IONIZATION OF INERT GASES BY MULTIPLY CHARGED IONS

N. V. FEDORENKO, I. P. FLAKS, and L. G. FILIPPENKO

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

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The total ionization cross sections (σ_-) for single collisions between Ne^+, Ne^{2+}, Ne^{3+}, Kr^+, Kr^{2+}, Kr^{3+}, Xe^+, Xe^{2+}, Xe^{3+}, Xe^{4+} ions and inert gas atoms have been measured. In the investigated voltage range from 3 to 30 kv, it was found that the ionization cross section is practically independent of the charge of the primary ion for equal ion energy. For all ion-atom pairs, a continuous growth of the cross section with increase in kinetic energy of the primary ions was observed. In most cases the absolute values of the measured cross sections were close to those computed by Firsov's formula.[1]

INTRODUCTION

INFORMATION on the ionization of atoms by multiply charged ions is limited to the single work of Sherwin,[2] in which the ionization cross sections of molecules of hydrogen and atoms of helium by singly, doubly and triply charged ions of certain metals were measured for two values of the accelerating voltage (6 and 24 kv). Sherwin came to the conclusion that the ionization cross section increased in proportion to the square of the charge of the primary ion. As is well known, such a charge dependence is given by the theory of shock ionization for regions of high energy, when the condition $v \gg v_e$ is satisfied, where v is the velocity of relative motion and v_e is the velocity of the outer electrons in the atom.

Studies carried out in recent years[3] on the ionization of the atoms of a gas by singly charged ions in the velocity region $v \lesssim v_e$ have shown that the collisions accompanied by ionization do not reduce to the interaction of the ion with the individual electrons of the atom. It was established that the processes of ionization are realized in "penetrating" atomic collisions and the interaction of the electronic shells of the colliding atomic particles possesses great significance.

Recently a theoretical paper by Firsov[1] appeared, in which the ionization cross section for atomic collisions was calculated, on the basis of a quasi-classical representation, as a function of the number of electrons in the shells of the two particles.

Thus it is not difficult to explain the data of Sherwin on the ionization by multiply charged ions on the basis of recently developed ideas on the mechanism of ionization of atoms by singly charged ions in the region of velocities $v \le v_e$. The present research was undertaken with a view toward carrying out a further investigation of ionization by ions of different charge with the application of a more reliable indication of monokinetic beams of multiply charged ions and with a method of measurement of the ion cross section developed in our laboratory.

In the present work, the study was limited to the ions Ne^+, Ne^{2+}, Ne^{3+}, Kr^+, Kr^{2+}, Kr^{3+}, Xe^+, Xe^{2+}, Xe^{3+}, Xe^{4+} and the atoms Ne, Kr, and Xe.

INVESTIGATION PROCEDURE

The study of ionization by ions of different charge was carried out on an experimental apparatus whose general description was given in our previous researches.[4-6] The primary ions were produced in an ion source which is a modification of the source of Ardenne.[7] A narrow ion beam of definite charge, composition and energy was separated by a magnetic mass spectrometer and passed through a rectangular slit S_1 (1×4 mm) in the collision chamber which was filled with the gas under investigation (Fig. 1). A sectionalized measuring capacitor, described in detail in reference 8, was placed in the collision chamber. The free electrons arising in the gas and the secondary ions were deflected by the electric field onto the plates 1 — 12 of the capacitor and the plates 13, 14 of the Faraday cage F_1. The plates of the measuring capacitor were screened by metallic grids with a transparency of about 90 per cent. A potential of ± 45 v relative to the ground,

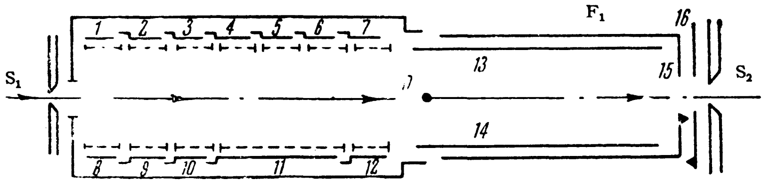

FIG. 1. Measuring capacitor.

was applied to the grids and to plates 13 and 14, while an additional +25 v relative to the grid was applied to plates 1 — 12. In this way, the secondary electrons emerging from the plates were held back by the electric field. Plates 5 and 6 in the middle part of the capacitor were chosen as the measuring plates. It was established by control experiments that the current of electrons and secondary ions were known to reach saturation, and only the charged particles appearing in the gas in a section equal to the length of the measuring plate were drawn off onto plates 5 and 6. A typical volt-ampere characteristic of the currents of the secondary ions and the free electrons on plates 5 and 6 is shown in Fig. 2.

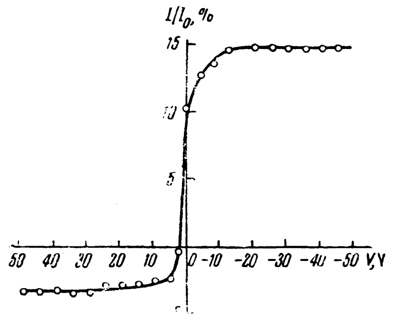

FIG. 2. Volt-ampere characteristic. The pair Kr^{2+}–Kr, energy 30 kev, gas pressure $p = 1.6 \times 10^{-4}$ atm, I – current in the measuring plates, I_0 – current of the primary beam.

The current of the primary beam was measured on the entire system of electrodes forming the measuring capacitor and the Faraday cage, and also on the suppressor plate 16, which covered the bottom of the cage at the time of measurement of the primary beam.

For correction of the beam inside the measuring condenser in the measurement of currents on plates 5 and 6, the collision chamber could be rotated about the axis O (which passes through the center of the chamber) by a small angle relative to the initial direction of the beam.[9] In this case the ion beam passed through the opening 15 into F_1 (8 × 12 mm) and the slit S_2 (6 × 10 mm), and was picked up on the monitor of the primary beam (not shown in Fig. 1).

To verify the recordings of the primary multiply-charged ions, measurements were made of the relative intensity of the lines of the different isotopes of the ions under study, which were then compared with the known tabular values. In addition, the presence of a magnetic analyzer after the collision chamber permitted us to observe the multiply-charged ions by means of the additional lines which appear in the ion beam after transmission through the gas [as the result of partial charge exchange (see references 4 — 6)].

The currents on plates 5 and 6 and the primary current were measured with an EMU-2 amplifier.

The experiments were carried out under conditions of single stage character of the collisions at gas pressures $\sim 1 \times 10^{-4}$ atm. The gas pressure drop amounted to $\sim 1:50$ at the slit S_1, and to $\sim 1:15$ at slit S_2. The residual pressure in the apparatus after shutting off admission of the gas into the collision chamber did not exceed 1×10^{-6} atm. The pressure was measured by an ionization manometer controlled by a McLeod gauge.

DETERMINATION OF THE CROSS SECTIONS

In the present research we measured the negative I_- and the positive I_+ saturation currents to the plates of the measuring condenser. Measurement of these currents permitted us to determine the total cross section for the formation of free electrons σ_- and the total cross section of formation of secondary ions σ_+ referred to a single charge:

$$\sigma_- = zI_- / NlI_0, \qquad \sigma_+ = zI_+ / NlI_0, \tag{1}$$

where I_0 is the current of the primary beam, z is the multiplicity of the charge of the primary ions, N is the number of atoms per cubic centimeter of gas, l is the length of the measuring electrodes.

The cross section σ_-, as also in reference 8, was of interest to us as the "total cross section" of ionization. Data on the cross section σ_+ were frequently used by us for the comparison with data on cross sections of capture of electrons by ions of the primary beam, obtained in references 4 — 6.

In the passage of a beam of singly charged ions through the gas, the cross sections σ_- and σ_+ are composed of the cross sections of the separate processes of three groups:

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

$$I^{i+} + A \rightarrow I^{f+} + A^{k+} + (f + k - i)e, \tag{II}$$

$$I^{i+} + A \rightarrow I^{l+} + A + (l - i)e. \tag{III}$$

The processes of group I have been named "pure" ionization of the gas atoms, those of the group II are known as ionization with capture, while those of group III as stripping of the primary ion. The processes of groups I and II can evidently exist simultaneously and lead to the release of electrons from the shells of both colliding atomic particles. Processes of group II are connected with the liberation of electrons from the shell of an atom and with partial neutralization of the primary ion. As the results of the existence of processes of groups I and II in the gas, secondary ions can be formed with different multiplicity of charge, and in the stripping (processes of group III), ions of much higher charge than that of the primary ions appear in the primary beam.

Complete account of all possible processes leads to the result that

$$\sigma_+ - \sigma_- = \sigma_0 - \sigma_l, \qquad (2)$$

where σ_0 is the "total cross section" of ionization with capture, while σ_l is the "total cross section" of stripping, referred to a single charge. In the energy region under investigation, $\sigma_l \ll \sigma_0$ and

$$\sigma_+ - \sigma_- \approx \sigma_0. \qquad (3)$$

In the general case of primary ions with i-fold charge,

$$\sigma_0 = \sum_{f=i-1}^{f=0} (i-f)\,\sigma_{if}, \qquad (4)$$

where σ_{if} is the cross section for formation of ions with charge from $i-1$ to 0. The separate components of the sum (4) can be determined by analysis of the charge composition of the ion beam after passage through the gas.[4-6]

The currents I_- and I_+ entering into Eq. (1) were measured as the difference of the saturation currents in the gas under study and after pumping out of the collision chamber to the limiting vacuum.

The relative crudeness of the measurement of the cross sections σ_- and σ_+ is estimated by us to be of the order of 20 per cent, starting from the reproducibility of the results. It is connected with random errors in the measurement of the currents and of the gas pressure, and with a certain instability of the operation of the ion source.

RESULTS

The dependence of the total cross section of ionization of the gases under study by the various ions is shown in Fig. 3 as a function of the kinetic energy of the primary ions T.

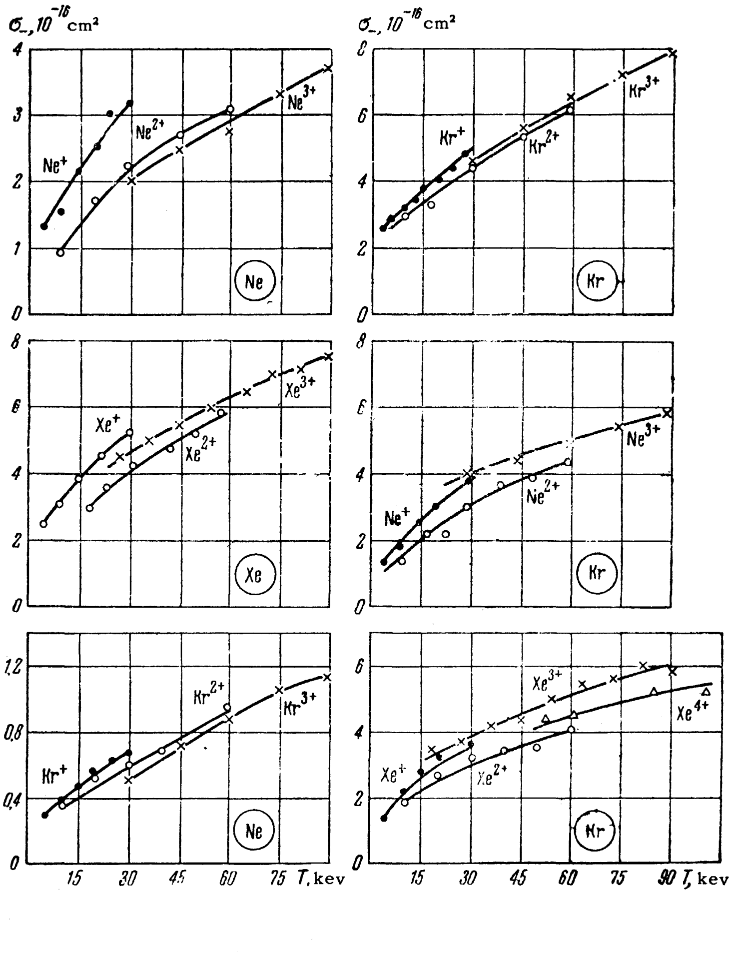

FIG. 3. Total ionization cross section of gases (shown in the circles) by ions (shown on the curves) as a function of the energy.

In the ionization of neon by neon ions, σ_- is somewhat larger for the Ne^+ ions at the same kinetic energy (or the same velocity of the ions). For the ions Ne^{2+} and Ne^{3+}, the cross sections are identical within the limits of error of the experiment.

In two other cases of ionization of a particular gas by ions of different charge, and in particular ionization of krypton by krypton ions and ionization of xenon by xenon ions, the cross sections σ_- are approximately the same for a given energy.

A similar conclusion can be reached from a consideration of the data for the ionization of gases by strange ions: krypton by neon ions, neon by krypton ions and krypton by xenon ions (see Fig. 3).

An uninterrupted increase in the ionization cross section was observed for all pairs of ions-atoms studied in the energy interval under consideration. In the middle part of the energy interval ($T \approx 50$ kev) the largest values were achieved for the ionization cross section of xenon by xenon ions and krypton by krypton ions ($\sigma_- \approx 6 \times 10^{-16}$ cm^2), and the smallest ionization cross section was that of neon by krypton ions ($\sigma_- \approx 8 \times 10^{-17}$ cm^2).

Two of the pairs studied by us, namely, Ne^+-Ne and Kr^+-Kr, were also measured by Gilbody and Hasted[10] in the energy ranges from 0.1 to 3 kev and from 5 to 40 kev. The cross section σ_- in reference 10 for the pair Ne^+-Ne is larger than ours by about 70 per cent, while that for Kr^+-Kr is 4 — 5 times larger. It should be noticed that these authors observed almost the same divergence for these pairs among the series of their own measurements completed in the region of small and high energies.

We carried out comparison of the difference in the cross sections σ_+-σ_- determined in our present work with the charge-exchange cross sections σ_{10} measured by us in reference 4 by the method of recording fast neutral atoms. This comparison was made for the pairs Ne^+-Ne and Kr^+-Kr. The results of the comparison are shown in Fig. 4. A similar comparison is shown in Fig. 5 for the pair Kr^{2+}-Ne, for which the cross section for capture of two electrons σ_{20} is very small in comparison with the cross section of capture of a single electron σ_{21} (see reference 5) and, consequently, by (4), σ_+-$\sigma_- \approx \sigma_{21}$. As is seen from Fig. 5, in the case of doubly charged ions, a close correspondence is also observed between the results of independent measurements.

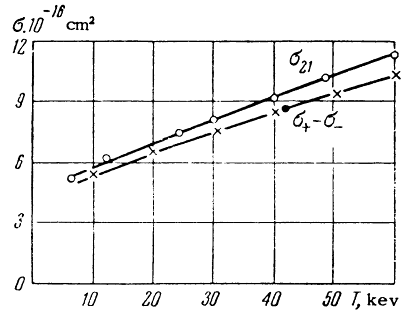

FIG. 5. Comparison of the capture cross section measured by the potential method $\sigma_+ - \sigma_-$ and by the mass spectrometer method (σ_{21}). The pair is Kr^{2+}−Ne.

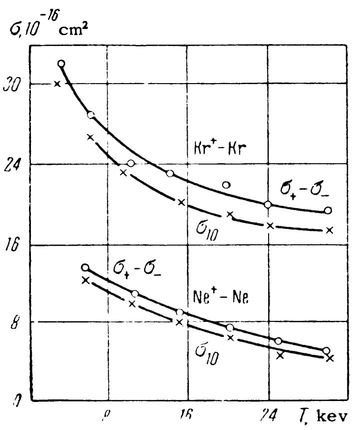

FIG. 4. Comparison of the capture cross section measured by the potential method $\sigma_+ - \sigma_-$ and by the method of registering fast atoms (σ_{10}). Pairs are Ne^+−Ne and Kr^+−Kr.

DISCUSSION

The results of our investigation of the ionization of inert gases by multiply charged ions do not substantiate the quadratic dependence of the cross section on the charge of the primary ion found by Sherwin. It should be noted in passing that the cross sections in the work of Sherwin were compared, not for identical kinetic energy of ions of different charge, but for the same accelerating voltage. Thus a weaker dependence of cross section on charge actually follows from the data of Sherwin. In several cases (Cu^+, Cu^{2+}-He; C^+, C^{2+}-He), the cross sections are practically identical for singly and doubly charged ions. Some shortcomings in the method of the research of Sherwin were noted in reference 11. The absence of a reliable indication of multiply charged ions and the measurement of cross sections for comparatively high gas pressure ($\sim 5 \times 10^{-5}$ atm) also reduced the reliability of the data of Sherwin.

In the theoretical work of Firsov[1] which appeared recently, an estimate is given of the ionization cross section in atomic collisions, starting out from a quasiclassical consideration of the problem of the motion of electrons in the region of overlap of the shells of the colliding atomic particles. The ionization cross section is represented in the form of the approximate formula

$$\sigma = \frac{32.7 \cdot 10^{-16}}{(Z_1 + Z_2)^{2/3}} \left\{ \left[\frac{(Z_1 + Z_2)^{5/3} v}{23.3 \cdot 10^6 \varepsilon_i} \right]^{1/5} - 1 \right\}^2, \qquad (5)$$

where ϵ_i is the ionization energy, while Z_1 and Z_2 are the number of electrons in the ion and in the atom, respectively.

Introducing the characteristic velocity v_0 and the cross section σ_0 for each pair of colliding particles

$$v_0 = \frac{23.3\varepsilon_i}{(Z_1 + Z_2)^{1/6}} \cdot 10^6 \text{ cm/sec},$$

$$\sigma_0 = \frac{32.7}{(Z_1 + Z_2)^{2/3}} \cdot 10^{-16} \text{ cm}^2, \qquad (6)$$

Firsov obtained a universal formula for the ionization cross section for an arbitrary pair of colliding particles:

$$\sigma / \sigma_0 = [(v / v_0)^{1/5} - 1]^2. \qquad (7)$$

The dependence of σ/σ_0 on v/v_0, given by Eq. (7), is represented in Fig. 6 by the dashed curve. The experimental data for all ion-atom pairs that we investigated are also plotted in the same drawing. As is seen, for heavy ions and atoms (curves 10 — 19), and also for ions and atoms of neon (curves 1, 2, 3), the experimental data are grouped about the theoretical curve. Departures from the theoretical curve, as also for singly-charged ions [1,3] lie within a factor of 2.

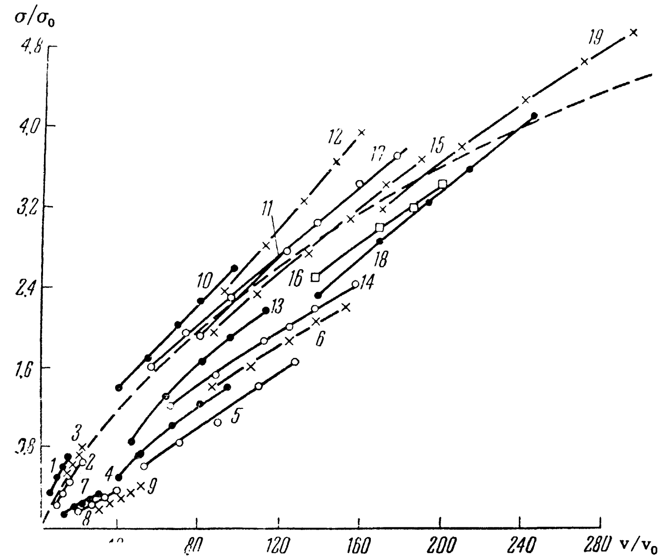

FIG. 6. Universal curve of Firsov (dashed) and experimental data on the ionization of atoms by ions of different charge

1 — Ne^+ — Ne; 2 — Ne^{2+} — Ne; 3 — Ne^{3+} — Ne; 4 — Ne^+ — Kr; 5 — Ne^{2+} — Kr; 6 — Ne^{3+} — Kr; 7 — Kr^+ — Ne; 8 — Kr^{2+} — Ne; 9 — Kr^{3+} — Ne; 10 — Kr^+ — Kr; 11 — Kr^{2+} — Kr; 12 — Kr^{3+} — Kr; 13 — Xe^+ — Kr; 14 — Xe^{2+} — Kr; 15 — Xe^{3+} — Kr; 16 — Xe^{4+} — Kr; 17 — Xe^+ — Xe; 18 — Xe^{2+} — Xe, 19 — Xe^{3+} — Xe

The essential experimental fact obtained in the present work, namely the absence of a significant effect of the charge of the primary ion on the magnitude of the ionization cross section, is in qualitative agreement with the Firsov theory. Actually, inasmuch as Z_1 and Z_2 are large numbers in Eq. (5), a decrease in Z_1 by several units in the case of multiply charged ions should not have a noticeable effect on the magnitude of the ionization cross section. Here it is interesting to observe that the ionization cross section for singly charged ions is as a rule always somewhat larger than for ions with a charge larger than unity (see Fig. 3).

For the cases of ionization of neon by krypton ions, and of krypton by neon ions, the Firsov formula gives cross sections that are too high by a factor of 2 — 4. It is possible that this is connected with the fact that individual peculiarities of the electron structure of the colliding atomic particles are not taken into account in the theory.

With the aim of further investigation of the phenomenon of ionization of gases by ions of different charge, we propose to extend the investigation to ions of elements of other groups of the periodic table, in particular to metallic ions.

In conclusion, the authors express their gratitude to Professor V. M. Dukel'skiĭ for discussion of the results of the research and to A. M. Shchenkov for practical assistance in carrying it out.

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