SYMMETRIC RESONANCE CHARGE EXCHANGE OF MULTICHARGED IONS

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An experimental method is developed for studying inelastic atomic collisions involving low-energy multicharged ions. The method is based on registration of the slow ions produced in a gas by a pulsating multicharged ion beam passing through it. The slow ions are extracted from the gas target by means of a pulsating electric field. The pulses of the primary ion current and of the field extracting the slow ions are separated by a period of time such that slow-ion formation occurs in absence of the field and their extraction by the field occurs in the absence of the primary beam. The method is used to measure the effective cross sections for symmetric resonance charge exchange of doubly and triply charged neon, argon and xenon ions at accelerating voltages of 200–3000 V. The measured effective cross sections for doubly charged ions are compared with the data of other authors and with the theoretical calculations. No information is available in the literature on cross sections for symmetric resonance charge exchange in the indicated energy range.

INTRODUCTION

EARLIER investigations[1,2] have established experimentally the existence of processes of symmetrical resonant charge exchange of doubly- and triply-charged ions:

$$X^{2+} + X \rightarrow X + X^{2+}, \quad (1)$$

$$X^{3+} + X \rightarrow X + X^{3+}. \quad (2)$$

Process (1) was investigated for the ions Ne, Ar, Kr, and Xe, while process (2) was investigated for Ne and Kr ions in the region of medium ion energies (T = nU = n · (3–30) keV, where U—accelerating voltage and n—multiplicity of ion charge). The method of determining the cross sections of the processes (1) and (2), used in[1,2], is based on the registration of neutral particles—reaction products that deviate little from the direction of the primary beam. Subsequent investigations[3-6] were limited to the process (1), and only McGowan and Kerwin[6] made an attempt to observe the existence of the process (2). The cross section $_{20}\sigma_{02}$ of the process (1) was determined in the cited investigations by using the method of Cermak and Herman[3] (ion energy 75–175 eV), the potential method based on registration of slow ions[4] (7–62 keV) and [5] (400–3600 eV), and the Aston-band method[6] (600–2400 eV). The process (1) was considered theoretically in [7-9]. In [7,9], the calculations of the cross section $_{20}\sigma_{02}$ are based on the theory of the single-electron symmetrical resonant charge exchange, previously developed by Firsov[10], and the results of the calculations agree well with the known experimental data. The cross sections $_{20}\sigma_{02}$ and $_{30}\sigma_{03}$ calculated by Murakhver[8] turned out to be much higher than those obtained experimentally.

Investigations of atomic collisions at low energies are of interest for a number of applications and for the development of theory of atomic collisions. For the development of the theory of multielectron transitions, particular interest may attach to experimental data obtained with n > 2.

The purpose of the present investigation was to develop an experimental method of investigating processes and atomic collisions in which relatively slow multiply-charged ions take part. The method described below was used to measure the cross sections of processes (1) and (2), and for comparison purposes we determined also the cross sections of single-electron charge exchange $_{10}\sigma_{01}$ at two or three values of the energy T. The investigations were made with Ne, Ar, and Se ions in the energy interval T = n · (200–3000) eV. The lower limit of the ion energy was set by the low intensity of the beam of multiply-charged ions extracted from the ion source at an accelerating voltage lower than 200 V.

INVESTIGATION METHOD

The use of the existing methods for determining the charge-exchange cross sections in the region of low accelerating voltages is hindered by a number of factors. Thus, in methods based on the registration of fast particles, difficulties arise in guiding a low-intensity beam of multiply-charged ions through a long drift region. The Aston-band method makes it possible only to estimate the cross sections, in view of the difficulties connected with the determination of the effective thickness of the target. In the extensively used potential method, the presence of a constant field transverse to the primary ion beam introduces a number of uncertainties in the experiment, owing to the deflection of the primary beam in this field.

The proposed measurement method is based on the registration of slow ions produced in the gas when a pulsating beam of multiply charged ions pass through the gas under conditions of single collisions. The slow ions are extracted from the gas target with a pulsating electric field. The pulses of the primary ion beam and of the field extracting the slow ions are shifted in time in such a way that the slow ions are produced in a field-free volume, whereas the extraction of the ions by the fields takes place in the absence of the primary beam. Such a pulse method makes it possible to investigate the ionization and charge-exchange processes in atomic collisions at low energies.

Figure 1 shows a diagram of the collision chambers and of the electrodes controlling the ion beams. The ions are obtained by ionizing the gas by electron impact in a Pierce-type ion source and are accelerated, analyzed with respect to e/m in a magnetic mass spectrometer, and are focused on the plane of the entrance slit S_0 of the collision chamber. The beam J of the primary ions then passes through slit S_1, the collision region O, the slit S_2, and strikes the ion receiver C. The intensity of the primary beam in the collision region pulsates at a frequency 5×10^4 Hz from zero to its maximum value; this is effected by applying to the plate A of a parallel-plate capacitor a blocking positive voltage pulse with amplitude v_{bl} = 800 V. The parameters of the blocking pulse are as follows: duration of the leading front—less than 0.05 μsec, duration at 0.5 amplitude level—0.4 μsec, slant of top—not more than 10% of the amplitude. An expelling positive voltage pulse with frequency 5×10^4 Hz and amplitude Z_{ex} = 100 V is applied to the capacitor plate B with a delay of 0.1 μsec relative to the blocking pulse. The expelling pulse has the following parameter: leading front—less than 0.05 μsec, pulse duration at 0.5 amplitude level—0.3 μsec, slant of top—10% of amplitude.

The source of the synchronizing pulses is a GI-3M generator with pulse amplifier at the output. In the absence of voltage pulses on electrodes A and B, the primary beam passes through the gap-filled region O and the slow ions are produced. Application of the blocking pulse prevents the primary beam from entering the collision region, and the expelling voltage pulse accelerates the ions in the direction of the grounded electrode with slit S_3. The slow ions passing through slits S_3–S_8 are accelerated by the constant voltage (2 kV), are analyzed with respect to e/m in a magnetic mass analyzer, and enter the detector. The intensity of the primary ion beam is determined from the average ion current in the collector C, measured with the aid of a U-1-2 electrometric amplifier. To suppress secondary electron emission from collector C, a negative voltage of 100 V is applied to the electrode D. The degree of focusing of the primary beam and its scattering in the gas target are controlled by varying the width of the slit S_2. The intensity of the slow-ion beam is measured by an ion detector developed in our laboratory,[11] consisting of an ion–electron converter, a scintillation counter, and a counting channel. The counting channel contains a USh-10 broadband amplifier, an amplitude discriminator, an ISS-3 counting-rate meter, and an ÉPP-09 automatic-recording potentiometer.

The system of slits S_0, S_9 and S_4–S_8 makes it possible to maintain the required vacuum in the setup ($\sim 6 \times 10^{-6}$ mm Hg) when the collision chamber is filled with gas to a pressure $\sim 3 \times 10^{-4}$ mm Hg. The gas pressure is measured by an LM-2 ionization manometer. The slit dimensions are as follows: S_0—12 × 1 mm, S_1—8 × 1 mm, S_2—15 × 1 mm, S_3—11 × 1.4 mm, S_4–S_8—15 × 4 mm, S_9—14 × 1 mm (the widths of slits S_1 and S_2 can be varied).

The distance between the electrodes S_1 and S_2 is $\sim$30 mm, and 6 mm between S_3 and B, so that edge effects due to the electric fields near the slit S_3 when the expelling pulsed voltage is applied can be neglected. At the maximum amplitude of the expelling pulse, the field produced between the electrodes S_3 and B is equal in magnitude and in direction to the constant field between the electrodes S_3 and S_4, so that the electric field near the slit S_3 should be homogeneous. Then, in the presence of an expelling pulse, the slow ions are gathered on their way to analysis and registration from a volume equal to the product of the area of the slits S_3 by the width of the primary ion beam $(S_1 + S_2)/2$. It should be noted that if the number of ions leaving the effective volume V in one expulsion act is $N' > 1$, then these ions will be registered as a single pulse. Therefore to ensure correct registration of N' it is necessary that not more than one slow ion be produced in the volume V during the length of the passage of the primary-beam pulse $t' = 2 \times 10^{-5}$ sec, i.e.,

$$N' = \frac{2.21 \cdot 10^{35}}{n} Jp\sigma Vt' \leqslant 1, \tag{3}$$

where J—density of the primary-beam current in A/cm^2, n—ion charge, p—gas pressure in mm Hg, σ—cross section of the process in cm^2. Condition (3) can be easily realized by suitable choice of the product Ip for each concrete case (depending on the value of σ). Here I is the intensity of the primary beam in amperes. The present measurements were made in the region $Ip \leq 10^{-14}$ A-mm Hg. As a control of the correctness of the measurements, we used the linear dependence of the signal on the intensity of the primary-ion beam.

The charge-exchange cross sections were determined from the formula

$$\sigma = \frac{(N - N_b) n \cdot C}{2.21 \cdot 10^{35} Ip l \alpha}, \tag{4}$$

where l—effective thickness of the gas target in cm, N—intensity of slow-ion beams in counts/sec, produced as a result of charge exchange, and N_b—background in counts/sec. α is the fraction of the slow ions produced during the time of passage of one pulse (2×10^{-5} sec) and remaining in the collision region at the instant of application of the expelling pulse. The constant of the instrument is $C = 1/\alpha_1\alpha_2$, where α_1 is the transmission of the mass analyzer and of the accelerating gap S_4–S_8, and α_2 is the efficiency of registration of the ions by the detector, is close to unity for all ions.[11]

When determining the cross sections ${}_{n0}\sigma_{0n}$, particular attention was paid to the question of identical efficiency of the system with respect to the gathering of the slow ions with different charges, or the independence of the constant C/α of n. To this end, we determined in special experiments the quantity $\alpha = (N - N_b)/(N - N_b)'$, where $N - N_b$ is the intensity of the slow-ion beam measured in the pulsed mode of the instrument, and $(N - N_b)'$ is the intensity of the ion beam without pulsation of the primary beam, obtained with a constant expelling voltage on the capacitor B (Fig. 1). The magnitude of this voltage equals the amplitude of the expelling pulse. The quantity α is obtained at an accelerating voltage $\sim$3 keV, in order to avoid a no-

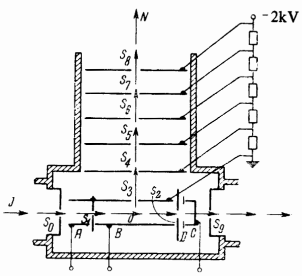

FIG. 1. Diagram of collision chamber and of the electrodes controlling the ion beams.

ticeable deflection of the beam in the field of the capacitor B. Thus, for example, we obtain $\alpha(Ar^{+}) = \alpha(Ar^{2+}) = 0.55$ and $\alpha(Xe^{2+}) = \alpha(Xe^{3+}) = 0.63$. The independence of α of the multiplicity of the charge of ions of the same atom is due to the fact that at low space-charge densities of the slow ions this charge decays only as a result of thermal motion. Indeed, calculations of α with allowance for the thermal motion yield $\alpha(Ar) = 0.6$ and $\alpha(Xe) = 0.88$. Calculations with allowance for the Coulomb repulsion yield $\alpha = 1$.

In the charge exchange processes (1) and (2), slow ions with different charges can acquire different momenta. In order for the difference in the distribution of the initial velocities to have no influence on the transmission of the mass analyzer and of the accelerating gap S_4–S_8, it is necessary, as shown in [12], to apply expelling and accelerating potentials that are high enough so that the geometrical width of the ion beam at the analyzed output is smaller than the width of the output slit. A study of the dependence of the effective cross sections for the charge exchange of ions with different charges on the accelerating voltage V_{acc} and the amplitude of the expelling pulse V_{ex} indicates that the cross section ratios ${}_{20}\sigma_{02}/{}_{10}\sigma_{01}$ and ${}_{30}\sigma_{03}/{}_{10}\sigma_{01}$ are constant in the potential regions $V_{acc} > 1000$ V and $V_{ex} > 50$ V. Thus, in this region of potentials V_{acc} and V_{ex}, the value of α does not depend on the charge of the slow ions. When these relations were obtained, the variation of V_{acc} was accompanied by a simultaneous variation of V_{ex}, in order to maintain homogeneity of the field in the collision region (Fig. 1). For subsequent measurements, the optimal potentials were chosen to be $V_{acc} = 2000$ V and $V_{ex} = 1000$ V.

The constant C/α was determined by calibrating the instrument with the aid of thoroughly investigated processes of symmetrical resonance charge exchange of the ions Ne^{+}, Ar^{+}, and Xe^{+}. The calibration was performed at those ion energies for which the agreement between the data of various authors was best, namely: 900 eV—for Ne^{+}–Ne,[13–15] 900 eV—for Ar^{+}–Ar,[15–18] and 800 eV for Xe^{+}–Xe.[14,18]

When determining the charge exchange cross sections by registering the slow ions, it is necessary to take into account also the possibility of production of slow ions as a result of ionization processes. According to estimates based on a number of experimental papers,[1,13,20] the relative contribution of the slow-ion current due to the ionization, to the total slow-ion current at T = 3 keV amounts to ~15% for Ne^{+} and Ar^{+} and ~5% in the case of Xe^{+}. When the energy T decreases, the error in the determination of the cross section ${}_{10}\sigma_{01}$, connected with the ionization processes, is much lower than the accuracy of the present measurements. The contribution of slow doubly- and triply-charged ions produced ionization to the resonance charge exchange cross sections ${}_{20}\sigma_{02}$ and ${}_{30}\sigma_{03}$ can certainly be neglected.

The use of an ion source of the Pierce type to obtain multiply charged ions makes it possible to ascertain the influence exerted on the effective charge exchange cross section by the energy state of the ions of the primary beam. To this end, we investigated the dependence of the effective cross sections of symmetrical resonant charge exchange on the energy E_e of the bombarding electrons. The width of the electrons changed from the ionization threshold of the atoms to 600 eV. As shown by the measurements, the presence of excited ions in the primary beam does not cause a noticeable change of the charge exchange cross sections for all the investigated pairs, since it is probable that the effective cross sections of symmetrical resonant charge exchange exceed the cross sections of all other possible competing processes.

By measuring the energy of the bombarding electrons it was also possible to exercise additional control over the correctness of the identification of the multiply charged ions.

MEASUREMENT RESULTS AND DISCUSSION

The accuracy with which the effective cross sections were measured, with allowance for the statistical errors and the reproducibility of the measurements, is estimated at ±20%. The absolute accuracy of the obtained cross sections depends on the accuracy of the data with the aid of which the calibration was performed.

Figures 2–4 show the measured effective cross sections ${}_{10}\sigma_{01}$, ${}_{20}\sigma_{02}$, and ${}_{30}\sigma_{03}$ of the processes $Ne^{n+} + Ne \rightarrow Ne + Ne^{n+}$, $Ar^{n+} + Ar \rightarrow Ar + Ar^{n+}$, and $Xe^{n+} + X \rightarrow Xe + Xe^{n+}$ (n = 1, 2, 3) functions of the relative velocity of the particles v and the ion energy T (solid lines), and also data obtained by others, for comparison (dashed lines). As seen from Figs. 3 and 4, the cross sections of two-electron charge exchange and their dependences on the velocity v are in good agreement with the experimental data,[1,4,5] and also with the calculated curve from [9]. Taking into account the error in the measurement of the cross sections ${}_{20}\sigma_{02}$ for Ar^{3+}, which equals ±20% in our work, ±15% in [1], ±20% in [5], and exceed ±20% in [6], we can regard the experi-

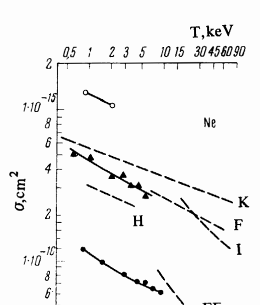

FIG. 2. Dependence of the effective cross sections ${}_{n0}\sigma_{0n}$ of Ne ions on the velocity v and on the kinetic energy T of the ions. Cross section ${}_{10}\sigma_{01}$: ○—present data. Cross section ${}_{20}\sigma_{02}$: ▲—present data, H—data from [5], F—from [1], I—from [4], K—calculated data from [9]. Cross section ${}_{30}\sigma_{03}$: ●—present data, FF—from [2].

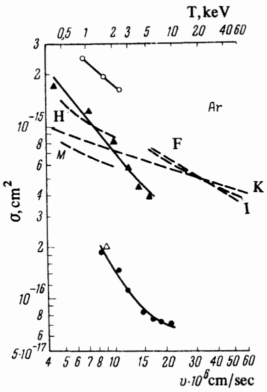

FIG. 3.

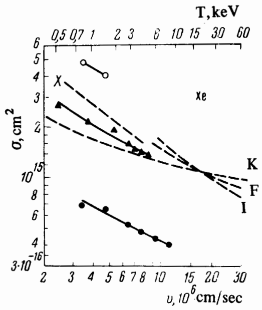

FIG. 4.

FIG. 3. Dependence of the cross sections $_{n0}\sigma_{0n}$ of Ar ions on the velocity v and the energy T. Cross section $_{10}\sigma_{01}$: ○—present data, cross section $_{20}\sigma_{02}$: ▲—present data, H—data from [5], M—from [6], F—from [1], I—from [4], K—calculated data from [9]. Cross section $_{30}\sigma_{03}$: ●—present data, Δ—from [6].

FIG. 4. Dependence of the cross sections $_{n0}\sigma_{0n}$ of Xe ions on the velocity v and the energy T. Cross section $_{10}\sigma_{01}$: ○—present data. Cross section $_{20}\sigma_{02}$: ▲—present data, H—from [5], F—from [1], I—from [4], K—from [9]. Cross section $_{30}\sigma_{03}$: ●—present data.

mental data as being in agreement. However, the $_{20}\sigma_{02}(v)$ curve has a larger slope than in the cited papers. The reason for the stronger increase of the cross section $_{20}\sigma_{02}$ with decreasing velocity of the Ar^{2+} ions is still unclear.

The effective charge exchange cross sections of triply-charged ions, with transition of three electrons in each collision act at energy 0.75–9 keV, which were measured in the present investigation, are new data. It can be noted only that a very rough estimate of the value of $_{30}\sigma_{03}$ for the case of Ar^{3+}, given by McGowan and Kerwin[6] at a single value of the energy T = 1.8 keV, agrees within the limits of the measurement error with the corresponding value obtained in the present work (Fig. 3). The variation of the $_{30}\sigma_{03}(v)$ curve and of the values of the cross sections for the Ne^{3+}–Ne pair, obtained in the present paper, agree within the limits of measurement accuracy with the previously obtained data.[2] Just as in the case of single- and two-electron resonant charge exchange, the cross sections $_{30}\sigma_{03}$ of three-electron charge exchange increase with decreasing velocity v, and at a fixed value of the velocity of the cross sections $_{30}\sigma_{03}$ increase with the increasing atomic number of the colliding particles. Thus, for example, at $v = 1 \times 10^7$ cm/sec we have $_{30}\sigma_{03} = 1.1 \times 10^{-16}$ cm² for the Ne^{3+}–Ne pair, 1.5 $\times 10^{-16}$ cm² for the Ar^{3+}–Ar pair, and 4.3×10^{-16} cm² for the Xe^{3+}–Xe pair. Thus, relatively large values of the cross sections $_{30}\sigma_{03}$ and the drooping character of the $_{30}\sigma_{03}(v)$ curves lead to the conclusion that the three-electron charge exchange of Ne^{3+}, Ar^{3+} and Xe^{3+} has the character of symmetrical resonant charge exchange in the investigated energy interval.

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