Shake-off and direct collisions in atoms with photoionization, β^--decay and electron-impact ionization

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High resolution primary and secondary K_α x-ray spectra of Si are investigated with the purpose of elucidating the mechanisms of shake-off and direct collisions in KL-ionization of the atoms. Slowing down of the electrons in the thick target is taken into account by introducing an effective electron energy. Control measurements are carried out for a number of elements in thick targets. The incident-electron energy dependence of the probability for L-shell ionization involving the formation of vacancies in the K-shell differs significantly from the dependence for K-shell ionization during β^--decay. It is shown that the parameter which determines the probability for two-electron KL-ionization is the energy transmitted to the atom by the incident particle. For moderate energies the main role in all cases of ionization is played by the shake-off mechanism.

The absorption of a photon or by electron impact can result in multi-electron ionization of an atom. If the electrons, ejected from the atom, belong to different shells, then for the description of the process of multi-electron ionization the models of shake-off (SO) and direct collisions (DC),[1–8] first introduced by Migdal and Feinberg for the calculation of the ionization probability of the atom by α and β-decay,[9–11] are usually used.

The processes of ionization of the atom by β^--decay and by two-electron ionization have much in common. Thus, for the electrons, for example of the L-shell, both in β^--decay, and in ionization of the K-shell, the self-consistent field changes (SO ionization mechanism), and either a β^--particle or an electron is scattered, leaving the K-shell (DC ionization mechanism).

An estimate of the contributions of the SO and DC mechanisms in the ionization process of the K-shell of an atom during β^--decay, carried out for various elements, yields results differing by several orders of magnitude.[12–16] This is connected with the essentially different limiting energies of the β^--particles. The role of the SO and DC mechanisms in two-electron KL-ionization process is not clear. Thus, in Ref. 5 the probability P_L for vacancy formation in the L-shell was calculated for the inert gases without considering DC. At the same time, the experimental investigation of the probability P_L for KL–ionization was basically conducted for electron excitation, assuming[7,8] that the direct collision mechanism plays a vital role.

In comparison with the analysis of the L-shell ionization by β^--decay, the study of the two-electron KL-ionization of an atom has the advantage that for a single element the energy E_0 of the incoming particles can be varied. In this connection it appears possible in principle to estimate the contribution of P_L (SO) and P_L (DC) to the probability P_L for various energies E_0, since these partial probabilities depend differently on E_0. The present work is aimed at carrying out this idea.

EXPERIMENTAL RESULTS

The probability P_L of two-electron KL-ionization of an atom relative to one-electron K-ionization may be determined experimentally[4] by measuring the relative intensity of the $K_{\alpha 3,4}$ x-ray lines: $\varkappa = I(\alpha_{3,4})/I(\alpha_{1,2})$, since they are emitted by the radioactive decay of atomic states with vacancies in the K- and L-shells, while the $K_{\alpha 3,4}$ lines are associated with a vacancy in the K-shell.

The x-ray fluorescent K_α - spectra of (Si) were obtained with a DRS-2 spectrograph in monochromatic and polychromatic excitation. The method of obtaining and processing the experimental data is analogous to that described earlier.[17] The object of the analysis (Si) was chosen with a view to the necessity for obtaining spectra of the highest resolution, as large a range of the ratio $\hbar\omega/E_{KL}$ as possible (where $\hbar\omega$ is the energy of the excited photon and E_{KL}-KL is the ionization threshold), and good $K_{\alpha 3,4}$ line intensity.

The conditions under which the experimental points shown in Fig. 1, (open circles) were obtained are as follows: With a ZR anode, the voltage in the tube was 16 kV; with N6, 17 kV; with Mo, 17 kV; with Pd, 23 kV with Ti, 23 kV, with Cr, 23 kV; with Cu, 23 kV; with Cu, 23 kV and monochromatic $K_{\alpha 1,2}$ radiation; with Pd, 35 kV and mochromatic $K_{\alpha 1,2}$ radiation. The corresponding points in Fig. 1 are distributed from left to right.

The primary x-ray spectra of Si was found using the rebuilt EMMA-2 device.[8] The acceleration voltage varied over the range 3–100 kV. The slowing-down of the electrons in a thick target was taken into account by introducing an effective electron energy $E_{\rm eff}$ (see the Appendix). Corrections were made to the experimental values of $\varkappa$, obtained as the ratio of the areas under the contours of the lines $K_{\alpha 1,2}$ and $K_{\alpha 3,4}$. By this means it was determined that the excitation of the two- and one-vacancy state of an atom was brought about by electrons with various effective energies. The corrected values of $\varkappa$ are shown in Fig. 1 (filled circles).

In Fig. 2 experimental values are shown (in units of $\varkappa_{\max}$), derived from both thick and thin Ti and Cr targets. The merging of the experimental points to form a smooth curve justifies introducting $E_{\rm eff}$ in order to calculate the influence of the electron slowing-down and the corrections in the experimental results obtained from thick targets.

The values of P_L for Si, shown in Fig. 1, are derived from the formula[4]

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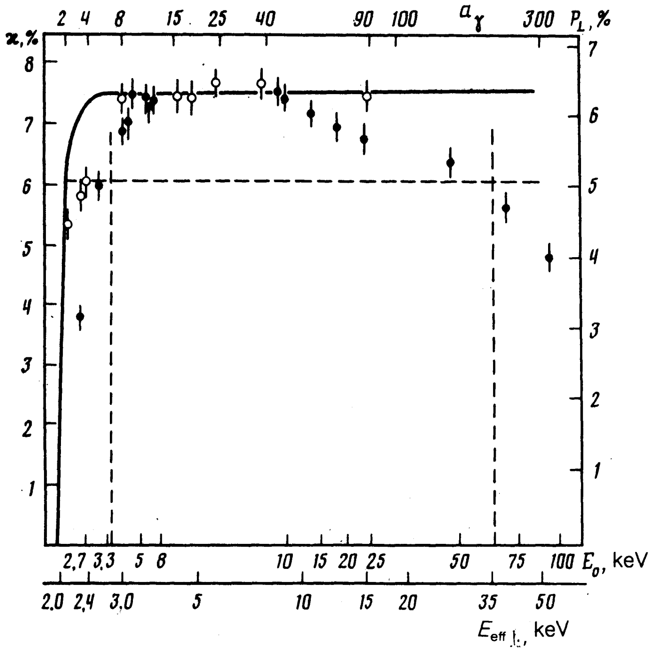

FIG. 1. Relative intensity of the $K_{\alpha 3,4}$ satellites of Si versus the energy of the incident electrons (●) and photons (○). The solid curve represents calculation in Ref. 19. See the text concerning E_{eff}, P_L, and a_γ, and the broken lines.

$$P_L = {}^4/_5\varkappa(1+B), \qquad (1)$$

where $B = 0.045$ is the ratio of the natural widths of the L_3^- and K-levels, the values of which are obtained from Ref. 18.

DISCUSSION

In analogy with the ionization of an atom by β^--decay, the application of the SO approximation to describe the process of two-electron KL-ionization in photoabsorption and electron impact is based on the possibility of dividing the ionization process into two stages: fast (ejection of a K-electron) and slow (shake-off of the L-electron)[1,2] In β^--decay the ratio P_L(DC)/P_L(SO), like P_L, is determined by the energy of the β^--particle, in photoionization and electron-impact ionization, by the energy E of the electron exiting from the K-shell.

It is important that P_L(SO) and P_L(DC) depend in different ways on E. In accordance with the calculation for the two-electron KL-photoionization,[19] the probability P_L(SO) abruptly increases in the threshold region and subsequently remains constant as a function of E (Fig. 1, solid line). At the same time P_L(DC), like the dependence of the ionization cross section of the inner shell of the atom on the energy of the incident electron, is a curve with a maximum at $E \approx 3E_L$.[20] It thus follows, that it makes sense to speak of an essential role of direct encounters in the process of two-electron KL-ionization for large E, only when the probability P_L is observed experimentally to decrease as E increases.

The photoionization data are the easiest to interpret. This is a consequence of the fact that in the absorption of monochromatic radiation the energy $E \approx \hbar\omega - E_K$ and the parameter $a_\gamma = E/E_L$, determining P_L and P_L(DC)/P_L(SO), are exactly known and are identical for each ionization event. As follows from Fig. 1, near the threshold, the probability P_L quickly increases as a function of energy of the absorbed photons and beginning even at $\hbar\omega \approx 1.5E_{KL}$, which corresponds to $a_\gamma = 8$, becomes constant. The result agrees qualitatively with a calculation carried out in the SO approximation.[19] Similar results are also obtained for Ti and Cr (see Fig. 2), but for a narrower interval of a_γ and with less reliable values of the relative intensity ϰ.

Since in the region $a_\gamma > 8$ (see Fig. 1) the probability P_L does not decrease as a function of E, it is possible to conclude that P_L(SO) $\gg P_L$(DC). More precisely, the experiment shows that the ratio P_L(DC)/P_L does not exceed the relative error of the experiment (10%) in the determination of P_L. Consequently, P_L(DC)/P_L for $a_\gamma = 8$ (the basis of the region $P_L \approx$ const) is at most 0.1, and must still be even smaller for $a_\gamma > 8$. In the region $4 < a_\gamma < 8$ the change of P_L is produced mainly by the SO mechanism, since as a_γ increases the probability P_L(SO) also increases, while P_L(DC), on the contrary, decreases. Note, that for $a_\gamma < 4$ estimating the ratio P_L(DC)/P_L(SO) from the experimental dependence of $P_L(a_\gamma)$ becomes more difficult, since as a_γ increases both partial probabilities increase. In addition, it should be remembered that near the threshold, the SO and DC mechanisms are not independent, i.e., the interference term may be comparable with P_L(SO) and P_L(DC), as in the case of the ionization associated with β^--decay.[12-14]

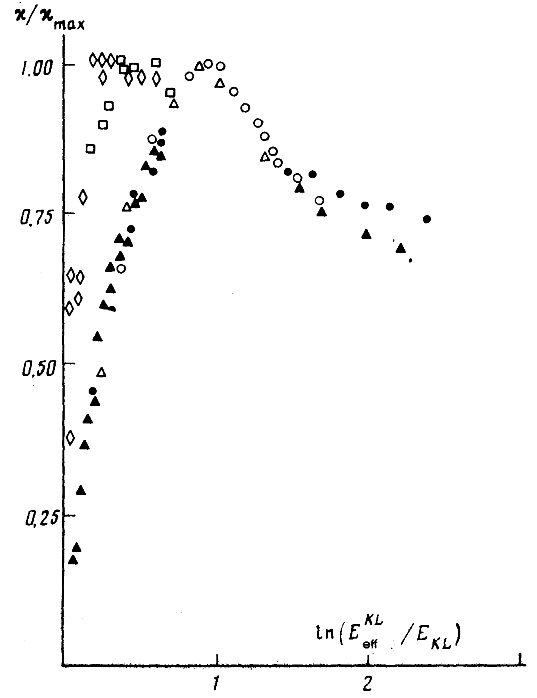

FIG. 2. Relative intensity of the $K_{\alpha 3,4}$ satellites of Ti and Cr versus the energy of the incident electrons (Ti: ○—thin target; ●—thick target; Cr: △—thin target, ▲—thick target) and photons (Ti—□, Cr—◇).

Since there is a profound analogy among the processes of two-electron KL-photoionization and ionization in β^--decay, it is reasonable to expect that the dependence of the probability for ionization of the inner shell on the energy of the β^--particles is similiar to $P_L(a_\gamma)$. However, in attempting to confirm this assumption we encounter two difficulties. First of all, for each isotope the measured values of P_K and P_L correspond, as a rule, to a single β^--particle limiting energy. In the second place, the β^--particles energies have a probability distribution.

To overcome the first of these problems we noted that in the case $E_\beta \to \infty$, the probabilities P_K (DC) and P_L (DC) go to zero, and P_K (SO) and P_L (SO) are well calculated and may serve as scale values. As far as the probability distribution of the β^--particle energy is concerned, it may be disregarded, since the error of the results increases greatly only in the range of small energies. As a parameter, determining the ionization probability of the inner shell we can use the quantity a_β (the ratio of the limiting energy of the β^--particle to the binding energy of the electron of the corresponding shell).[9–14] Thus, it is possible to assume, that the normalized experimental results of the ionization probability of the inner shell by β^--decay for various isotopes are described by some universal curve.

Experimental and theoretical values of the probability P_K for β^--decay ionization of the K-shell, which are known for many isotopes,[12–16,21] are conveniently represented in the form of the dependence $P_K^{\rm exp}/P_K^{\rm theor} = f(a_\beta)$. In the present case the parameter $a_\beta = E_\beta/E_K$ determines both P_K and $P_K({\rm DC})/P_K({\rm SO})$. For each isotope, $P_K^{\rm exp}$ corresponds to some E_β, while the probability $P_K^{\rm theor}$, calculated in the sudden approximation,[22] corresponds to $E_\beta \to \infty$. Thus, Fig. 3 actually shows the dependence of $P_K(a_\beta)$ for some imaginary isotopes with varying β^--particle energy limits. It is clear that $P_K^{\rm exp}/P_K^{\rm theor}$ increases in the interval $1 < a_\beta < 20$ as a function of a_β, but subsequently remains constant right up to $a_\beta \approx 800$.

The experimental data for L-shell ionization by β^--decay,[23] which are in fact relatively sparse, give rise to the same dependence of $P_L^{\rm exp}/P_L^{\rm theor}$ on the parameter $a_\beta = E_\beta/E_L$. Based on this it may be deduced that the ionization of the L-shell, and also the K-shell by β^--decay in the region $a_\beta > 20$ is essentially accounted for by the SO mechanism.

Note, that in linking a_β with the energy limit of the β^--particle, we raise the value of the parameter, determining the complete and partial probabilities for the ionization of an atom. This evidently explains the slower increase of P_K and

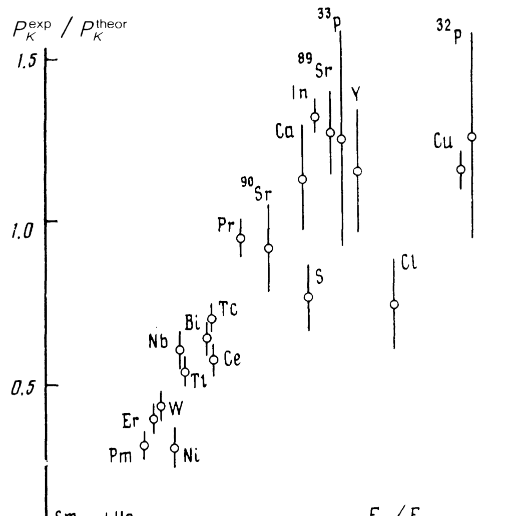

FIG. 3. Probability of ionization of the K-shell versus E_β, the extreme energy of the β^--particles. The experimental values P_K are normalized with respect to the theoretical values[22], E_β is normalized with respect to electron binding energy of the K-shell of the element $Z + 1$.

P_L with a_β (see Fig. 3) in comparison with the increase of P_L as a function of a_γ (see Fig. 1).

The most difficult data from the point of view of interpretation are those obtained by electron impact. In this case, in contrast to β^--decay ionization and two-electron photoionization, the final state has an ion and three, not two electrons, with a continuous energy spectrum. The energy of one of them (ejected from the L-shell) is close to zero,[19] the distribution of the energy between the scattered electron and that knocked out from the K-shell is unknown. Consequently, the energy E_1 transmitted to the atom by electron impact is also unknown.

The decrease of $\varkappa$ (and P_L), observed experimentally as the incoming electron energy E_0 increases in the range $E_0 > 3E_{KL}$ (see Figs. 1 and 2), is commonly connected with the contribution of the DC mechanism. The expression for the ratio of the cross sections two- and one-electron ionization by electron impact serves as the basis for this[3]:

$$\sigma_{KL}/\sigma_K \approx a + b(\ln E_0)^{-1}. \qquad (2)$$

Here a = const is identified with the contribution of SO, and $b(\ln E_0)^{-1}$ with that of DC. In recent work,[24] the decrease in the relative intensity of the hypersatellite $I(K_{\alpha_2}^H)/I(K_{\alpha,1,3})$ with increasing E_0 was accounted for in a similar manner.

However, in the two-level approximation to the KL-ionization, in which the SO and DC channels are independent, the terms in the binomial (2) signify P_L (SO) and P_L (DC) only, if in place of E_0, E(the average energy of the electron leaving the K-shell) is used. Therefore, the interpretation of the decrease of P_L as a function of E_0 as an occurrence of the mechansim of DC[3,7,8] is essentially based on two assumptions: a) E also increases with E_0; b) the contribution of P_L (DC) to P_L is so large, that it gives rise to the maximum in the experimental curves of $P_L(E_0)$.

If we now take into account, that when atoms interact with fast electrons the problem of the double KL-ionization by electron impact becomes analogous to the problem of photoionization, then it appears that assumptions a) and b) does not agree with our experimental data for photoionization (see Fig. 1). Actually, although as the frequency of the absorbed radiation increases as a function of the energy of the electron exiting from the K-shell, but no decrease in P_L is observed, in contrast to KL-ionization by electron impact. All this permits us to conclude that the interpretation of the decrease in P_L in the range $E_0 > 3E_{KL}$ as a decrease in P_L (DC) with P_L (SO) = const (Refs. 3,7,8) appears doubtful.

We note that the fast incident electron does not directly influence the processes occuring in the atoms after the formation of the primary vacancy, and no additional ionizations takes place, since in the opposite case, the values of $\varkappa_{\max}$, obtained from photoionization and ionization by electron impact would not be identical. Consequently, the only parameter, affecting P_L/P_L (SO) and P_L (DC) is found to be the energy E transferred to the atom by the interaction of these particles.

Our interpretation of the results, obtained from electron impact, shown in Figs. 1 and 2, is based on the following assumption: If the same energy E_1 is transferred to the atom by photoionization and electron-impact ionization then the probability of ionization of the L-shell with the formation of a vacancy in the K-shell is also identical, i.e.,

$$P^{el}(E_1) = P^{phot}(E_1). \quad (3)$$

Evidently when condition (3) is satisfied, the same relation holds as well for $P_L(\mathrm{DC})/P_L(\mathrm{SO})$.

Since in photoionization close to the threshold the probability P_L decreases only when E_1 decreases, then in our picture, the decrease in P_L, observed in electron impact in the range $E_0 > 3E_{KL}$ is linked with the fact, that an increase in the energy of the incident electron causes the energy transmitted to the atom by the impact to decrease on the average. In other words, with an increase in E_0 in the region $E_0 > 3K_{KL}$ the maximum of the electron distribution as a function of the energy transfer shifts toward lower energy. This conclusion, which implies that in real experiments with $E_0 \gg E_{KL}$ the conditions of a sudden perturbation are not realized, might be verified by comparison with $(e^-, 2e^-)$-experiments.

For convenience in estimating $P_L(\mathrm{DC})/P_L(\mathrm{SO})$ for electron impact we introduce by analogy with a_γ and a_β, the parameter $a_1 = (E_1 - E_K)/E_2$. Then the conditions in (3) are written as

$$P_L(a_l) = P_L(a_\gamma). \quad (3')$$

Condition (3') permits us to define a, for each value of energy of the incident electron, using the experimental dependence $P_L(a_\gamma)$, where to each value of P_L corresponds an exact value of a_γ. As a consequence of this the change in P_L in the range $E_0 > 3E_{KL}$ associated with electron impact and close to the threshold for photoionization are interpreted similarly.

The experimental values of P_L, displayed in Fig. 1 above the horizontal dashed line correspond to the case $a_l > 4$ and the vertical dashed line indicates the corresponding range of energy of the incident electron. It is possible to conclude, that the decrease in P_L as a function of E_0 in the range $12 \lesssim E_0 \lesssim 60$ keV, as in the region $4 < a_\gamma < 8$ for photoionization, is mainly due to the SO process. At the same time, for $E_0 \gg 60$ keV it is difficult to distinguish the contributions of $P_L(\mathrm{SO}) < P_L(\mathrm{DC})$ to P_L for the same reasons as in the case $a_\gamma < 4$ for photoionization.

Thus, the experimental results, obtained by photoabsorption, β^--decay, and by electron impact, allow us to conclude that the basic mechanism for ionization of the atom in all cases appears to be SO, provided only that the energy transferred to the atom by the collision is not too small.

APPENDIX

Disregarding the statistical of the electron energy losses due to nonelastic collisions with atomic targets we can express the intensity of a given line of the K-series of the primary x-ray spectrum obtained in a thick anode of element z in the following form[25]:

$$I = \mathrm{const} \int_{E_K}^{E_0} \frac{Q_k}{S} dE, \quad (A1)$$

where

$$S = -\frac{1}{\rho}\frac{dE}{dx} = \mathrm{const}\frac{1}{E}\ln\frac{1.166E}{J}$$

is the specific slowing-down effectiveness,

$$Q_K = \mathrm{const}\frac{1}{E}\ln\frac{E}{E_K}$$

is the cross section for K-shell ionization, and J is the average ionization potential.

We introduce $Q_K^{\mathrm{eff}} = \mathrm{const}\, E_{\mathrm{eff}}^{-1} \ln(E_{\mathrm{eff}}/E_K)$ in order to satisfy the condition

$$\int_{E_K}^{E_0} \frac{Q}{S} dE = Q_K^{\mathrm{eff}} \int_{E_K}^{E_0} \frac{dE}{S}. \quad (A2)$$

The substitution Q_K, Q_K^{eff} and S in $(P,2)$ yields

$$E_{\mathrm{eff}} = C \ln(E_{\mathrm{eff}}/E_K), \quad (A3)$$

where

$$C = \int_{E_K}^{E_0} \frac{E\, dE}{\ln(1.166E/J)} \Big/ \int_{E_K}^{E_0} \frac{\ln(E/E_K)\, dE}{\ln(1.166E/J)}. \quad (A4)$$

Equations (A3) and (A4) were solved numerically; the functions $E_{\mathrm{eff}}^{K}(E_0, E_K = \mathrm{const})$ and $E_{\mathrm{eff}}^{KL}(E_0, E_{KL} = \mathrm{const})$ were calculated for Si, Ti, Cr and other elements in the energy interval $E_K < E_0 \lesssim 100$ keV.

Thus, the introduction of E_{eff} accounts for the influence of electron deceleration in a thick target on the relative intensity of the lines of the x-ray spectra. Obviously, for a thin target (thickness on the order of the electron mean free path) $E_{\mathrm{eff}} \approx E_0$.

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