Electron capture from helium atoms into various electronic states by multiply charged argon ions

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(Submitted 30 January 1986)
Zh. Eksp. Teor. Fiz. **91**, 465–476 (August 1986)

The cross sections $\sigma_{0,1}^{z,z-1}$ for one-electron capture $Ar^{+z} + He \rightarrow Ar^{+(z-1)} + He^{+}$ and $\sigma_{0,2}^{z,z-2}$ for two-electron capture $Ar^{+z} + He \rightarrow Ar^{+(z-2)} + He^{+2}$ were measured directly, for the first time ever, by recording the coincidences between the particles participating in one Ar^{+z}–$He(z = 3 - 8)$ collision act. The range of the kinetic energies of the incident ions was $E = (1–40)z$. The populations of different electronic states of the $Ar^{+(z-1)}$ ions produced in one-electron captures, and Ar^{+4} produced in two-electron capture, were determined by measuring the kinetic-energy spectra of the particles after the interaction. The cross sections for production of Ar^{+5} ions in various electronic states in the process $Ar^{+5}(3s^2) + He(1s^2) \rightarrow Ar^{+5}(n,l) + He^{+}(1s)$ are calculated on the basis of the quasimolecular model of particle interaction, using two-electron wave functions. It is shown that the procedure proposed for the calculation of the cross sections of the processes in the interaction of many-electron particles yields results that agree well with the experimental data.

I. INTRODUCTION

When multiply charged ions collide with atoms at approach velocities $v < 1$ a.u. the probability of particle production in definite final charge and electronic states is determined to a considerable degree by the potential energy of the excited quasimolecular system produced when the particles come close together. The excited states of the quasimolecule are due to the presence of unoccupied electronic levels of the multiply charged ion. As a result, electronic transitions from the initial state into a host of final excited states are possible even at low approach velocities. The study of such a multichannel interaction between a multiply charged ion and an atom is of interest for the physics of atomic collisions and for its applications. Most available data, however, are experimental cross sections for the capture of one or several electrons by a multiply charged ion, without analysis of the ion's final electronic states and without recording the slow-ion gas target produced from the atom.[1,2] In recent studies, optical spectroscopy methods were used to determine the emission cross sections for individual transitions in excited multiply charged ions, produced when an electron is captured from a helium atom or hydrogen molecule by an ion of charge $z = 6$ (Ref. 3). Determination of the populations of the electronic states by this method calls for the use of rather intense beams of multiply charged ions and for mandatory measurement of the cross sections for the emission of a whole set of individual lines. The number of such studies is therefore small. The use of another method, based on analysis of the change of the kinetic energy of ions that capture electrons, yields directly the pattern of population of the individual electronic states,[4] but the absolute values of the cross sections were not determined in an overwhelming majority of the studies.[5]

To verify and further develop the theoretical premises concerning collisions, it is important to measure the absolute values of the cross sections for population of individual electronic states during the actual change of the charge states of both colliding particles. It is also necessary to study the dependence of these quantities on the particles approach velocity v for different charge states z of the incident ion.

The theoretical papers deal mainly with collisions between nuclei and hydrogen atoms. The reason is that programs for the calculation of the molecular terms and of the interaction matrix elements are available for a one-electron system with two Coulomb centers.[1] The problem is much more complicated in the case of a many-electron system. One method of solving it is to use a model potential that comes closest to describing the real form of the potential, but permits the variables to be separated in the equation, just as in the case of the one-electron system with two Coulomb centers. This method was used by Harel and Salin[6] to calculate the cross sections for one- and two-electron capture from helium atoms by O^{+8} nuclei, and by Nikulin and Samoylov[7] to calculate the terms and matrix elements of the interaction of states of a system.

In the present paper, a coincidence method is used to record both interacting particles,[8] in order to isolate the one- and two-electron capture processes; the electronic states of the particles after the collision were determined by analyzing the collision kinematics. The absolute values of the cross sections of these processes were measured for collisions between Ar^{+2} atoms with charges z from 3 to 8 and helium atoms, in the velocity range $v = (1 - 12) \cdot 10^7$ cm/s corresponding to an incident-ion kinetic energy $E = (1 - 40)z$ keV. We determined the population of the electronic states produced in the course of one-electron capture of the ions $Ar^{+2} - Ar^{+6}$, and also in the course of two-electron capture of Ar^{+4} ions.

The one-electron wave functions of the quasimolecule were obtained by using a model potential[7] that yields correct values of the electronic levels inside the combined atom and the separated particles, and permits the variables of the Schrödinger equation to be separated. These wave functions

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were used to construct two-electron [Σ states of $(ArHe)^{+6}$] wave functions with which the populations were calculated of the $4d$, $4s$, and $4p$ states of the Ar^{+5} ion produced in the process $Ar^{+6}(3s^2) + He(1s^2) \rightarrow Ar^{+5}(3s^2,nl) + He^{+}(1s)$. The validity of this approach is confirmed by the good agreement between the calculated and experimental data.

II. EXPERIMENTAL PROCEDURE

1. Measurement of the cross sections for the elementary changes of the particle charge states

The Ar^{+z} ions were produced in a source in which atoms were ionized by an electron beam, accelerated to a specified energy E in the range $(1–40)z$ keV, their charges z analyzed with a mass monochromator, and guided to two collision chambers aligned in tandem with the beam and filled with helium to pressures $\sim 10^{-5}$ Torr at which only single collisions were produced. In the first chamber, the cross sections for production of positive ions were measured by attaching the slow helium atoms produced in the target gas to the plates of a capacitor. In the second chamber, the cross sections of the elementary changes of the charge states of the particles were measured by a coincidence method. To this end, the slow He^{+} and He^{+2} ions were extracted from the chamber by an electric field and charge-separated by a magnetic analyzer attached to the chamber. The fast scattered ions $Ar^{+(z-k)}$, that passed through the chamber were charge-analyzed with an electrostatic analyzer. Both the fast and the slow ions were recorded in the individual-particle counting regime. The pulses corresponding to particles in definite charge states were fed to a coincidence circuit to separate the partners of one and the same collision act. From the measured numbers of coincidences for different charge states of the argon and helium particles it was possible to determine the ratio of the cross sections of the elementary processes. These data and the absolute values of the total cross sections for the production of positive He^{+} and He^{+2} ions yielded also the absolute values of the cross section for the elementary changes of the particle charge states.

The gas target used in the experiment was the isotope ^3He; this permitted the H_2^{+}-ion background to be eliminated when the He^{+2} ions were recorded.

The measurement errors in the cross sections for the elementary processes are $\sim 15\%$. About two-thirds of this value are due to errors in the measurement of the absolute values of the cross sections of the produced He^{+} and He^{+2}, and one-third is due to errors in the measurement of the cross-section ratios of the various elementary processes.

2. Measurement of populations of various electronic states of particles produced in the capture process

The electronic states were determined by measuring the kinetic energies of the particles before and after the collision. The initial and final energies of the fast particle were determined with an electrostatic analyzer accurate to $\sim 2.5 \cdot 10^{-4}$. The initial (thermal) energy of the target particles could be neglected, while the final energy was found from the mass ratio of the colliding particles and from the scattering angle of the fast particle. The difference between the summary kinetic energies before and after the collision yielded the inelastic energy loss. The states responsible for the formation of individual peaks in the inelastic-energy-loss spectrum were identified on the basis of Moore's tables[9] of ion-excitation energies. The largest number of peaks was observed in collisions between Ar^{+z} ions with incompletely removed $3p$ shells, viz., Ar^{+3}, Ar^{+4}, and Ar^{+5}.

Figure 1 shows the excitation-energy spectrum of the Ar^{+4} ions produced when Ar^{+5} ions capture one electron each from He atoms. It shows a wide peak corresponding to an excitation energy 15–18 eV and narrower peaks corresponding to 23, 27, and 37 eV. In addition, the 27-eV peak has a singularity near 31 eV. The excitation-energy range 15–40 eV corresponds to three electron-configuration states: $3s3p^3$, $3s^23p3d$ and $3s^23p4s$. The peak corresponding to the lowest excitation energy is related to the production of the

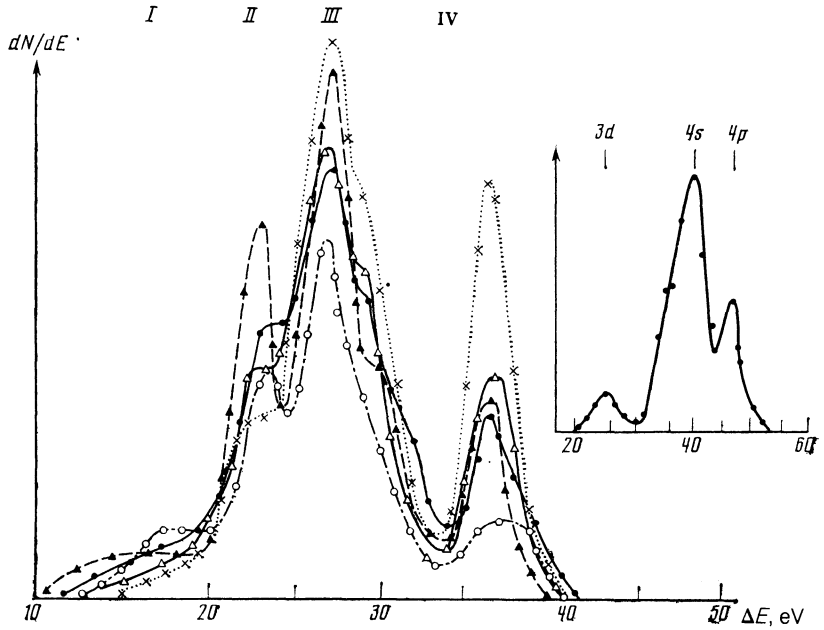

FIG. 1. Excitation-energy spectrum of Ar^{+4} and Ar^{+5} (inset) produced in the process $Ar^{+z} + He \rightarrow Ar^{+(z-1)} + He^{+}$. Peak I—$3s3p^3(^3D,^3P)$ states, peak II-$3s3p^3(^3S,^1P,^1D)$, peak III-$3s^23p3d(^3P,^3S,^3F)$ and $3s^23p3d(^1P,^1D,^1F)$, peak IV-$3s^23p4s(^3P,^1P)$; ×—incident-particle energy 5 keV, Δ—7.5 keV, ●—10 keV, ○—12.5 keV, ▲—15 keV.

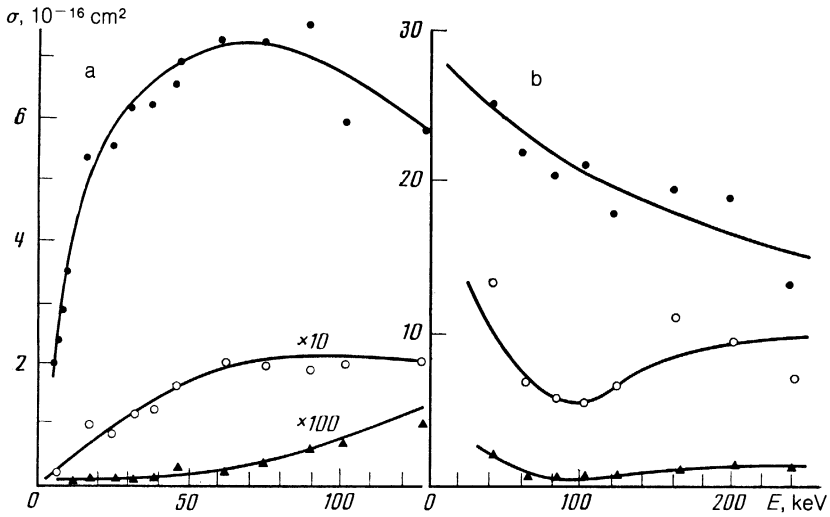

FIG. 2. Cross sections for the elementary changes of paticle charge states: ●—one-electron capture, ○—two-electron capture, ▲—capture of electron + ionization; a—Ar^{+3}–He pair, b—Ar^{+8}–He pair.

states $3s3p^3\ ^3D$ (excitation energy 15.06 eV) and $3s3p^3\ ^3P$ (17.57 eV), the second peak corresponds to the states $3s3p^3\ ^3S$(23.74, eV, $3s3p^3\ ^1P$(24.21 eV) and $3s3p^3\ ^1D$; the latter state is close in energy to the $3s3p^3\ ^1P$ state. The state $3s3p^3\ ^5S$ should be produced in the investigated collision-velocity region with low probability in view of the need for a change of the total spin of the system of colliding particles.

The third peak corresponds to ion production in the states (27.06 eV), $3s^23p3d\ ^3D$ (27.83 eV). Since the excitation energy $\approx$37 eV corresponding to the position of the fourth peak agrees well with the excitation energy of the states (37.35, eV the singularity near $\approx$31 eV can be attributed to formation of the states $3s^23p3d\ ^1D$ and $3s^23p3d\ ^1P$ whose excitation energy should be higher than that of the triplet states of the same ^3P and ^3D configuration, but lower than the excitation energy of the states $3s^23p4s\ ^3P$ and ^1P. Thus, the spectrum (Fig. 1) reflects all the possible Ar^{+4} electronic states possible in this energy region, and the energy positions of all peaks agree accurately with the tabulated Ar^{+4} excitation energies.

The identification of the peaks in the kinetic-energy spectra of Ar^{+2} and Ar^{+3} produced in collisions of Ar^{+3} and A^{+4} with helium atoms were similarly identified. In interactions of Ar^{+6} and Ar^{+7} ions with He, the spectra are simpler and consist of three well-resolved peaks (inset of Fig. 1). The excitation energies determined by measuring the kinetic energies of heavy particles agree well with the tabulated values. This procedure made it possible to determine directly the populations of levels with different excitation levels from several to hundreds of eV by measuring the absolute cross section of the elementary processes of the change of the charge states and of the ratio of the area under the peaks in the kinetic-energy spectra of the fast particles produced in the given elementary process. The resolving power of the procedure depends on the width of the instrumental function in the analysis of the energy spectrum of the particles. This width is determined by the number of components of the beam of multiply charged ions and by the resolving power of the analyzer. In our experiment the instrumental function, at an incident-ion energy $E \leqslant 3z$ keV, was determined mainly by the energy spread $\sim 0.4z$ eV of the primary particles extracted the ion source; at higher kinetic energies it was determined by the analyzer resolving power, which amounted to $\sim$6000.

III. MEASUREMENT RESULTS AND THEIR DISCUSSION

A characteristic feature of the electron capture by multiply charged ions is the high values of the capture cross sections (Fig. 2). In Ar^{+z}–He collisions the process with the largest cross sections, reaching $3\cdot10^{-15}$ cm^2, is one-electron capture. The role of two-electron capture increases rapidly with increasing ion charge z. Whereas for Ar^{+3} this cross section is several percent of the one-electron capture cross section, for Ar^{+8} it reaches $\sim$60%. The cross section for the capture + ionization process $Ar^{+z} + He \rightarrow Ar^{+(z-1)} + He^{+2} + e^-$ at large E reaches 10% of the one-electron capture cross sections for the Ar^{+8}–He pair. In the case of atom or molecule targets with electron binding energies lower than in He, the contribution of the capture + ionization process to the formation of $A^{+(z-1)}$ ions can be substantial. Therefore the use of a coincidence method to separate the actual change of the charge states of both particles after the collision ensures a correct determination, by the "collisional spectroscopy" method, of the populations of the electronic states of the ions produced in this process.

1. One-electron capture processes

Values of the cross sections

The cross section $\sigma_{0,1}^{z,z-1}$ for capture of one electron reaches a maximum in the velocity region $v = (2-4)\cdot10^7$ cm/s (Fig. 3). At $v > 7\cdot10^7$ cm/s a regular growth of the cross section is observed with increase of the incident-ion charge. However, while the cross sections have a general tendency to increase with the incident-ion charge, a noticeable deviation from the power-law dependence $\sigma \sim kz^{1+\alpha}; \alpha < 1$ (Ref. 10) is observed. This shows that the use of the scaling rules[10] to estimate the cross sections, espe-

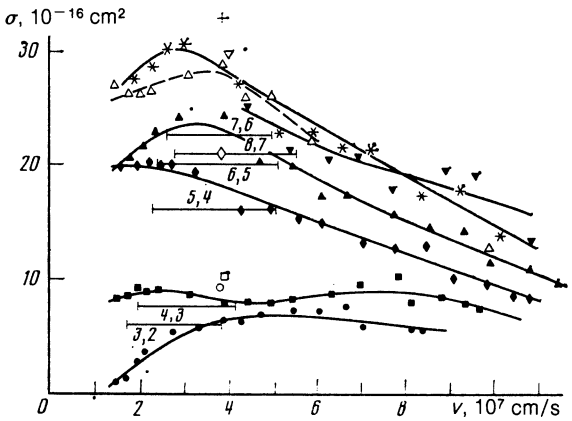

FIG. 3. Cross sections for one-electron capture in collision of Ar^{+2} ions with He atoms: Ar^{+3}—(●), Ar^{+4}—(■), Ar^{+5}—(◆), Ar^{+6}—(▲), Ar^{+7}—(▼), Ar^{+8}—(*) present data; Ar^{+3}—(○), Ar^{+4}—(□), Ar^{+5}—(◇), Ar^{+6}—(△), Ar^{+7}—(▽), Ar^{+8}— + data from Ref. 10 for $v = 4 \cdot 10^7$ cm/s and from Ref. 11 for $v = 10^8$ cm/s; segments $z,z-1$-data from Ref. 12.

cially near their maxima, can lead to substantial errors.

Our measured cross sections $\sigma_{0,1}^{z,z-1}$ for the capture of one electron and $\sigma_{0,2}^{z,z-1}$ for capture + ionization can be compared with the published data on the capture cross sections $\sigma^{z,z-1}$ obtained by analyzing the charge composition of only the fast particles, since $\sigma^{z,z-1} = \sigma_{0,1}^{z,z-1} + \sigma_{0,2}^{z,z-1}$ Figure 3 show the data of Müller and Salzborn[10] and of El-Sherbini *et al.*[11] Notice the good agreement between their results and our data. The cross sections measured by Bliman[12] also agree satisfactorily with our data, but the fact that in Ref. 12 the cross section is independent of velocity indicates that these cross sections have been measured with considerable errors exceeding the cross-section range in the velocity region investigated in Ref. 12.

State populations

Experiment. The populations of the electronic states of the ions produced in the capture process were measured for all the Ar^{+z}–He pairs (Fig. 4). The cross-section values indicate that in the investigated particle approach-velocity region the electron is effectively captured at large internuclear distances, resulting in formation of lower-charge ions in excited states. It should be noted that for ions with electrons in the $3p$ shell (Ar^{+3},Ar^{+4},Ar^{+5}) electron capture from a helium atom is accompanied with high probability by excitation of the core of the multiply charged ion, i.e., the capture is via a two-electron transition. Since even electron capture by an Ar^{+5} ion with ionization potential $\approx$91.3 eV is the result of such a two-electron transition, caution must be exercised when the laws governing the cross sections obtained in experiments with collision of many-electron particles[2] are extended to include capture processes accompanying collisions of elemental nuclei with hydrogen atoms.

In the case of collisions of Ar^{+7} (Fig. 4d) and Ar^{+6} (see Fig. 6 below), which have electrons only in the $3s$ subshell, the capture is exclusively via a one-electron transition. The proximity, observed in Ref. 3, of the cross section for emission resulting from $nl - n'l'$ transitions between similar levels of C^{+5},N^{+5},O^{+5} is evidence that in this case, too, one-electron capture does not lead to excitation of the core, and the cross section for electron capture by O^{+6} and N^{+6} is close to that for capture by a carbon nucleus. At the same time, the proximity of the cross sections for electron capture by multielectron-atom ions having the same charge[2] cannot serve as a basis for estimating the cross sections for electron capture by nuclei of the same charge, in view of the high probability that in the former case we have a two-electron process.

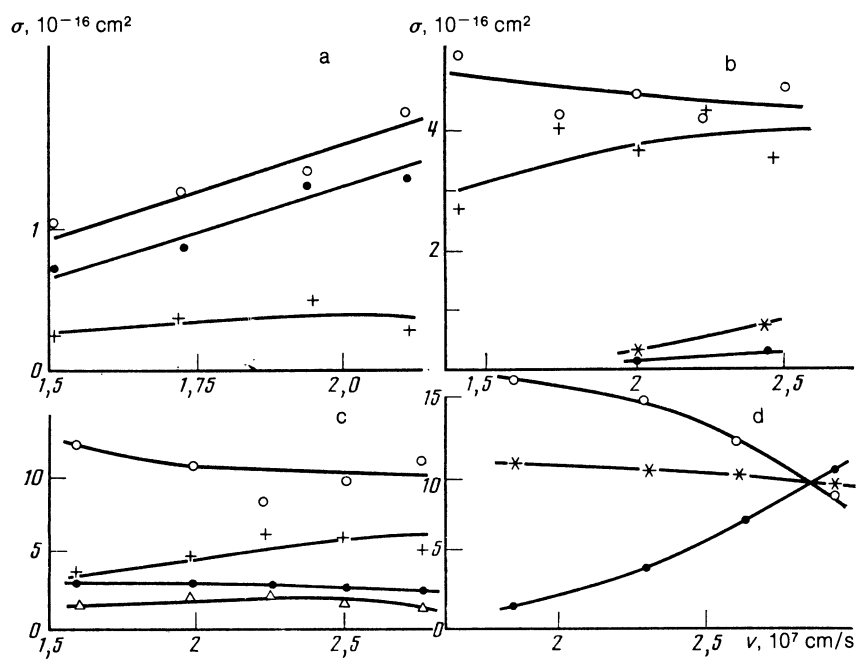

FIG. 4. Populations of various electronic states in one-electron capture: a) Ar^{+3}–He: + —states with configuration $3s3p^5$, ●—$3s^23p^4$, ○—$3s^23p^33d$, $3s^23p^34s$; b) Ar^{+4}-He: ●—$3s^23p^3$, *—$3s^23p^24s$, + —$3s3p^4$, ○—$3s^23p^23d$; c) Ar^{+5}-He: ●—$3s^23p4s$, + —$3s3p^{31}P$, ○—$3s^23p3d$, ▲—$3s3p^{33}P$; d) Ar^{+7}-He: ●—$4s$,*—$4p$, ○—$4d$.

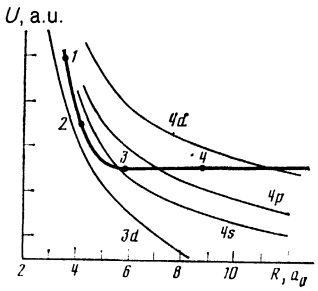

FIG. 5. Diabatic-term scheme of the quasimolecular system $(ArHe)^{+8}$: thick line—entrance term $Ar^{+6}(3s^2) + He(1s^2)$, thin lines $3d$, $4s$, $4p$, and $4d$ terms of the states $Ar^{+5}(3d) + He^+(1s)$, $Ar^{+5}(4s) + He^+$, $Ar^{+5}(4p) + He^+$, $Ar^{+5}(4d) + He^+$, respectively, corresponding to one-electron capture. Points 1, 2, 3, 4—locations, calculated in Ref. 14, of the crossing points of the entrance term and of the terms respective terms $Ar^{+4}(3p3d) + He^{+2}$, $Ar^{+4}(3p4s) + He^{+2}$, $Ar^{+4}(3d^2) + He^{+2}$, $Ar^{+4}(3d4s) + He^{+2}$, corresponding to two-electron capture.

One-electron capture accompanied by excitation of an ion core was observed by El-Sherbini and de Heer[11] in Ar^{+3}–He interactions. Emission was recorded of lines due to the decay of $3s3p^5\ ^1P$ and $3s3p^5\ ^3P$ states of Ar^{+2}. The populations of these levels, obtained in Ref. 11, agree with the populations measured by us. It can be seen from Fig. 4a, however, that the main contribution to the capture is made by channels connected with formation of other electronic configurations, viz., $3s^23p^4$, $3s^23p^33d$ and $3s^23p^34s$. Their emission was either below the sensitivity of the spectrometer of Ref. 11, or was nonexistent, just as in the case of Ar^{+2} formation in the ground state $3s^23p^4\ ^3P$. For all the remaining pairs, one-electron capture leads only to formation of $Ar^{+(z-1)}$ ions in excited states (Figs. 4b, c, d).

An advantage of our present procedure is that it permits measurement of the probabilities of populating all the electronic states observed in the course of ion capture, regardless of their excitation energy. This makes it possible to compare directly the calculated probabilities of the electronic transitions with the experimental data.

Calculation. The particle interaction in the investigated range of mutual approach velocities has a quasimolecular character. The transitions take place in the vicinities of points r_n of the quasicrossings of the initial $Ar^{+z} + He$ term with the terms of the final states $Ar^{+(z-1)} + He^+$. In the case chosen for the calculation of the Ar^{+6}–He pair, these regions are separated from one another (Fig. 5) and the electronic transitions can be treated in the two-level approximation. A system of diabatic terms was calculated for this pair on the basis of Ref. 13 and is shown in Fig. 5. The calculation took into account the interaction, in the course of the collisions, between two electrons initially contained in the He atom, with an He^{2+} nucleus, and the electronic core $Ar^{+6}(3s^2\ ^1S)$, assumed to be "frozen," as well as interaction of electrons with one another. (This is valid for Ar^{+6} core, as shown by experiment.) To construct the two-electron states of the quasimolecule $(Ar\,He)^{+6}$ we used the basis of diabatic molecular orbitals obtained by numerical solution of the two-center problem:

$$\left[-\frac{1}{2}\nabla^2-\left(\frac{z_1}{r_{1i}}+\frac{z_2}{r_{2i}}\right)+V_{\text{eff}}(\mathbf{r}_i,R)\right]\psi_i(\mathbf{r}_i,R)=\varepsilon_i\psi_i(\mathbf{r}_i,R), \tag{1}$$

where r_{1i} and r_{2i} are the distances from the electrons to the nuclei z_1 and z_2 of the ith electron, $\mathbf{r}_i$ are the electron coordinates ($i = 1,2$), R is the distance between the atoms, and V_{eff} is the effective potential, which takes into account the screening of the nuclei by two electrons, but admits of separation of the variables in Eq. (1), expressed in elongated spheroidal coordinates.[13]

The quasimolecular states were obtained in the one-configuration approximation, and their energy was calculated with allowance for the many-electron interaction.[7] It can be seen from Fig. 5 that the final-state terms corresponding to the $4s$, $4p$, and $4d$ states of the Ar^{+5} ion cross the entrance term at internuclear distances R_n equal to (5.8, 7.2, and 11.9) a_0. The term corresponding to the population of the $3d$ state of Ar^{+5} does not cross the entrance term but is close to it in the region $R \approx (4\text{–}5)a_0$. The diabatic crossings obtained agree with the calculation results of Ref. 14. Since the quasicrossing points are well separated, the difference ΔU between the adiabatic energies of the states in the vicinity of the points R_n were calculated in an approximation with only these two interacting configurations.

To determine the electronic-transition probabilities at the quasicrossing points R_n we used, for two interacting terms, Nikitin's model[15] in which the difference between the adiabatic energies in the vicinity of the points R_n was described by the expression

$$\Delta U(R)=\Delta\varepsilon\{1-2\cos\theta\exp[-\alpha(R-R_n)]+\exp[-2\alpha(R-R_n)]\}^{1/2} \tag{2}$$

The transition probability is then determined from the formula

$$P=\exp[-\xi(1-\cos\theta)]\ \text{sh}[\xi(1-\cos\theta)]\ \text{sh}^{-1}2\xi, \tag{3}$$

where $\xi = \pi\Delta\varepsilon/2\alpha v_R$ is the Massey parameter, $\Delta\varepsilon$ the resonance defect of the considered states, $v_R = v_0(1 - b^2/R_n^2)^{1/2}$ the radial velocity, b the impact parameter, and R_n the quasicrossing point. The parameters θ and α of the model were chosen such as to make the quantity ΔU, defined by Eq. (2), closest in the vicinity of the R_n point to the adiabatic-energy difference calculated by the method of Ref. 7.

The overall probability $P_{n,l}(b)$ of transitions to a definite state in one-electron capture with formation of an $Ar^{+5}(n,l)$ ion was calculated with allowance for the influence of the transitions to other states.[16] After integration with respect to the impact parameter, we determined the populations of various states of the Ar^{+5} ion (Fig. 6). Figure 6 shows also data from Ref. 14, where the potential curves and the splittings were obtained using a model potential similar to the one presented above. To simplify the calculations the $3d$-state population was determined with the aid of the

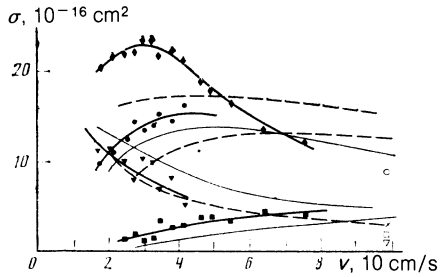

FIG. 6. Cross sections for formation of $Ar^{+5}(n,l)$ ions in one-electron capture: ●—state 4*s*, ▼—4*p*, ■—3*d*, ◆—total cross section: 4*s* + 4*p*; ○,▽, □—data from Ref. 11 for 4*s*, 4*p*, and 3*d* states, respectively. Thin continuous lines—calculation data from Ref. 14, dashed lines—our calculation data, the thick continuous lines are drawn through our experimental points.

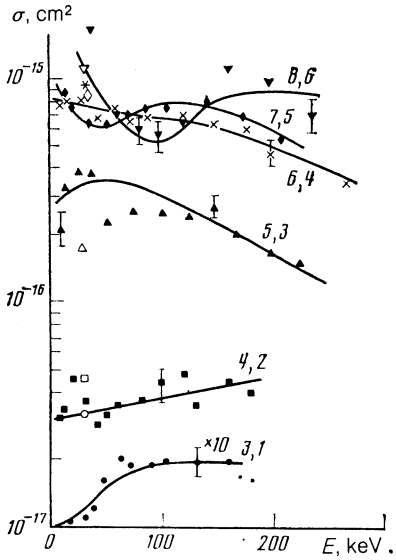

FIG. 7. Cross sections for capture of two electrons: ●—by ions Ar^{+3}, ■—Ar^{+4}, ▲—Ar^{+5}, ×—Ar^{+6}, ◆—Ar^{+7}, ▼—Ar^{+8}; ○, □, △, *, ◇, ▽—data from Ref. 10.

Landau-Zener model, while the 4*s*, 4*p*, and 4*d* states were determined by solving a system of coupled equations that takes into account the interaction of these terms with the initial-state term.

Comparison of the experimental and calculated data. It is seen from Fig. 6 that the results of our calculations and experiments agree satisfactorily with each other and with the calculations of Ref. 13. The main contribution to the cross section for electron capture is made, in the investigated velocity range, by the channel connected with the quasicrossing of ther terms at $R_n = 5.8a_0$ and leading to formation of $Ar^{+5}(s)$ ($\Delta\varepsilon = 0.94$ a.u., $\theta = 0.12$, $\alpha = 0.4$). The cross section for capture with formation of the ions $Ar^{+5}(4p)$ ($\Delta\varepsilon = 0.72$ a.u., $\theta = 0.04$, $\alpha = 0.21$) is determined by the quasicrossing at $R_n = 7.2a_0$ and reaches its maximum at velocities lower than $1\cdot10^7$ cm/s. The channel connected with the population of the 3*d* state makes a contribution that is small but increases with increasing velocity. Such a velocity dependence is due to absence of crossing of diabatic terms of the initial and final states. When the particles come close to the internuclear distance $R < 5a_0$, an appreciable energy gap remains between the terms, and it is this which causes the low probability of the electronic transition. No formation of ions in the state $Ar^{+5}(4d)$ is observed in experiment. Our calculation of the 4*d*-state population ($\Delta\varepsilon = 0.42$ a.u., $\theta = 0.002$, $\alpha = 0.23$) has shown the cross section for ion production in this state decreases with increasing approach velocity from $\sigma_{4d} = 10^{-17}$ cm² at $v = 2\cdot10^7$ cms to $\sigma_{4d} = 3\cdot10^{-18}$ cm² at $v = 10\cdot10^7$ cm/s. The low value of the cross section for ion formation in the 4*d* state, compared with the cross sections for other states, is attributed to the small term splitting at $R_n = 11.9a_0$, and as a result the system passes through this quasicrossing "diabatically" at the velocities investigated.

2. Two-electron capture processes

Values of cross sections

Data on the cross sections for these processes are shown in Fig. 7. In view of the high binding energy of the electrons in the helium atom ($\approx$79 eV) all the channels of the two-electron capture process are endothermal in the case of Ar^{+3} + He and the cross section of the process is relatively small ($\sim10^{-18}$ cm²). For the Ar^{+4} + He pair the exothermal process realizable in practice is capture of two electrons with formation of an Ar^{+2} ion in the ground state $3p^4$ ($\Delta E\approx21.5$ eV, quasicrossing point $R_n \approx 20a_0$). The second quasicrossing point, corresponding to formation of an Ar^{+2} ion in the lower excited state, is at a distance $R_n \approx 70a_0$, where the transition probability is small at the investigated collision velocities. The situation for the Ar^{+5} + He pair, however, is radically changed, in view of the presence of a large number of quasicrossings in the internuclear distance region where the electronic transitions are effective, and this leads to a strong increase of the cross sections for two-electron capture. For ions with $z \geqslant 6$ the number of possible process channels becomes large and its cross section reaches $\sim10^{-15}$ cm².

Population of states

We have measured the populations of various electronic states of Ar^{+4} ions produced by two-electron capture in the collision $Ar^{+6}(3s^2)$ + He (Fig. 8). The energy spectrum of the excited Ar^{+4} ions has peaks in the regions ΔE and this corresponds to formation of ions with electron configuration $3p^2$, $\Delta E\approx27$ eV–$3p3d$, $\Delta E\approx37$ eV–$3p3l$ and a broad peak in the region $\Delta E\approx50$ eV, corresponding to formation of Ar^{+4} ions in the states $3p5l$ and $3d^2$.

It follows from the data of Fig. 2 that the cross section for capture with ionization is small compared with the cross section for capture of two electrons, so that it can be assumed that capture of two electrons with formation of autoionizing states of Ar^{+4} ions (excitation energy $\Delta E\approx59$ eV) has low probability.

The excitation-energy spectrum (inset in Fig. 8) of Ar^{+4} ions produced by two-electron capture does not have

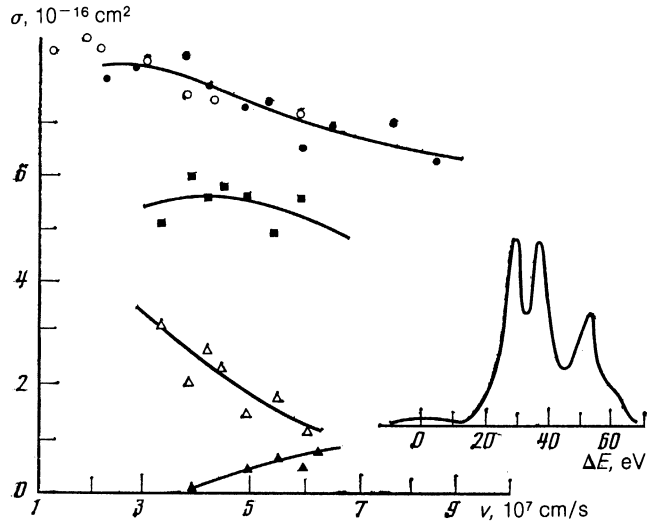

FIG. 8. Populations of various electronic states of Ar^{+4} ions produced in the course of the two-electron capture $Ar^{+6} + He \rightarrow Ar^{+4} + He^{+2}$: ●—total two-electron capture cross section, ○—the same from Ref. 10; ▲—cross section for production of Ar^{+4} ions in the state $3p^2$, ■—$3p3d$ and $3p4s$, △—$3p5l + 3d^2$. Inset—excitation-energy spectrum of Ar^{+4} ions.

some of the peaks present in the energy spectrum of the same ions but produced by one-electron capture by Ar^{+5} ions (Fig. 1). Figure 8 has no peak in the region of ≈ 15 eV, only the high-energy part corresponding to the singularity position is left of peak III in the energy region 25–35 eV, and there is no peak IV. This can be attributed to the fact that at the velocities investigated the total spin of the system should be conserved in an overwhelming part of the $Ar^{+6} + He$ collisions, and the Ar^{+4} ions are produced in this case only in the singlet state.

Two-electron capture mechanism

Two-electron capture can result either from two one-electron transitions in one collision act, with participation of an intermediate term ($Ar^{+5} + He^{+}$), or from a direct two-electron transition at the quasicrossing points of terms of the initial and final states. An attempt to take into account the contributions of both possibilities was made in Ref. 17. On the basis of these two models, the total cross section was calculated in Ref. 17 for two-electron capture in the Ar^{+6}–He pair, and was found to agree well with the experimental cross section. The total cross section, however, turned out to be insensitive to the type of model. The model is therefore best chosen on the basis of experimental data on the populations of the individual electronic states. Thus, population of the $3p^2$,$3p5l$, $3p3d$, $3d^2$ states can result only from a one-step two-electron transition. Indeed, as can be seen from Fig. 6, there is either no population of the possible intermediate term ($3p$), or the population probability is too small ($3d$). Favoring the direct two-electron transition are also the observed velocity dependences, typical of two-lavel Landau-Zener transitions, of the cross sections for population of terms having points of quasicrossing with the principal term for different R_n. As expected, in this case an increase of the $3p^2$-state population is observed with increasing velocity. At the same time, for the dependences of the populations of the $3p3d$ and $3p4s$ states, which occur at intermediate distances between the nuclei (points 1 and 2 on Fig. 5), a flat maximum is observed, and the cross section for populating the $3p5l$ and $3d^2$ states which occur at larger approach distances, decrease with increase of the colliding-particle velocity. This justifies the conclusion that the main mechanism for capture of two electrons is in this case a one-step two-electron transition.

The experiments reported in this paper have shown that study of the interaction between multiply charged ions and atoms by the coincidence method, with simultaneous analysis of the particle kinetic energies, yields sufficiently detailed information on the electronic transitions. The method makes it possible to distinguish between processes corresponding to individual electronic transitions, and to measure, with equal efficiency and over the entire excitation energy range, their absolute cross sections as functions of the colliding-particle velocities. Such experimental data are most convenient for comparison with calculation results. The calculations reported here show that an analysis of the one-electron capture process within the framework of the quasimolecular model, using two-electron wave functions, permits a satisfactory description of the experimental data on multielectron systems.

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Translated by J. G. Adashko