order of magnitude and more; with increasing energy, this difference decreases because of the steeper variation of the $\sigma_{02}(T)$ curves.

For I^+–A pairs $\sigma_{02}(I^+)$ also increases continuously with the energy, but much more slowly than $\sigma_{02}(I^0)$. The great difference between $\sigma_{02}(I^+)$ and $\sigma_{02}(I^0)$ shows that ionization with capture plays the predominant role in the production of A^{2+} ions, since this process involves less energy loss than pure ionization, by an amount equal to the energy required to neutralize the fast single-charge ion.

With increasing charge of the fast ions, the cross sections σ_{02} increase and the character of their energy dependence changes. In the case of the like pairs Ne^{2+}–Ne and Kr^{2+}–Kr, doubly-charged slow ions are formed predominantly by resonant two-electron charge exchange, and the cross sections σ_{02} decrease monotonically with increasing energy. It is easily seen that $\sigma_{02}(I^{2+})$ exceeds $\sigma_{01}(I^{2+})$ in the initial energy range (6–40 kev), since the resonant charge-exchange cross section σ^{20} exceeds the charge-exchange cross section σ^{21}.[5]

For the Ne^{2+}–Kr and Ne^{2+}–Xe pairs, a greater contribution may be made to σ_{02} by ionization with capture

$$I^{2+} + A \rightarrow I^{+} + A^{2+} + e, \tag{9}$$

rather than by charge exchange ($I^{2+} + A \rightarrow I + A^{2+}$). This is confirmed by the fact that the cross sections σ_{02} for these pairs are greater than the charge-exchange cross sections σ^{20} measured in [6], and the $\sigma_{02}(T)$ and $\sigma^{20}(T)$ curves are different. This relation obtains between the cross sections because processes (9) are exothermic for these pairs.

For the remaining two pairs, Ne^{2+}–Ar and Kr^{2+}–Xe, processes (9) are endothermic and their contributions to σ_{02} is apparently smaller. For these pairs both the σ_{02} and the σ^{20} increase continuously with the energy T.

In the case of triply-charged fast ions the cross sections σ_{02} for the like pairs Ne^{3+}–Ne and Kr^{3+}–Kr are only 2 or 2.5 times smaller than the corresponding cross sections σ_{01}, while for the unlike pairs they differ by a factor 6 or 7. We obtained the same relation for the cross sections of two-electron (σ^{31}) and single-electron (σ^{32}) charge-exchange.[7] However, for the pairs Ne^{3+}–Ne, Ne^{3+}–Kr, and Kr^{3+}–Kr, which are comparable with the data of [7], the σ_{02} greatly exceed the capture cross sections σ^{31}, since the σ_{02} include ionization with capture

$$I^{3+} + A \rightarrow I^{2+} + A^{2+} + e, \tag{10}$$

a process that is exothermic for all the investigated pairs except Kr^{3+}–Kr.

C. Triply-Charged Slow Ions

The cross section σ_{03} for pure triple ionization was measured only for the pairs Ne^{0}–Ar, Ne^{0}–Kr, Ne^{0}–Xe, and Kr^{0}–Kr. For the remaining two pairs, Ne^{0}–Ne and Kr^{0}–Xe, the currents corresponding to triply-charged slow ions where below the sensitivity limit of the electrometer.

For I^{n+}–A pairs with $n \geq 1$, A^{3+} ions are also produced by ionization with capture, while when $n \geq 3$ three-electron charge exchange is added to these processes. An examination of Figs. 2–7 readily shows that the $\sigma_{03}(I^{+})$ differ from the $\sigma_{03}(I^{0})$ in the mean by only 1.5–2 times, since pure ionization and ionization with capture

$$I^{+} + A \rightarrow I^{0} + A^{3+} + 2e \tag{11}$$

are strongly endothermic processes and the relative decrease in the energy of reaction (11) is small compared with the energy of triple ionization of the gas atom. With increasing energy T, $\sigma_{03}(I^{0})$ and $\sigma_{0}(I^{+})$ increase continuously.

For I^{2+}–A pairs, the ionization with capture processes

$$I^{2+} + A \rightarrow I^{+} + A^{3+} + 2e, \tag{12}$$

$$I^{2+} + A \rightarrow I^{0} + A^{3+} + e, \tag{13}$$

are also endothermic and the cross sections σ_{03} increase with energy. However, the reaction energy of the process (12), and particularly of (13), is considerably less than the energy of the pure ionization process, and therefore the $\sigma_{03}(I^{2+})$ greatly exceed $\sigma_{03}(I^{0})$ for almost all the I^{2+}–A pairs.

In the collision of triply-charged ions with gas atoms, I^{3+}–A, the predominant role in the formation of A^{3+} ions is played in the case of the like pairs Ne^{3+}–Ne and Kr^{3+}–Kr by the resonant charge-exchange processes. The cross sections σ_{03}, like the three-electron charge exchange cross section σ_{30}, decrease with increasing energy.[7] For the Ne^{3+}–Ne pair at energies ~45–90 kev, the charge-exchange cross section σ^{30} is practically constant and the increase in the cross sections σ_{03} with increasing energy is due to the endothermal processes of ionization with capture

$$I^{3+} + A \rightarrow I^{2+} + A^{3+} + 2e, \tag{14}$$

$$I^{3+} + A \rightarrow I^{+} + A^{3+} + e, \tag{15}$$

and also to pure ionization.

A comparison of σ_{03} with σ^{30} for the Ne^{3+}–Kr pair shows that for the same energy σ_{03} is 4–5 times greater than σ_{30},[7] since the processes (14) are exothermic for this pair, as well as for the Ne^{3+}–Ar and Ne^{3+}–Xe pairs, and it is probable that these processes make a greater contribution to σ_{03} than the three-electron charge-exchange processes.

D. Quadruply-Charged Slow Ions

The cross sections σ_{04} were measured only for the collisions of Ne ions with Ar, Kr and Xe atoms. As can be seen from the data of Figs. 4–6, the cross sections σ_{04} increase sharply with increasing energy and with increasing charge of the fast ions. The cross section σ_{04} is on the order of 10^{-19} cm^2 for the Ne^{+}–Ar pair and on the order of 10^{-18} cm^2 for the Ne^{+}–Xe pairs. With increasing fast-ion charge, the number of ionization processes with capture increases and the energies of these reactions are greatly reduced, thereby increasing σ_{04}. For the Ne^{3+}–Xe pair ionization with capture ($Ne^{3+} + Xe \rightarrow Ne^{0} + Xe^{4+} + e$) is an

Charge-exchange cross section σ^{10} (in units of 10^{-16} cm^2) calculated from (18) and (19) (in parentheses) and determined by measuring the difference $(\sigma_+ - \sigma_-)$ [3]

Energy	Ne+ — Ne		Ne+ — Ar		Ne+ — Kr		Ne+ — Xe		Kr+ — Kr		Kr+ — Xe	
T, kev	σ^{10}	$\sigma_+ - \sigma_-$	σ^{10}	$\sigma_+ - \sigma_-$	σ^{10}	$\sigma_+ - \sigma_-$	σ^{10}	$\sigma_+ - \sigma_-$	σ^{10}	$\sigma_+ - \sigma_-$	σ^{10}	$\sigma_+ - \sigma_-$
6	—	—	2,4	2.5	3,6	3.6	5.8	5.6	—	—	10.8	8,4
15	6.7 (5.4)	5.6	2,75	3.0	4.0	4.2	5,4	5.3	20,1 (17.8)	18	12.5	9.3
25	5.7 (4,1)	4.2	3.3	3.45	4,25	4,3	5,0	4.8	19,1 (16,4)	16,5	12,8	9,3

exothermal process and this is apparently why σ_{04} reaches a value close to 1×10^{-16} cm^2 for this pair.

E. Estimate of the Cross Section for Ionization with Capture

By examining the experimental data for the simplest cases, when the fast particles are singly-charged ions and σ_{0k} is the sum of only two cross sections, we can attempt to estimate quantitatively the contribution of the individual cross sections to the overall cross sections σ_{0k}, starting from the assumption that the pure-ionization cross sections are the same for fast neutral atoms and for singly-charged ions

$$\sigma^{11}_{0k}(I^+) = \sigma^{00}_{0k}(I^0). \tag{16}$$

In this case the charge-exchange cross section σ^{10}_{01}, which enters in $\sigma_{01}(I^+)$, and the cross sections for ionization with capture, σ^{10}_{02} and σ^{10}_{03}, which enter in σ_{02} and σ_{03}, are differences of the cross sections

$$\sigma^{10}_{0k} = \sigma_{0k}(I^+) - \sigma_{0k}(I^0). \tag{17}$$

The assumption (16) can be verified by comparing the summary charge-exchange cross sections, obtained by adding the cross sections calculated from (17)

$$\sigma^{10} = \sigma^{10}_{01} + \sigma^{10}_{02} + \sigma^{10}_{03} \tag{18}$$

and measured in [3] by determining the difference of the cross sections $(\sigma_+ - \sigma_-)$.

The values of σ^{10} and $(\sigma_+ - \sigma_-)$ are compared in the table for 6, 15, and 25 kev (the cross sections are given in units of 10^{-16} cm^2).

As can be seen from the table, assumption (16) is valid only for the pairs Ne$^+$–Ar, Ne$^+$–Kr and Ne$^+$–Xe. For the remaining cases, the cross sections for pure ionization of gas atoms by singly-charged ions and by fast ions may differ appreciably. In particular, in the ionization of gas atoms by fast atoms of the same gas, the same amount of energy is consumed in ionization and in "stripping," and the cross sections of these processes should be equal. [3]

In ionization by ions with relatively low energy (3–30 kev), we can neglect the "stripping" of the ions and instead of (16) we can assume for the like ion-atom and atom-atom pairs

$$\sigma^{11}_{0k}(I^+) \approx 2\sigma^{00}_{0k}(I^0), \tag{19}$$

thereby obtaining a good agreement between σ^{10} and $(\sigma_+ - \sigma_-)$ [the values of σ^{10} determined from (19) are shown in the table in parentheses].

By way of an example, Fig. 8 shows the estimated charge-exchange cross sections σ^{10}_{01} and

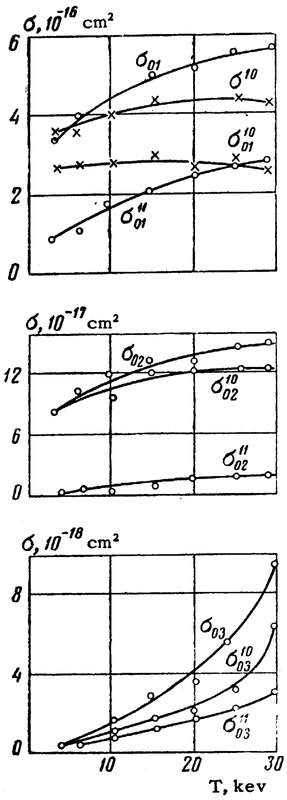

FIG. 8. Dependence of the cross sections of different processes in Kr on the energy of the fast Ne$^+$ particle. σ_{01}, σ_{02}, σ_{03} – cross sections for the production of singly-, doubly-, and triply-charged ions; σ^{11}_{01}, σ^{12}_{02}, σ^{11}_{03} – cross sections for pure ionization: σ^{10}_{01}, σ^{10}_{02}, σ^{10}_{03} – cross sections for ionization with capture; σ^{10} – charge-exchange cross section.

the cross sections for ionization with capture σ_{02}^{10} and σ_{03}^{10} for the pair Ne^+–Kr. The same figure shows also the summary cross sections σ_{01}, σ_{02}, and σ_{03} as well as the pure-ionization cross sections σ_{01}^{11}, σ_{02}^{11} and σ_{03}^{11}, determined under the assumption (16), and also the summary charge-exchange cross sections σ^{10}. It is seen from these data that in the initial energy range the relative contribution of charge exchange to σ_{01} predominates over the contribution from pure ionization. But since the pure-ionization cross section increases more rapidly with energy than does the charge-exchange cross section, the contributions from both processes are approximately the same at the limiting energy value. Pure ionization plays a small role in the formation of doubly-charged slow ions, while pure ionization and ionization with capture are approximately of equal weight in the production of triply-charged slow ions. All these conclusions are in good agreement with remarks in the preceding sections, based on energy considerations, concerning the relative probability of various processes.

In conclusion, the author is deeply grateful to Professor V. M. Dukel'skii for a discussion of the results of the work.

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194