Resonant charge exchange of alkali-earth ions

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The total cross sections for resonant charge exchange of Hg^+, Ca^+ Sr^+, and Ba^+ ions are investigated experimentally in the 6–1000 eV energy range. The measurements are performed by the intersecting ion and atomic beam technique, in which a new method of extracting and recording the slow charge-exchange ions is used. On the whole, the resonant charge-exchange cross sections increase monotonically with decrease of interaction energy, and at an energy of 6 eV they reach 1.5×10^{-14}, 2.2×10^{-14} and 2.7×10^{-14} cm^2 for Mg^+ Ca^+, Sr^+, and Ba^+, respectively. Regular oscillations with frequencies 1.57×10^8, 1.57×10^8, and 0.86×10^8 cm/sec are observed for the $Ca^+ + Ca$, $Sr^+ + Sr$, and $Ba^+ + Ba$ processes respectively. The results are compared with calculations performed on basis of the Smirnov, Firsov, and Rapp–Francis formulas. It is suggested that the oscillations are due to the contribution of inelastic transitions to the total cross section.

Many experiments have been recently performed on resonant charge exchange of the ions of hydrogen [1], helium [2], and alkali elements [3-8]. The energy dependences of the cross sections of the process have in many cases presented an oscillatory structure [9-12]. However, in spite of the seeming abundance of studies, the experimental data on resonant charge exchange, especially at electron-volt energies, are still scanty, and none are available at all, for example, for Ca^+, Sr^+, and Ba^+ ions. The purpose of the present study was to investigate resonant charge exchange of the ions Mg^+, Ca^+, Sr^+, and Ba^+ at energies below 1000 eV.

EXPERIMENTAL TECHNIQUE

A block diagram of the setup is shown in Fig. 1. Its principal units are a metallic vacuum chamber, an ion source, an ion-beam shaping system, an ion receiver, a neutral-atom source, a system for extracting and registering the slow charge-exchange ions, and a power supply.

The ion source (see [13]) operates either in the arc-discharge regime (in the case of Mg) or in the surface-ionization regime (Ca, Sr, Ba). The ions produced in the source are extracted, accelerated, and focused into a cylindrical beam by an ion-optical system consisting of an immersion objective, two single lenses, and a set of collimating diaphragms. Such a system permits good focusing of the ion beam in the energy interval ~ 10—1000 eV. The atomic beam is produced by an effusion-type source, is shaped by a system of slits, and, after passing through the collision region, is condensed on a trap cooled with liquid nitrogen.

In our experiment, the slow charge-exchange ions were for the first time collected with a quadrupole capacitor (see Fig. 1). A small constant potential difference was applied to the quadrupole electrodes in such a way that the potentials of two neighboring electrodes were the same in magnitude but opposite in sign. This leads to the appearance of an electric field in the space of the quadrupole capacitor; this field is transverse to the ion beam and has a hyperbolic axially-symmetrical distribution. Since there is practically no electric field along and near the quadrupole axis, it is possible to pass an ion beam with very low energy, on the order of several electron volts, through the quadrupole.

On the other hand, the slow charge-exchange ions produced in the collisions (and also the electrons and ions produced as a result of the accompanying inelastic processes) are extracted by the electric field, which increases towards the quadrupole electrodes, and are gathered by the corresponding electrode pairs. Thus, the method realized in our experiment for the detection of the charge-exchange ions combines the advantages of the parallel-plate capacitor method and those of the cylinder method, and eliminates the shortcomings that these methods possess when used alone [14].

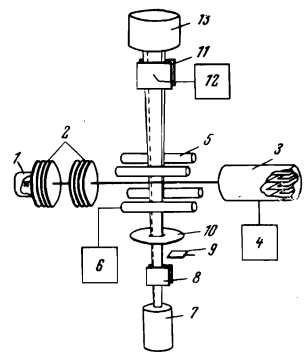

FIG. 1. Block diagram of experimental setup: 1—ion source, 2—ion-optical system, 3—ion receiver, 4—block for registration of primary ions, 5—quadrupole capacitor, 6—block for registration of charge-exchange ions, 7—atom source, 8 and 11—parallel-plate capacitors, 9—shutter to block the atom beam, 10—collimating diaphragm, 12—block for registration of the ions produced by electron impact, 13—liquid-nitrogen-cooled trap.

The electronic block for the measurement of the currents includes electrometric amplifiers with sensitivity ~ 10^{-13} A and electronic digital voltmeters and ammeters.

The charge exchange was investigated in two stages: 1) we measured the dependence of the efficiency of the process on the bombarding-ion energy; 2) we determined the absolute values of the cross sections. The errors in the determination of the energy dependence of the charge-exchange cross sections can result from incomplete gathering of the charge-exchange ions, from changes in the geometry of the ion beam with changing energy, from background effects, and from the occurrence of competing processes such as ionization. The extent to which all the charge-exchange ions were gathered was assessed from the saturation of the charge-exchange current following an increase in the potential difference between the quadrupole electrodes; clear-cut saturation was observed at a potential difference as low as 3—4 V. The influence of the ion-beam geometry was eliminated, inasmuch as the width of the atom beam exceeded the diameter of the ion beam by 2.5 times at the intersection point. The background effects due to charge exchange of the ions by the residual gas were eliminated

with the aid of the shutter. The signal/noise ratio at energies above 100 eV was approximately 3—4, and dropped to 0.1 with decreasing energy. The fraction of the ions produced by the competing process of ionization of the target atoms by ion impact was taken into account by measuring the corresponding electron current.

The absolute charge-exchange cross sections were obtained by calibrating the measuring apparatus against the known cross sections for the ionization of the investigated atoms by electron impact. This method was chosen because, first, direct and highly reliable determination of the concentration of the atoms in the beam is a subject for a separate investigation and, second, there are sufficiently reliable data on the effective cross sections for the ionization of Mg, Ca, Sr, and Ba atoms by electron impact [15,16]. In this case the procedure for determining the absolute cross sections reduces to measurement of the current of the charge-exchange ions and the current of the ions produced by electron impact. To this end, under the same experimental conditions, the ions are drawn out from the ion source first and passed through the interaction region, and are followed by the electrons. The cross section for the resonant charge exchange is then determined from the simple formula

$$Q=Q_{\text{ion}}Ji_e/J'i_i, \qquad (1)$$

where Q_{ion} is the known cross section for the ionization of the atom by electron impact, J is the current of the slow charge-exchange ions, J′ is the current of the atoms produced by ionization, i_i is the ion-beam current, and i_e is the electron-beam current. The ions produced by ionization were gathered on one of the electrodes of a parallel-plate capacitor placed in the path of the atomic beam (see Fig. 1).

RESULTS AND DISCUSSION

The measured cross sections for resonant charge exchange of the ions Mg^+, Ca^+, Sr^+, and Ba^+ in the energy interval E = 6—1000 eV are shown in Fig. 2. The measurements were performed at a primary-ion beam current $(2—8)\times 10^{-8}$ A. The energy scatter of the ions in the beam was 2—4 eV at the half-width of the distribution

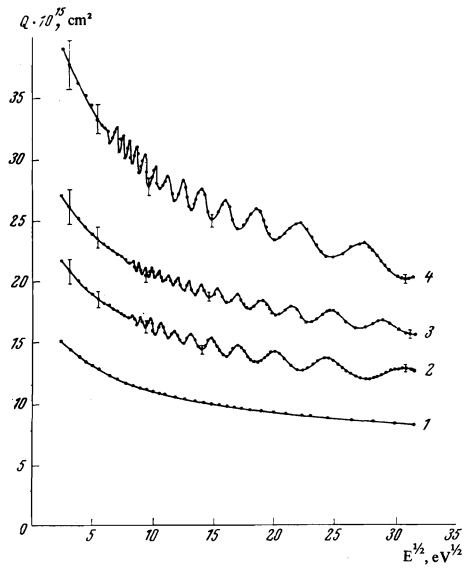

FIG. 2. Cross sections of resonant charge exchange of the ions Mg^+ (curve 1), Ca^+ (2), Sr^+ (3), and Ba^+ (4).

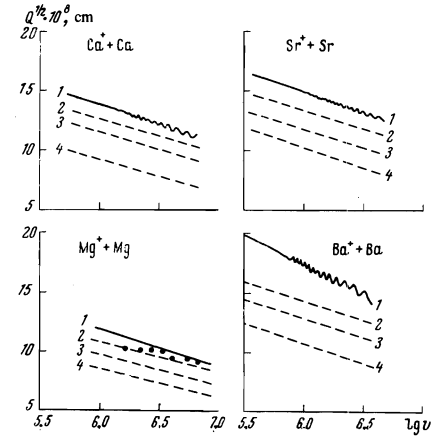

FIG. 3. Comparison of experiment with theory: 1—present result, 2—calculation in accordance with the Smirnov formula [18], 3—calculation in accordance with the Firsov formula [17], 4—calculation in accordance with the formula of Rapp and Francis [19], dark circles—experimental data on magnesium, taken from [20].

curve. The concentration of the atomic beam at the intersection point was $\sim 10^{11}$ cm^{-3}. The experiment was performed under high-vacuum conditions: the pressure in the collision chamber was $(2—3)\times 10^{-7}$ Torr. The error in the determination of the relative value of the cross section was 2% at energies above 100 eV, and increased to 6—7% with decreasing energy. Each of the curves shown in Fig. 2 is the result of averaging 10—15 individual measurements. The absolute values of the cross sections were determined with an error of 30%, which includes the error in the initial data (see [16]) on the ionization of the Mg, Ca, Sr, and Ba atoms by electron impact.

It is seen from Fig. 2 that the effective charge-exchange cross section increases in general with decreasing interaction energy and increases gradually on going from Mg^+ to Ba^+. However, whereas in the process $Mg^+ + Mg$ the cross section increases smoothly with energy, in the processes $Ca^+ + Ca$, $Sr^+ + Sr$, and $Ba^+ + Ba$ the energy dependences of the cross section consist of two components, monotonic and oscillating. The amplitude of the oscillations is approximately 3—4% of the average cross section. At energies 40—200 eV, the amplitude slightly exceeds the limits of the measurement errors, so that it is impossible to assess the amplitude in this case with assurance. At energies below 40—60 eV, on the other hand, it is very difficult to discern the structure.

Without allowance for the oscillatory structure, the measured curves agree well with the known expressions for resonant charge exchange [14], which reduce to the relation

$$Q^{1/2}=a-b\lg v, \qquad (2)$$

where Q is the cross section of the process, v is the relative velocity of the colliding particles, and a and b are constants. Moreover, the absolute values of the cross sections are in satisfactory agreement with calculations performed on the basis of the formulas of Firsov [17], Smirnov [18], and Rapp and Francis [19]. Experiment was compared with theory in Fig. 3 (which also shows the experimental data of [20] on the charge exchange of Mg^+). It is seen that from the point of view of absolute value, experiment agrees best with the curves calculated in accordance with Smirnov, whereas

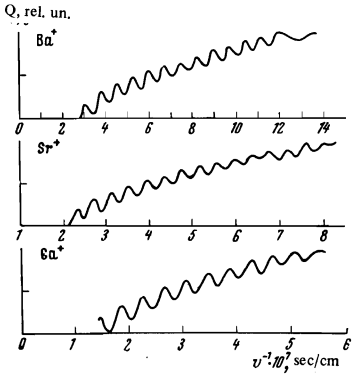

FIG. 4. Cross sections of resonant charge exchange of Ca^+, Sr^+, and Ba^+ as functions of the reciprocal velocity: a–Ba^+, b–Sr^+, c–Ca^+.

Element	$\beta \cdot 10^{-8}$ cm/sec	δ
Ca	1.57	−1.51
Sr	1.57	−2.04
Ba	0.86	−4.39

calculations in accordance with Rapp and Francis underestimate the results significantly. It is typical that the experimental values of the cross sections are higher than the theoretical ones for all the elements. The difference between the experimental and theoretical values increases on going from Mg^+ to Ba^+. This result does not contradict the tendency of the experimental cross sections to exceed the theoretical ones with decreasing ionization potential of the target atom [21].

An important feature of the observed oscillatory structure is its regularity. Figure 4 shows the experimental results on Ca^+, Sr^+, and Ba^+ as functions of the reciprocal velocities. We see that the extrema are distinctly equidistant. The oscillating component of the cross section is then well described by the analytic expression

$$Q_{osc}=cv^{\alpha}\cos(\beta v^{-1}-\delta), \tag{3}$$

where v is the incident-ion velocity, c and α are constants, β is the oscillation frequency, and δ is a phase constant. The values of β and δ are given in the table.

On the basis of the foregoing, the dependence of the total cross section for the resonant charge exchange of the ions Ca^+, Sr^+, and Ba^+ on the velocity, obtained by us above, can be represented in the form

$$Q=Q_M-Q_{osc} \tag{4}$$

(here Q_M is the monotonic component of the cross section).

The mechanism that gives rise to the experimentally observed oscillations can be explained from two fundamentally different points of view. It can be assumed that inelastic transitions are not very likely in resonant charge exchange processes, or, to the contrary, it can be assumed that they play an essential role. On the basis of the first assumption, Smith [9] showed that the oscillations can appear in the total cross sections of resonant processes if an extremum exists for the difference between the terms of the odd and even states of the produced quasimolecule. In light of this model, however, it seems strange that there are no oscillations in the $Mg^+ + Mg$ process.

It is perfectly possible that the observed oscillatory structure is due to transitions to an excited state, i.e., to the presence of nonresonant reaction channels which have small energy defects. In this case the oscillations can be due either to an extremal difference between the terms of the quasimolecules [11], or to competition of inelastic transitions of the type

$$A^+ + A \begin{cases} \nearrow A^* + A^+ + \Delta\varepsilon_1 \\ \searrow A^{+*} + A + \Delta\varepsilon_2, \end{cases} \tag{5}$$

where the asterisk designates excited states of the particles. In our experiment, the energy defects of the processes (5) amount to 0.6–2 eV (with the exception of the $Mg^+ + Mg$ pair), so that the probability of a transition to the excited state can be appreciable (see [22]). In view of the fact that the difference of the energy defects of the processes (5) is equal to 0.18, 0.04, and 0.25 eV for the pairs $Ca^+ + Ca$, $Sr^+ + Sr$, and $Ba^+ + Ba$, respectively, it is quite probable that the quasimolecule terms interact at large internuclear distances when the particles move apart, and this leads to regular oscillations (see [23]).

As to the $Mg^+ + Mg$ pair, the energy defects of the processes (5), as well as their difference, are much larger in this case. This lowers the contribution of the charge exchange to the excited state. The absence of an oscillatory structure in the $Mg^+ + Mg$ process thus becomes natural. A direct confirmation of the last mechanism would be the observation of regular oscillations on the energy dependences of the processes (5). Such experiments, however, entail great difficulties, since the deepest states of Mg I and Mg II are metastable.

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