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Free electrons formation during adiabatic collisions of He^{2+} ions with xenon atoms

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The cross sections of various channels of the process of formation of free electrons occurring during adiabatic collisions of He^{2+} with xenon atoms $Xe(5s^25p^{61}S)$, that is, at velocities significantly lower than the velocities of electrons in the target atom outer shell, have been calculated. The dependences of the cross sections for direct ionization (DI) and quasi-molecular ionization or Auger ionization (QA) on the kinetic energy of the resulting electrons and the distance of approach of atomic particles at the moment of ionization have been determined. The calculated values of the cross sections of these ionization channels are compared with experimental data obtained from measurements of the cross sections of elementary processes of change charge states in collisions between He^{2+} and Xe.

Keywords: adiabatic collisions, direct ionization, quasi-molecular autoionization, diabatic terms.

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Introduction

Adiabatic collisions of alpha-particles with xenon atoms are accompanied by various elementary processes leading to the formation of electrons, and which can be written:

$$He^{2+} + Xe \rightarrow \begin{cases} He^{2+} + Xe^{n+} + ne^- & n\text{-multiple ionization } -220n \\ He^{(2-m)} + Xe^{n+} + (n-m)\ e^- & \text{capture } m\text{-electrons with ionization } -2(2-m)0n \end{cases}$$

In literature relating to ion-atom collisions to exclude long verbal names to designate the processes of changing the charge states of particles, we use values of particle charges before and after interaction. In our case, it is $2m0n$. First two numbers — charge states of the bombarding particle before and after the collision, the next two numbers — similar values of the charge states of the target particle.

The present paper objective is calculation of free electron formation cross-sections in direct ionization and Auger ionization processes, and their comparison with the experimental measured previously cross-sections of processes of charge states change of atom particles for pair $He^{2+}-Xe$, leading to the free electrons formation. Determination of applicability of available theoretical approaches to multielectron systems.

Processes of free electrons formation during interaction of alpha-particle with multi-electron xenon atom were studied by experiments using the various methods. Many of them are based on the analysis of state characteristics of heavy atomic particles associated with the process of free electrons formation. Registration of final charge states of atomic particles gives the number of formed free electrons. Measurement of kinetic energy of heavy particles after interaction provides data on energy used for ionization and kinetic energy of electrons. Experiments relating measurement of cross-sections differential by scattering angle of the ionization processes ensure determination of particles distance of approach, at which the formation of free electrons occurs.

In region of energies ($E < 10$ keV), corresponding to the adiabatic interaction of alpha-particles with Xe atoms, we measured absolute values of cross-sections of elementary processes of change in charge states of interacting particles leading to the formation of free electrons in ionization processes $\{220n\}$ $(n = 1-5)$, ionization with one electron capturing $\{210n\}$ $(n = 2-6)$ and ionization with two electrons capturing $\{200n\}$ $(n = 3-6)$ [1]. It is shown that basic mechanisms of formation of free electrons are processes of capturing of one or two electrons by the bombarding ion He^{2+} with formation of Xe^{n+} $n = 2, 3$. Total cross-section of formation of free electrons in the studied region of the collision energies increases upon decrease in speed of colliding particles. Analysis of measured differential cross-section of scattering He atoms formed during processes $\{200n\}$ $(n = 3-6)$ shows that ionization is effectively performed at distances of approach corresponding to maximum density $5p$- and $5s$-electrons of external shell of Xe atom [2].

The precision measurement of spectra of kinetic energies of He^+ ions formed during capturing of one electron with ionization {2102} showed that at energies of the bombarding ions He^{2+} $E < 10$ keV during this process the ion is formed in state $He^+(1s)$ [3]. The experimentally observed continuous spectrum of kinetic energies of ions He^+ shows that formation of free electron occurs in quasi-molecular $\{HeXe\}^{2+}$, at that the electron energy depends on internuclear distance at the time of its formation.

The alternative experimental approach associated with direct registration of free electrons ensures measurements of full cross-section of formation of free electrons and their energy spectrum. During ionization by photons the spectrum reflects the energy levels of atom [4,5], and during ionization during collision of ions with atoms or molecules — change in electronic levels of quasi-molecule depending on the internuclear distance, and electronic transitions between them with further ionization or interaction of terms with continuous spectrum. The measurements were performed for distribution of free electrons by their kinetic energies for ion-atom pairs [6–9], and upon interaction of ions and molecules with low number of electrons [10–13].

At collision speeds corresponding to the adiabatic interaction of partners, the basic channels of electrons formation can be direct ionization (DI) due to disintegration of quasi-molecular state exiting to continuous spectrum [14–21], and quasi-molecular autoionization or Auger-ionization (QA) [22,23]. The quasi-molecular autoionization occurs as result of resonant correlated two-electron transition between terms of the quasi-molecule.

The process of impact ionization contributes to the cross-section of the formation of free electrons only at collision speeds exceeding the speeds of electrons belonging to the outer shell of the atom. At the kinetic energy of ions He^{2+} below 10 keV, discussed in the present paper, the cross-section of this process is small.

1. Calculation procedure

During ion He^{2+} collision with xenon atom the quasi-molecular system $\{HeXe\}^{2+}$ is formed in excited state, which potential energy is sufficient for its ionization. Upon particles approaching the level of helium $1s$ correlates with level $4f$ of combined ion Ba^{2+} and at particles approaching crosses $5l$ terms of quasi-molecule. In region of these crosses the electron transitions in quasi-molecule occur. As a result of electron transition from the outer shell of Xe atom to $1s$-level of He^+ ion the energy is generated sufficient to remove one more electron from Xe atom. In this case the process of formation of free electrons is exothermic process and occurs without expenditure of kinetic energy. Possibility of transitions occurs only when definite distance of approach of nuclei of particles is achieved. At some distance of approach some quasi-molecular term can exit to the continuous spectrum. As result of disintegration the system direct ionization occurs.

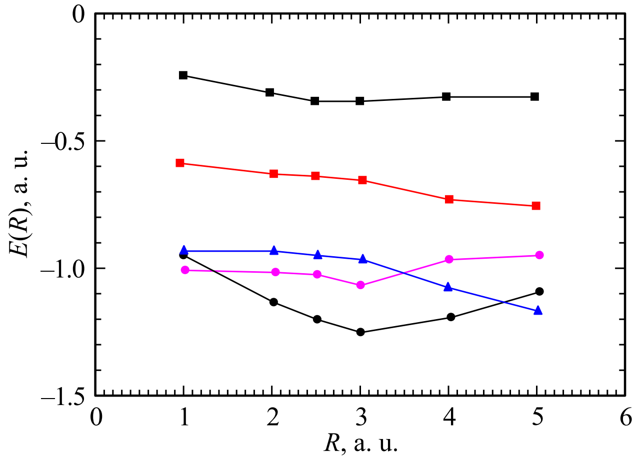

Figure 1. Two-electron adiabatic terms of quasi-molecule $\{HeXe\}^{2+}$. Term responsible for the Coulomb repulsion of particles is subtracted. ■ — $6d\sigma^2$, □ —$4f\sigma 6d\sigma$, Diabatic terms of this electronic configurations exit to the continuous spectrum (see references ([10,12]) and are responsible for the system direct ionization, △ — $4f\sigma^2$; ○ — $3d\sigma^2$, term responsible for quasi-molecular ionization QA1; ○ — $3p\sigma 6d\sigma$ — term responsible for quasi-molecular ionization QA2.

For analysis of the ionization mechanism during collision of xenon atoms with He^{2+} ion the results of paper [17,22] were used. Such analysis of formation of free electrons was performed in our paper for collision $^3He^{2+}$–Ar, in energy range of ions $5 - 10$ keV [24], and was used in present paper to analyze the mechanism of ionization processes in the quasi-molecular system $\{HeXe\}^{2+}$. Note that for this system the most effective process is capturing of one electrode with formation of ion $He^+(n = 2)$ due to low resonance defect [3] and exclusion of formation of free electrons.

To review processes resulting in the formation of free electrons we made calculations of terms of quasi-molecule and probabilities of transitions between them. Terms were calculated by solving the Schrödinger equation in prolate spheroidal coordinates as in our previous work [24]. The calculated two-electron diabatic terms of the quasi-molecule $\{HeXe\}^{2+}$ are given in Fig. 1.

The energy spectra of electrons were calculated using expressions obtained under the modified adiabatic approximation for direct ionization and perturbation theory for quasi-molecular autoionization.

Cross-sections differential by energy of electrons of the direct ionization process [17] are described by expressions (1), (2):

$$\frac{d\sigma}{dE} = \frac{4\pi|R(E)|^2 \mathrm{Im}R(E)}{\alpha(E)} \exp\left[-\frac{\alpha(E)}{V_R}\right], \qquad (1)$$

$$\alpha(E) = 2\int_{E_0}^{E} \mathrm{Im}(R(E')dE', \qquad (2)$$

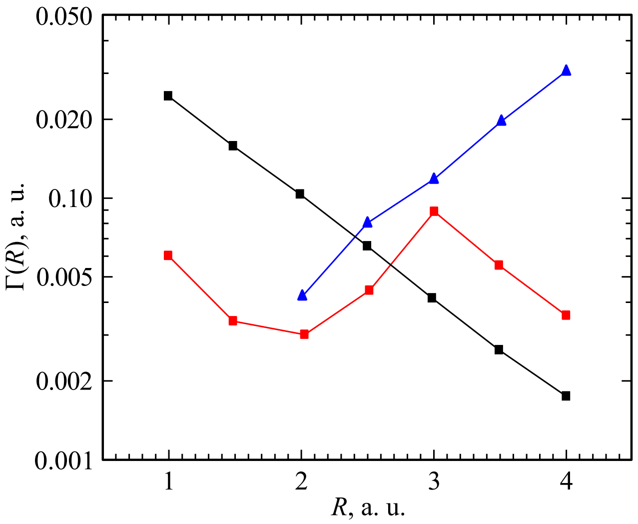

Figure 2. Widths of autoionization states: ■ — $6d\sigma^2{-}3d\sigma$, — direct ionization DI; □ — $4f\sigma 6d\sigma{-}3d\sigma$, quasi-molecular ionization QI; △ — $3p\sigma 6d\sigma{-}3d\sigma$ quasi-molecular ionization Q2.

where $R(E)$ — inverse function of a term $E(R)$; $\mathrm{Im}R(E)$ — imaginary part of this function; E_0 — limit value of energy (first quasi-intersection of the protruding diabatic term); V_R — radial speed of approaching of atomic particles.

Differential cross-section of the process of quasi-molecular autoinizatuion is described by expressions (3), (4) [22]:

$$\frac{d\sigma}{dE} = 2\pi \int 0^{\infty} N(b, R(E))W(E, b)bdb, \qquad (3)$$

$$W(E, b) = \left| \int_{\square}^{t} \sqrt{\frac{\Gamma(t)}{2\pi}} \exp\left\{ iEt - i \int_{\square}^{t} E_0(t')dt' \right\} dt \right|^2, \qquad (4)$$

where $N(b, R)$ — autoionization term population, $W(E, b)$ — probability of disintegration, t — time, $\Gamma(t)$ — natural width of term $E(t)$, b — impact parameter. During $\Gamma \ll 1$ the probability of disintegration can be presented as equation (5):

$$W(E, b) \approx \frac{\Gamma(t_0)}{E_t'(t_0)} = \frac{\Gamma(R_0)}{V_R E_R'(R_0)}, \qquad (5)$$

where t_0 and R_0 — time and internuclear distance corresponding to region of autoionization transition, V_R — speed, the apostrophe means the derivative.

Data on width of terms $\Gamma(R_0)$, leading to the formation of free electrons as function of internuclear distance are given in Fig. 2.

2. Results and discussion

Results of calculation of two times differential cross-sections of various channels of free electrons formation during collision of He^{2+} ions with Xe atoms are given in Fig. 3.

One of sources of free electrons formation is quasi-molecular autoionization [22] (Auger-ionization) which corresponds to the experimentally isolated process of capture with ionization {2012}. Formation of autoionization molecular ion can occur either as result of simultaneous transition of two electrons in point of quasi-intersection of terms of quasi-molecule $\{HeXe\}^{2+}$ (QA1) or during successive transition of two electrons in two different by internuclear distance points of quasi-intersection followed by Auger-disintegration of this quasi-molecular state of autoionizations (QA2):

$6d\sigma^2 \to 3d\sigma + e$, quasi-molecular autoionization (QA(1), Fig. 3)

$6d^2 \to 3p\sigma 6d\sigma \to 3d\sigma + e$, quasi-molecular autoionization (QA(2), Fig. 3).

The electrons formed during simultaneous transition of two electrons, contribute to low energy part of spectrum of electrons, and differential cross-section of the process of successive transition of two electrons has wide maximum with most probable energy of electrons ~ 25 eV.

The third source of the process of capturing with ionization is disintegration of the autoionization state of $He(2l^2)$ at internuclear distances corresponding to the limit of separated atoms, formed during capturing of two electrons. This is confirmed by peaks in the experimental measured curves of energy spectrum of electrons, corresponding to energy of state disintegration of $He(2s2p^{1,3}P)$ [23].

The calculated integral value of cross-sections of the process of one electron capturing with ionization leading

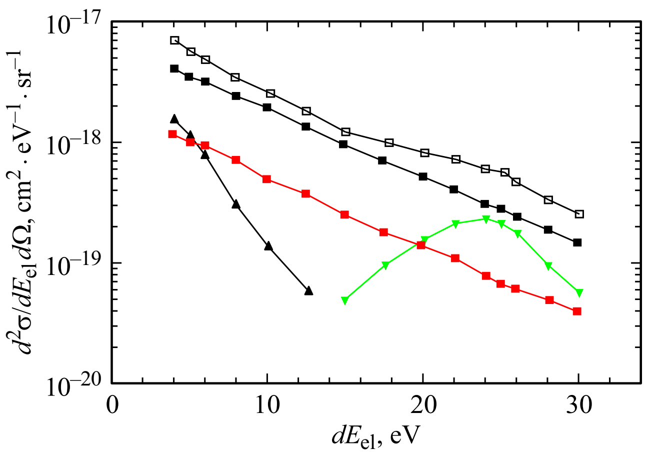

Figure 3. Differential cross-sections of various channels of free electrons formation during collision of He^{2+} ions with Xe atoms. Ion energy 10 keV. □ — Total, ■ — cross-section of direct ionization from initial state DI(1); □ — cross-section of direct ionization from state with one captured electron DI(2); ▲ — QA (1) — cross-section of Auger-ionization in case of simultaneous transition of two electrons to the autoionization term; ▽ — QA (2) — cross-section of Auger-ionization during successive transition of electrons.

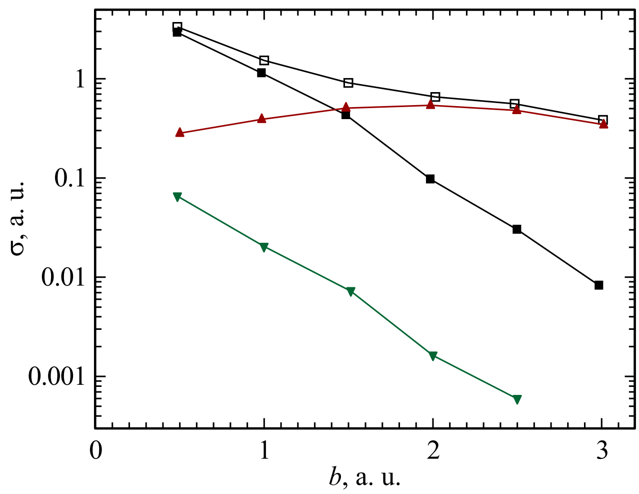

Figure 4. Differential cross-sections of various channels of free electrons formation vs. impact parameter. □ — Total, total cross-section of direct ionization and Auger-ionization; ■ — D1, cross-section of direct ionization — result of interaction of quasi-molecular term and free spectrum; ▲ — QA1, cross-section of Auger-ionization during successive transition of electrons to autoionization term; ▽ — QA2, cross-section of Auger-ionization during simultaneous transition of electrons.

to the formation of He^+ ion, Xe^{2+} ion and one free electron is $3.1 \cdot 10^{-16}\,cm^2$ at ion energy 10 keV.

The following channels contribute to the cross-section of capture with ionization: successive transition of two electrons — $2.4 \cdot 10^{-16}\,cm^2$, simultaneous transition of two electrons in quasi-molecule — $0.15 \cdot 10^{-16}\,cm^2$ (almost 100% per orbital $3d\sigma$) and capture of two electrons into autoionization state of $He(2l^2)$ — $0.55 \cdot 10^{-16}\,cm^2$. Value of cross-section of last channel of the process is determined experimentally [4] based on area of peaks corresponding to energy of electrons during disintegration $2l^2$ of states of helium atom.

Experimental measurements of the cross-section of capture with ionization {2012}, obtained by registration using coincidence method of formed ions Xe^{2+} and He^+ [1], give the value $2.7 \cdot 10^{-16}\,cm^2$. Note that during experiment we register only cases of interaction when only one electron is formed, and there is no additional ionization of xenon atom as result of multi-electron transitions. The theoretical calculation also does not take into account multiple ionization. As result good agreement between the theory and experiment is observed.

The direct ionization occurred as a result of single electron transitions from diabatic term to continuum state results in formation of electrons, which energy distribution has form of a decreasing curve with maximum at energy $E_{el} = 0$ eV. Based on the made calculations we can make the following picture of evolution of discussed system. State $6d\sigma^2$ of two-electron system upon approaching of colliding particles is subjected to capture of one electron $6d\sigma \to 3p\sigma$ due to Demkov bond ($R_c \approx 4$ a.u., probability of transition is $P = 0.25$). During further approaching of the particles the following processes occur, leading to ionization:

$6d\sigma^2 \to 6d\sigma + e$, direct ionization (DI(1), Fig. 3)

$3p\sigma 6d\sigma \to 3p\sigma + e$, direct ionization (DI(2), Fig. 3)

Calculations of cross-section of the direct ionization as per solutions of equations (3) and (4), at energy of ions He^{2+} 10 keV give value $\sigma(202n) = 6.4 \cdot 10^{-16}\,cm^2$. Theory of direct ionization — term disintegration upon exit to continuous spectrum — does not include the multi-electron processes.

The theoretical calculation shows (Fig. 3) that the largest contribution to the cross-section of electrons formation of direct ionization belongs to the process of state disintegration ($6d\sigma^2$), and the lower contribution is — from state with one captured electron ($3p\sigma 6d\sigma$). The probabilities of these ionization channels occurring as function of the impact parameter are presented in Fig. 4.

But such description not considering the multi-electron processes [17] is true only for systems with very limited number of electrons [7–10]. Experiment of measurement of cross-sections of elementary processes shows that the formed multielectron ion $\{HeXe\}^{2+}$ formed during the direct ionization further disintegrates with formation of ions of helium and xenon with different charge, and some additional free electrons. The value of cross-section of direct ionization theoretically obtained based on solution of equations (1) and (2) exceeds the sum of all experimental measured values of cross-sections of processes leading to the formation of free electrons.

The sum of cross-sections of such processes, obtained based on the registration of charge states of the formed ions, leading to formation of free electrons, is $4.49 \cdot 10^{-16}\,cm^2$, this is 70% only of calculated cross-section of the direct ionization. The significant difference in cross-sections can be due to inaccurate width calculation of the diabatic term exiting to the free spectrum (Fig. 2).

According to measurements of cross-sections of elementary processes [1], the major part of cross-section of free electrons formation is contributed by the process of capture of one electron leading to the formation of helium ion He^+ and having total cross-section $4.77 \cdot 10^{-16}\,cm^2$. The next process as per contribution to the cross-section value is process of capture of two electrons, their total cross-section is $4.4 \cdot 10^{-16}\,cm^2$. The minimum contribution belongs to the process of ionization {2021} without charge change of bombarding ion He^{2+} — $0.315 \cdot 10^{-16}\,cm^2$.

Conclusion

The theoretical calculation of cross-section of capture with ionization as result of population of the autoionization two-electron term and its further disintegration, and experimental measurement of the cross-section of one electron capture with formation of ion He^+, singly charged ion Xe^+ and one electron clearly distinguish one definite elementary process. In this case the obtained theoretical and experimental cross-sections show good agreement. However, the formation of free electrons as result of direct ionization of multi-electron quasi-molecule — term disintegration upon exiting into continuous spectrum — is only part of the cross-section of free electrons formation. The developed for systems with low number of electrons method of for determination of cross-section of ionization requires additional consideration of multi-electron process for the case of quasi-molecule $HeXe^{2+}$. The experimental data on absolute values of elementary processes of charge states change ensure distinguishing in its pure form of any process of the charge states change of particles associated with the formation of free electrons.

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Conflict of interest

The authors declare that they have no conflict of interest.

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