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Auger transition probabilities and electron emission cross sections for vacancy decay into the $2p\pi$-orbital in the Ne^+-Ne quasimolecule

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Received September 6, 2021
Revised September 6, 2021
Accepted October 4, 2021

The paper reports the calculation of the probabilities of Auger transitions taking place in the process of filling a vacancy on the $2p\pi$-orbital in an Ne^+-Ne quasimolecule, a short-lived system which arises when the ion and atom approach each other and decays when they fly apart. Calculations for various ionization degrees of the quasimolecule particles were performed for the first time. It was found out that the system ionization degree increases very significantly (from 2 to 6) with increasing collision energy and decreasing distance of the particles closest approach. Using of the quantum mechanical approach and taking into account the collision dynamics made it possible to quantitatively describe for the first time the experimental Auger electron spectra of a complex many-electron quasimolecule. The contribution of the transition from the initial $3d\pi-3d\pi$ state to the $2p\pi$ orbital was shown to be predominant among the whole variety of possible Auger decay channels.

Keywords: atomic collisions, quasimolecule, Auger transitions, electron emission cross sections, vacancy decay

DOI: 10.21883/TPL.2022.01.52475.19018

Auger transitions in quasimolecules were discovered in studying the Kr^+-Kr collisions at particle energies of a few keV [1]. It was established that, despite the short collision time ($10^{-15}-10^{-16}$ s) and, hence, short quasimolecule „lifetime“ there is possible Auger decay of vacancies on molecular levels whose energies change during the collision. Hence, the electron spectra exhibit a broad continuous band, unlike the spectrum of characteristic electrons emitted from atoms after collision, i.e., after scattering, which is of the discrete–line type. It became immediately apparent that spectroscopy of electrons (or photons) emitted during the collision is a unique source of information on processes in an extremely short–lived object just to which the quasimolecule belongs (all other methods provide information on the atoms – collision partners after flying apart when relaxation of excited states is already finished).

As our earlier paper [2] showed, electron spectra for the Ne^+-Ne collisions possess two components. When the electron energies are low, a spectrum exponentially decreasing with increasing electron energy is observed. This component makes the dominant contribution to the ionization cross section and is connected with the decay of autoionization states arising when the $4f\sigma$ orbital is promoted. When the electron energies exceed 2 a.u., electrons associated with the Auger decay of vacancies in the quasimolecule dominate in the spectra. While the particles approach each other, the $2p\pi$-orbital energy changes, due to which the electron spectrum becomes continuous. Among other studies of the continuous component of electron spectra in atomic collisions, works [3–10] should be noticed.

The goals of this study included calculation of probabilities of Auger transitions to the $2p\pi$ orbital for various Auger decay channels with participation of electrons of various outer orbitals, and the comparison between the calculations and measurements.

As shown in [1–3], analysis of the spectrum shape and its variation with increasing collision energy allows extracting from experimental data the dependence of the Auger transition energy on the achieved internuclear distance. Data of papers [2,3] are in good agreement with calculations of the molecular orbital energies [11] (Fig. 1). The $2p\pi$ orbital has a vacancy. The $2s\sigma$ orbital is fully occupied since it is formed from fully occupied $2s$ levels of the colliding atoms.

Let us use the data of paper [11]where diagrams of molecular orbitals were calculated for different charge states

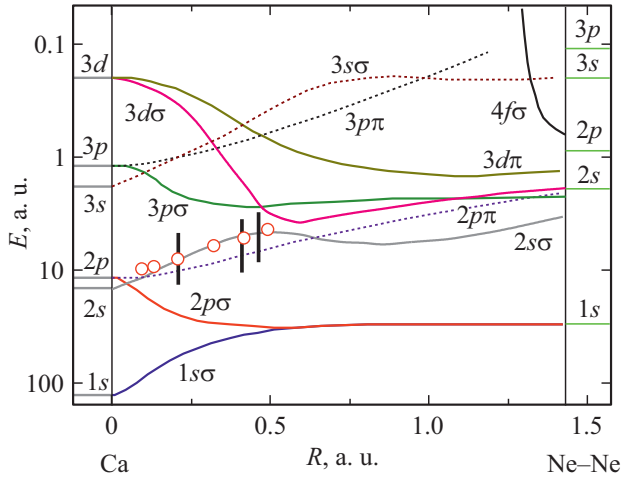

Figure 1. The experimentally obtained position of the effective term (point) [2,3] and the diagram of molecular orbitals for the $Ne^{2+}-Ne$ system [11].

of the Ne^{q+}–Ne ($q = 0, 2, 4$) system. Vacancies on the $2p\pi$ level may be filled from the $3p\sigma$ orbital (two electrons on the orbital), $3d\sigma$ orbital (two electrons), and $3d\pi$ orbital (four electrons). We assume that orbitals $3s\sigma$, $3p\pi$ and $3d\delta$ do not contain electrons since they are formed at large internuclear distances from unoccupied levels. Orbital $4f\sigma$ is emptied by electron transitions to continuum. Thus, there are six variants of Auger transition initial states: $3p\sigma^2-2p\pi\varepsilon$ (letter ε designates the outgoing electron), $3d\sigma^2-2p\pi\varepsilon$, $3d\pi^2-2p\pi\varepsilon$, $3p\sigma 3d\sigma-2p\pi\varepsilon$, $3p\sigma 3d\pi-2p\pi\varepsilon$ and $3d\sigma 3d\pi-2p\pi\varepsilon$. Further the Auger decay channels are designated by the initial states. The Auger transition energy E_i was calculated as a difference in energies of relevant orbitals $E_i(R) = E_1(R) - E_2(R) - E_3(R)$. Here $E_1(R)$ is the energy of the orbital whereto the electron transits, $E_2(R)$ is the energy of the orbital wherefrom the electron transits, $E_3(R)$ is the energy of the orbital wherefrom the electron is emitted to continuum.

The Auger transition probability per time unit may be defined as

$$W = \frac{2\pi}{\hbar}\left|\iint \chi_f^* \varphi_f^* \frac{e^2}{r_{1,2}} \chi_i \varphi_i d\tau_1 d\tau_2\right|^2,$$

where χ_i, φ_i are single–electron wave functions of each of two excited atomś electrons, χ_f is the wave function of the electron on the $2p\pi$-orbital, φ_f is the wave function of the Auger electron, i. e., of the free electron leaving the atom. Operator $\frac{e^2}{r_{1,2}}$ describes the Coulomb interaction of two electrons. In calculations for two electrons with identical spin directions, the direct and backward transition amplitudes and their interference were taken into account. After that, the probabilities of transitions from the states with different spins and momentum projections were summarized. As wave functions of the electron stationary states, hydrogen–like wave functions were used; in this case, effective charge Z_i changed with the internuclear distance in accordance with the changes in the orbital energy. Selection of hydrogen–like wave functions was reasonable because the considered transitions occur near the united atom limit while electrons stay in the field of a considerable effective charge of the united atom. The partial emission cross section of electrons with energy E may be calculated as follows [2,12]:

$$\frac{d\sigma}{dE} = 2^{5/2}\pi^2 f \int_0^{b(R)} \alpha^{-2/3} W(R) A_i^2\left\{\alpha^{-1/3}\left[E - E_0(R)\right]\right\} b db.$$

Here $E_0(R)$ is the internuclear distance dependence of the Auger transition energy, $W(R)$ is the transition probability, $f = 1/3$ is the probability of the vacancy presence on the $2p\pi$-orbital, $A_i(x)$ is the Airy function, b is the impact parameter. Parameter α is defined as

$$\alpha = \frac{1}{4}\frac{dE}{dR} v^2\left(-\frac{dU(R)}{dR}\frac{1}{E_{cm}} + 2\frac{b^2}{R^3}\right),$$

i. e., α depends on the term derivative dE/dR, increases with increasing collision velocity v, and also depends on impact parameter b and derivative of the interaction potential $U(R)$, E_{cm} is the collision energy in the center–of–mass system. For convenience, let us rewrite the emission cross section as follows:

$$\frac{d\sigma}{dE} = W(R) f G,$$

where factor G depends only on the term parameters, achieved internuclear distance R and impact parameter b.

As Fig. 2 shows, at the energy of 3 keV there is a good agreement between the calculations and measurements of the Auger electron emission cross sections in case the Auger transition probabilities and Auger electron energies calculated for the Ne^{2+}–Ne system at 6.25 keV are used; the agreement takes place also at the electron energies of 3–4 a.u. in case parameters for the Ne^{2+}–Ne system are used, while at the electron energies of 5–6 a.u. a

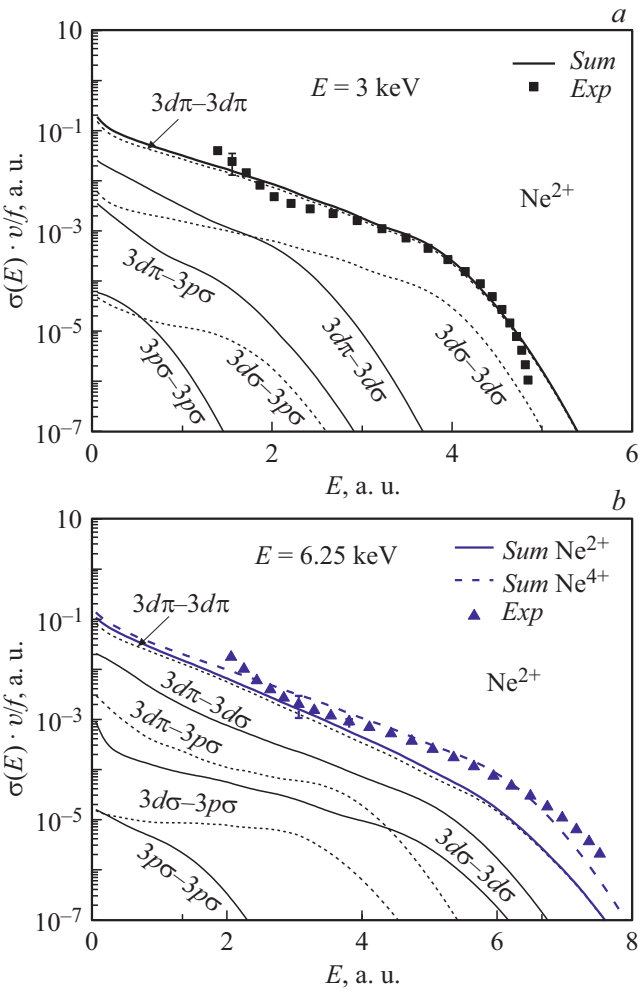

Figure 2. Comparison of the experimentally measured spectra with our calculations for the energies of 3 (*a*) and 6.25 keV (*b*). Fine solid and dotted lines represent the contributions of different channels for occupying the $2p\pi$ vacancy. Bold lines represent the total contribution of all the channels. Calculations at the energy of 6.25 keV are presented for systems Ne^{4+}–Ne and Ne^{2+}–Ne.

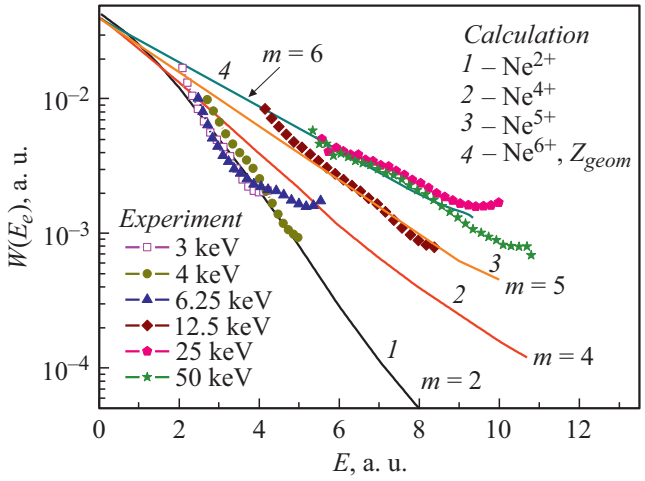

Figure 3. Comparison between calculations of the Auger transition probabilities for the dominant channel $3d\pi^2-2p\pi\varepsilon$ at different extents of the system ionization m and measurements at different collision energies.

higher agreement is reached by using the Auger transition probabilities for the $Ne^{4+}-Ne$ system. At the energies of 12.5–50 keV, the agreement with the experiment may be reached only in case the quasimolecule ionization degree is raised to $m = 6$. In this case, quantity m means the total number of electrons removed from the system.

Therefore, observation of electron emission in a quasimolecule and comparison of the experimental data with the calculations allowed us to establish that vacancy decay on the $2p\pi$-orbital occurs if the system is highly ionized and excited, and essential enhancement of particle ionization with increasing collision energy takes place.

The main contribution at low collision energies comes from the $3d\pi^2-2p\pi\varepsilon$ transition. When the collision energy increases, contribution of channel $3d\pi 3d\sigma$ gets considerable, and the spectrum shifts to higher energies. However, the channel $3d\pi^2$ contribution dominates at all the considered energies (up to 50 keV). Contribution of this channel exceeds 80%. This enables determination of the probability of the indicated Auger transition directly from the experiment with the aid of the following relation:

$$W(R) = \left(\frac{d\sigma}{dE}\right)\frac{1}{fG}.$$

The accuracy of this estimation is better than 20%; the error connected with taking into account other channels is of the systematic character.

Fig. 3 illustrates the comparison between the calculations and experimental data. As seen from Fig. 3, experimental probabilities of occupying the $2p\pi$ vacancy show that at low collision energies Auger transitions occur in the $Ne^{2+}-Ne$ system, while at high collision energies an essential growth (to $m = 6$) of the particle ionization degree (number of removed electrons) takes place. With increasing ionization of the collision partners, energy positions of excited levels $3p\sigma$, $3d\sigma$ and $3d\pi$ come closer to the $2p\pi$ level. As shown by the calculation, the increase in the wave function overlap integrals connected with this fact results in considerable increase in the transition probabilities.

Quite natural is the conclusion that the system consisting of two colliding atoms at the moment of their closest approach gets in a highly excited (strongly „overheated“) state. What is different about it is that we can obtain relevant information only by analyzing the spectrum of electrons emitted in a short time interval of the quasimolecule existence, since some excited states arising in case of close approach of the colliding atoms at the expense of their kinetic energy can adiabatically decay without leaving any traces when the atoms fly apart. Only if the state decay is accompanied by emission of an electron (or photon) bringing energy away from the system, the collision becomes irreversibly inelastic.

Thus, probabilities of vacancy Auger decay during collision $W(R)$ were calculated for the first time for a many–electron quasimolecule Ne–Ne at different quasimolecule ionization degrees; the calculations were compared with experimental data.

The contribution of the transition from the initial $3d\pi-3d\pi$ state to the $2p\pi$ orbital was shown to be predominant among the whole variety of possible Auger decay channels, which has enabled refining the experimentally estimated probabilities of Auger transitions in the quasimolecule as well as direct comparison of the calculated and measured probabilities of Auger transitions in the quasimolecule.

The established regularity, namely, a great increase in the system ionization degree with increasing collision energy which is equivalent to reduction of the reachable closest approach distances should be taken into account in modeling particles stopping in the matter and in analyzing particles ionization during collisions, Auger electron emission and characteristic irradiation, and other collision processes.

Financial support

The study has been performed in the scope of State Assignment of the RF Ministry of Science and Education for the Federal State Budgetary Scientific Institution Ioffe Institute (RAS).

Conflict of interests

The authors declare that they have no conflict of interests.

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