Observation of new types of Auger transitions in atoms with two internal vacancies

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We have observed and investigated three-electron Auger transitions of the type *LL–MMM*. In such transitions, two internal vacancies in the atom are filled simultaneously by two external electrons, and the entire released energy is carried away by the third external electron.

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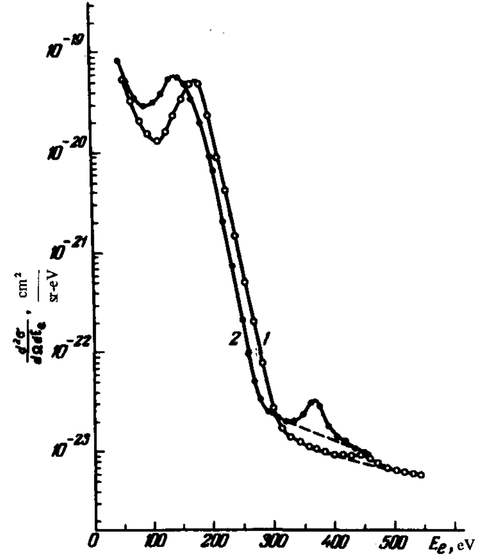
The question of the feasibility of three-electron Auger transitions has already been under discussion for a number of years. In[1] they reported observation of a peak corresponding to an energy $E_e \approx 500$ eV in the spectrum of the electrons produced in Ar^+—Ar collisions. These electrons were ascribed to *LL—MMM* transitions. In subsequent experiments,[2] however, it was shown that the peak observed in[1] is an apparatus effect. It could not be observed even when the sensitivity of the procedure was such that cross sections smaller than those indicated in[1] by three to four orders of magnitude could be measured in the region $E_e \approx 500$ eV.

In the present study we have investigated the energy spectra of electrons produced in collisions of the ions N^+, N_2^+, Ar^+, and Cl^+ with Ar atoms at incident-ion energies $E_0 = 25$ and 50 keV. The measurement procedure is described briefly in[3]. The electron emission angle amounted to 125° relative to the direction of the primary beam. The energy resolution of the spectrum analyzer was $\Delta E_e/E_e = 4\%$, the aperture was $\Delta\Omega = 0.05$ sr. The minimum measured cross section $d^2\sigma/d\Omega\, dE_e$ was $\sim 10^{-25}$ cm²/sr-eV, which is higher by one or two orders of magnitude than the sensitivity used in[2].

Measurements of the inelastic energy losses and the ionization in deep atomic collisions[4] have shown that in the Cl^+—Ar case there are produced, with high probability, two $L_{2,3}$ vacancies in the Cl ions, and in the remaining investigated cases there are produced two $L_{2,3}$ vacancies in Ar. These vacancies are filled, with an



Energy spectra of the released electrons: 1—Ar^+—Ar, 50 keV; 2—Cl^+—Ar, 50 keV.

overwhelming probability, as a result of two ordinary Auger transitions of the type L—MM, with each electron carrying away the transition energy per vacancy.

The figure shows the spectra of the electrons produced in Ar^+—Ar and Cl^+—Ar collisions. Intense peaks of electrons with energies 180 eV in the case of Ar^+—Ar and 140 eV in the case Cl^+—Ar correspond to ordinary L—MM transitions. The peaks at 445 eV for Ar^+—Ar and 370 eV for Cl^+—Ar correspond to three-electron LL—MMM transitions. The error in the relative measurements of the cross sections $d^2\sigma/d\Omega\, dE_2$ was 10—20%. The absolute values of the cross sections were obtained by calibration against the data of[5] for Ar^+—Ar collisions at 50 keV.

The electron energies E_e in three-electron transitions can be calculated for the Ar^+—Ar case by starting from the energies of the different electronic configuartions of the Ar ion, as given in the paper of Larkins.[6] It is known[4] that when two $L_{2,3}$ vacancies are produced in the Ar^+—Ar system, two electrons each are removed on the average simultaneously from the outer shell of each of the particles. Therefore the "average" initial configuration for three-electron Auger transitions is $2p^4 3s^2 3p^4$. The Auger-electron energy calculated for this configuration is 440 eV, in very good agreement with the experimental data. The change of the energy of the Auger electrons in the Cl^+—Ar case agrees fully with the change in the binding energy of the $L_{2,3}$ electrons on going from Ar to Cl.

The table lists, for different investigated cases, the energies E_e and the cross sections $\sigma(L—MM)$ and $\sigma(LL—MMM)$ of ordinary and three-electron transitions, and also the cross sections $\sigma(LL)$ for the production of two $L_{2,3}$ vacancies in one particle.

The cross sections $\sigma(LL)$ were determined on the basis of the preceding investigations of scattering and inelastic energy losses.[4] In[4] we investigated, for the cases N^+—Ar and Cl^+—Ar, the probability W, of production of two $L_{2,3}$ vacancies as a function of the internuclear distance r_0. Using the data on $W(r_0)$, we can find the dependence of W on the impact parameter p at each energy E_0, and determine the cross sections $\sigma(LL) = 2\pi \int_0^\infty pW(p)\, dp$. In the Ar^+—Ar case the situation is somewhat more complicated, since the two $L_{2,3}$ vacancies can be distributed among the collision partners or turn out to be in one of the particles. To determine $\sigma(LL)$, we used estimates of the probability of production of two L vacancies in one ion, obtained in[4,7] by analysis of the data on the probability distribution of the final charge state of the colliding particles.

It is seen from the table that the ratios $\sigma(LL—MMM)/\sigma(L—MM)$ differ in the Ar^+—Ar case from the remaining cases by almost one order of magnitude. It is obvious that this is connected with the different probability of formation of two vacancies in one particle in collisions of identical and different particles. At the same time, the ratios $\sigma(LL—MM)/\sigma(LL)$, which characterize the relative probability of the three-electron transition, differ little in all the investigated cases. The deviation in the case of Ar^+—Ar can be due entirely to the approximate character of $\sigma(LL)$ for these collisions. The large difference between the ratios $\sigma(LL—MMM)/\sigma(L—MM)$ at constant $\sigma(LL—MMM)/\sigma(LL)$ indicates that the considered peaks are indeed connected with formation of two frequencies in one particle, and are not apparatus satellites of the intense L—MM peak.

The probabilities of the three-electron transitions should not depend significantly on the collision velocity, if the transitions occur in isolated atoms. The dependence of the probability on the initial energy E_0 was investigated in N^+ and N_2^+—Ar collisions. The molecular structure of the N_2^+ ion does not influence the processes in the internal shells, and therefore the N_2^+ ion can be regarded as two independent atomic particles. As seen from the table, the ratio $\sigma(LL—MMM)/\sigma(LL)$ does indeed

Pair	E_0	$L_{2,3}$-MM		$L_{2,3}L_{2,3}$-MMM		$\sigma(LL\text{-}MMM)/\sigma(L\text{-}MM)$	$\sigma(LL)$	$\sigma(LL\text{-}MMM)/\sigma(LL)$
	keV	E_e eV	$\sigma, 10^{-18}$ cm²	E_e eV	$\sigma, 10^{-22}$ cm²	10^{-5}	10^{-18} cm²	10^{-4}
Ar^+—Ar	50	175	32.0	445	11.0	3.3 ±0.6	2.8	3.8
Ar^+—Ar	25	180	27.0	445	10.0	3.7 ±0.7	2.7	3.7
Cl^+—Ar	50	140	36.0	370	99.0	27 ± 5	16.0	6.2
N^+—Ar	50	180	7.1	470	16.0	23 ± 5	1.7	9.7
N_2^+—Ar	50	185	5.9	465	9.3	16 ±3	1.3	7.2
N_2^+—Ar	25	185	3.7	470	6.7	18 ±4	0.7	9.6

remain practically constant when E_0 changes by a factor of four, from 50 keV in the N^+—Ar case to 12.5 keV per incident atom in the case of N_2^+—Ar (25 keV).

The observed three-electron transitions are the consequence of multielectron correlations in the atom. The measured ratios $\sigma(LL-MMM)/\sigma(LL)$ lie in the interval $10^{-3}-10^{-4}$. If it is assumed that the lifetime of the state with two L vacancies relative to the ordinary $L-MM$ transition is of the same order of magnitude as in an atom with one L vacancy, i.e., $\sim 10^{-14}-10^{-15}$ sec, then the lifetime relative to the three-electron transition should be $\sim 10^{-10}-10^{-12}$ sec. It is of interest to study the influence of the number of electrons in the outer shell of the atom on the probability of the three-electron transition. One can expect the probability of the three-electron transition $\sigma(LL-MMM)/\sigma(LL)$ to decrease with decreasing number of outer electrons, owing to the decrease in the role of the correlation effect. Naturally, in the presence of only two internal electrons this transition is not possible at all.

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