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Theory of the Radiative Auger Effect*

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The probability of simultaneous emission of an x-ray photon and an electron is considered. It is shown that this process can occur as a consequence of the change in the average potential acting on the electrons when the x-ray hole is moving from an inner to an outer shell (shake-off) and as a consequence of interaction between single- and double-hole configurations in the final state. Relative shake-off probabilities of $K \rightarrow L^2$, $K \rightarrow M^2$, and $L_{2,3} \rightarrow M_1 M_{2,3}$ transitions are calculated for F^-, Ne, Na^+, Cl^-, Ar, and K^+ from Bagus's self-consistent-field wave functions of single-hole x-ray states. The relative probability of $K \rightarrow L^2$ and $K \rightarrow M^2$ transitions is estimated for the rest of the atoms in the range $9 \leq Z \leq 22$. The estimated $K \rightarrow L^2$ and $K \rightarrow M^2$ probabilities are of the same order of magnitude as the observed probabilities. However, for low Z, they overestimate the observed probabilities, which may be because the shake-off calculations do not account properly for the interaction between single- and double-hole configurations in the final state.

I. INTRODUCTION

Recent observations[1-5] of low-energy structures accompanying ordinary x-ray lines have renewed the interest in the radiative Auger effect.[6-8] In this effect an inner-hole state decays by the simultaneous emission of a photon and an electron. The electron may also be excited into a bound state during the x-ray emission process. So far the analysis of the low-energy spectra has been based on energy considerations and no attempt has been made to calculate the probability of the radiative Auger effect. Bloch[7] gave only a very rough estimate of this probability in a special case where radiative Auger transitions replace a dipole forbidden x-ray transition. However, the experiments indicate that the radiative Auger effect occurs in connection with allowed x-ray transitions as well.[1-4]

In this paper we consider probabilities of double-electron transitions of radiative Auger type and show that they should be observable in a variety of cases. Since the photon-electron interaction is described by a one-electron operator, the ordinary one-electron model where the electrons are assumed to move in a fixed central-field potential does not account for the radiative Auger effect. The first step to improve the fixed-potential model is to solve separate self-consistent-field (SCF) equations for the initial and final states.[9-13] This procedure describes the core rearrangement (shake-off) during the x-ray emission process and leads to a finite probability of the radiative Auger effect, as we shall show in Sec. IIA. However, shake-off is not the only mechanism which contributes to the radiative Auger effect in the first order. In Sec. IIB, we notice that configuration interaction (CI) between final single- and double-hole states is also a first-order effect. In Sec. IIIA, we estimate shake-off probabilities of transitions where a K hole is filled by an outer-shell electron and a photon, and another electron is emitted. Results are presented for atoms and ions in the range $9 \leq Z \leq 22$. In Sec. IIIB, we make a comparison with available experimental data and discuss $L \rightarrow M^2$ transitions briefly. In Sec. IV, we discuss the significance of shake-off calculations with regard to configuration interaction in the final state and relate our treatment to the theory[14] of electron-interaction effects on the emission of soft x rays from metals.

II. PROBABILITY OF DOUBLE-ELECTRON TRANSITIONS

We consider a dipole transition between two levels i and f in an atom. The transition probability A_{if} of the spontaneous emission of a photon is given in a.u. by

$$A_{if} = \tfrac{4}{3}\,\alpha^3 g_i^{-1} E_{if}^3 S_{if} , \tag{1}$$

where α is the fine-structure constant, g_i the degeneracy of the initial level, and $E_{if} = E_i - E_f$ the energy difference equal to the photon energy $\hbar\omega$.[15] The line strength S_{if} is given by

$$S_{if} = \sum_{a,b} \left| \langle b | \sum_{j=1}^{N} \vec{r}_j \, | a \rangle \right|^2 , \tag{2}$$

where the summation is carried out over the degenerate initial and final states a and b. The following treatment of the photon emission in the radiative Auger effect is based on Eqs. (1) and (2).

A radiative Auger transition is a double-electron transition where an outer-shell electron nl jumps into an inner-shell hole $n_0 l_0$ and where an another outer-shell electron $n'l'$ is simultaneously excited into a bound or continuum state $\epsilon\bar{l}$. Consequently these transitions correspond to a discrete and con-

tinuous distribution of photon energies $\hbar\omega$ given by

$$\hbar\omega = I_{n_0 l_0} - I_{nl,n'l'} - \epsilon \ . \tag{3}$$

Here $I_{n_0 l_0}$ is the $n_0 l_0$ ionization energy of the neutral atom and $I_{nl,n'l'}$ the energy of the *simultaneous* ionization of two electrons nl and $n'l'$ from the neutral atom. The variation of ϵ from zero to $I_{n_0 l_0} - I_{nl,n'l'}$ gives rise to a continuous distribution of photon energies. The excitation of the $n'l'$ electron into various bound states corresponds to discrete negative values of ϵ. The transitions $n_0 l_0 \to nl$ and $n_0 l_0 \to n'l'$ may both be allowed by dipole selection rules, one may be forbidden, or both may be forbidden. In the following we assume that at least the transition $n_0 l_0 \to nl$ is allowed. A case where both transitions are forbidden is briefly mentioned in Sec. III B.

A. Shake-Off

Suppose that separate SCF equations have been solved for the initial $(n_0 l_0)^{-1}$ single-hole state and for the final $[(nl)^{-1}(n'l')^{-1}]\epsilon\bar{l}$ double-hole states. Consequently, the overlap elements $\langle n_2 l_2 | n_1 l_1 \rangle$, where $n_1 l_1$ is an initial-state and $n_2 l_2$ a final-state orbital, are not necessarily equal to $\delta_{n_2 n_1}$ even if $l_2 = l_1$. However, the total initial- and final-state wave functions are still orthogonal since they must have opposite parity in a dipole transition.

The line strength (2) can be calculated using formulas given by Löwdin[16] for transition matrix elements of Slater determinants which are built up from nonorthogonal basic sets. If we consider only terms to the lowest nonvanishing order in the overlap elements $\langle n_2 l_2 | n_1 l_1 \rangle$ and put them equal to 1 whenever $n_2 = n_1$, we get

$$S \propto [D_0(\epsilon\bar{l}, n'l') D_1(n_0 l_0, nl)]^2 \delta_{\bar{l}l'} \ , \tag{4}$$

where

$$D_j(n_2 l_2, n_1 l_1) = \int R_f(n_2 l_2) R_i(n_1 l_1) r^{j+2} dr \ . \tag{5}$$

Here R_i and R_f are the initial- and final-state radial wave functions that represent the participating electrons. According to Eq. (4), the double-electron transition $(n_0 l_0)^{-1} \to [(nl)^{-1}(n'l')^{-1}]\epsilon\bar{l}$ can be interpreted as a monopole-dipole transition where the excitation is due to a screening effect corresponding to zero angular momentum transfer $\bar{l} = l'$.

B. Configuration Interaction in Final State

Double-electron transitions are also possible if there is an interaction between the single-hole configuration $(nl)^{-1}$ and the double-hole configuration $[(nl)^{-1}(n'l')^{-1}]\epsilon\bar{l}$ in the final state. Transitions to double-hole states would then get strength from the ordinary x-ray transition $(n_0 l_0)^{-1} \to (nl)^{-1}$ through the small mixing coefficients U_{1j} of the single-hole wave function φ_1 in the CI expansion

$$\Psi_j = \varphi_1 U_{1j} + \sum_{i \geq 2} \varphi_i U_{ij} \ , \tag{6}$$

where φ_i for $i \geq 2$ correspond to double-hole states and where U_{jj} is close to 1 ($\Psi_j \cong \varphi_j$).[17] If we assume LS coupling, the wave functions in Eq. (6) are characterized by the same set of quantum numbers $(LSM_L M_S, \pi)$. The φ_i's for $i \geq 2$ are characterized by ϵ and may correspond to several $\bar{l}$ values consistent with the parity π. The coefficients U_{ij} are independent of M_L and M_S.

If the wave functions φ_i and the initial-state wave function are built up from separate SCF orbitals, both shake-off and CI contribute to the double-excitation probability. If we retain terms to the lowest nonvanishing order in both overlap elements $\langle n_2 l_2 | n_1 l_1 \rangle$ and mixing coefficients U_{ij}, we get instead of Eq. (4)

$$S_j \propto D_1(n_0 l_0, nl)^2 [N D_0(\epsilon l', n'l') + U_{1j}]^2 \tag{7}$$

for $j \neq 1$ by putting $U_{jj} = 1$ and $\langle n_2 l_1 | n_1 l_1 \rangle = 1$ for $n_2 = n_1$. Since φ_1 and φ_i ($i \geq 2$) describe different open-shell configurations, $(nl)^{-1}$ and $[(nl)^{-1}, (n'l')^{-1}]\epsilon\bar{l}$, the factors appearing in front of D_1 are generally different for the transition amplitudes that correspond to $i = 1$ and $i \cong 2$. Consequently, the factor N in Eq. (7) is usually not equal to 1.

Although Eq. (7) represents a very crude approximation, it shows that both shake-off and CI in the final state give first-order contributions to the radiative Auger probability. We note also that the corresponding transition amplitudes may either add or subtract. In Secs. III and V we present evidence that the observed double-electron x-ray transitions[1-5] can be interpreted as shake-off and final-state configuration interaction transitions.

III. RELATIVE $K \to L^2$, $K \to M^2$, AND $L \to M^2$ INTENSITIES

A. $K \to L^2$ and $K \to M^2$ Shake-Off Probabilities

In this section we consider light atoms and ions ($9 \leq Z \leq 22$) and assume LS coupling. We estimate the shake-off probability by subtracting from 1 the probability that an electron will stay in its orbit during the x-ray emission. However, we treat the $(1s)^{-1}\,{}^2S \to (2p)^{-2}\epsilon p\,{}^2P^0$ and $(2s)^{-1}(2p)^{-1}\epsilon s\,{}^2P^0$ transitions in neonlike ions in detail in order to show what assumptions our shake-off calculations are based on.

The (p^4) core has three terms ${}^3P$, ${}^1D$, and ${}^1S$. For each term we can construct the properly symmetrized wave functions from the orbitals $(nlm_l m_s)$. The ϵp orbitals can then be coupled to each wave function of the core, which gives rise to six ${}^2P$ wave functions corresponding to $M_L = 0, \pm 1$, and $M_S = \pm\frac{1}{2}$. These wave functions are linear combinations of several Slater determinants whereas the $(1s)^{-1}\,{}^2S$ wave function is a single determinant. The probability of a $(1s)^{-1}\,{}^2S \to (2p)^{-2}\epsilon p\,{}^2P^0$ transition

can then be calculated from Eqs. (1) and (2) where the summation is carried out over M_L and M_S. The result is

$$A_{L'S'}(\epsilon p)=\tfrac{4}{9}\alpha^3(2L'+1)(2S'+1)$$
$$\times E^3_{L'S'}\begin{vmatrix} D_0(1s,1s) & D_0(1s,2s) \\ D_0(2s,1s) & D_0(2s,2s)\end{vmatrix}^2$$
$$\times D_0(2p,2p)^8 D_0(\epsilon p,2p)^2[D_0(2s,2s)D_1(1s,2p)$$
$$-D_0(1s,2s)D_1(2s,2p)]^2 . \quad (8)$$

The $D_j(n_2l_2,n_1l_1)$ elements are defined by Eq. (5). The approximation (4) follows from Eq. (8) if we put $D_0(nl,nl)=1$ and $D_0(1s,2s)=0$. The quantum numbers $L'S'$ refer to the *core* terms. If we assume that the orbitals of each core term are the same, then the intensity ratio $^3P:{}^1D:{}^1S$ is given by the statistical weight $9:5:1$. The energy $E_{L'S'}$ is given by

$$E_{L'S'}=I_{1s}-I_{2p^2}(L'S')-\epsilon , \quad (9)$$

where $I_{1s}-I_{2p^2}(L'S')$ is the $KL_{2,3}L_{2,3}(^{2S'+1}L')$ Auger electron energy. The $L'S'$ splitting of the core varies from 5 to 20 eV in the range $9\le Z\le 22$.[18]

The probability of a $(1s)^{-1}{}^2S\rightarrow[(2s)^{-1}(2p)^{-1};\ ^3P,\ ^1P]\epsilon s\ ^2P^0$ transition can be calculated in the same way as the probability (8). If $D_0(1s,2s)\cong 0$, then the ratio of the ^3P and ^1P transition probabilities is $3:1$. The term splitting of the $2s2p^5$ core varies from 10 to 30 eV in the range $9\le Z\le 22$.[18]

The probability $A(K\rightarrow L^2)$ of x-ray emission and simultaneous excitation to any of the excited $\epsilon p(s)$ states is given by

$$A(K\rightarrow L^2)=\sum_{\epsilon}\left(\sum_{L'S'}A_{L'S'}(\epsilon p)+\sum_{L''S''}A_{L''S''}(\epsilon s)\right) . \quad (10)$$

If we assume that the $2p(2s)$ orbital of the $2p^5$ core and the excited $\epsilon p(\epsilon s)$ orbitals move in the same SCF potential, the probability $A(K\rightarrow L^2)$ can be estimated by using the closure relation. Dividing $A(K\rightarrow L^2)$ by the transition probability of the normal $(1s)^{-1}\rightarrow(2p)^{-1}$ x-ray line yields the relative $K\rightarrow L^2$ probability

$$P(K\rightarrow L^2)=C\left(\frac{5[1-D_0(2p,2p)^2]}{D_0(2p,2p)^2}+\frac{2[1-D_0(2s,2s)^2]}{D_0(2s,2s)^2}\right) , \quad (11)$$

where C is the average of the ratio $E^3_{K\rightarrow L2}/E^3_{K\rightarrow L}$. In Eq. (11), $D_0(2l,2l)^2$ is the square of the overlap element with respect to the $2l$ orbitals of the K and $L_{2,3}$ states and represents the probability that a $2l$ electron stays in its orbit during the $(1s)^{-1}\rightarrow(2p)^{-1}$ x-ray emission process. Consequently $P(K\rightarrow L^2)$ is the L-shell shake-off probability of $K\alpha$ emission in neonlike ions.

TABLE I. Relative shake-off probabilities of $K\rightarrow L^2$ and $K\rightarrow M^2$ transitions in percentages. In column A, $C=1$ in Eq. (11) and there is no exchange correction. In column B, C has been estimated[a] and the exchange correction has been included.

Atom or ion	$P(K\rightarrow L^2)$ A	$P(K\rightarrow L^2)$ B	$P(K\rightarrow M^2)$ A	$P(K\rightarrow M^2)$ B
F^-	6.3	5.7		
Ne	4.4	4.0		
Na^+	3.3	3.0		
Cl^-	2.1	0.60	10.3	9.2
Ar	1.9	0.51	8.3	7.3
K^+	1.7	0.41	6.9	5.9

[a]For $K\rightarrow L^2$ transitions $C\cong 0.9$ ($Z=9$, 10, 11) and 0.75 ($Z=17$, 18, 19). For $K\rightarrow M^2$ transitions $C\cong 1$.

The exchange overlap element $D_0(1s,2s)$ has been neglected in the s part of the probability (11). However, if there are electrons outside the L shell, the contribution from the overlap elements $D_0(n_2l,n_1l)$, where $n_2\neq n_1$, may be important. In the case where there is a filled M shell, the relative probability $P(K\rightarrow L^2)$ can be estimated from Eq. (11) by replacing $D_0(2l,2l)^2$ by the sum $D_0(2l,2l)^2+D_0(3l,2l)^2$ for $l=0,1$. In the case of $K\rightarrow M^2$ transitions in argonlike ions, we must replace $D_0(2l,2l)^2$ by the sum $D_0(3l,3l)^2+D_0(2l,3l)^2$ and the ratio $E_{K\rightarrow L2}/E_{K\rightarrow L}$ by $E_{K\rightarrow M2}/E_{K\rightarrow M}$.

In order to evaluate the relative probability $P(K\rightarrow L^2)$ or $P(K\rightarrow M^2)$, we need SCF one-electron wave functions of the $(1s)^{-1}{}^2S$, $(2p)^{-1}{}^2P$, and $(3p)^{-1}{}^2P$ states. For F^-, Ne, Na^+, Cl^-, Ar, and K^+ we can use SCF wave functions calculated by Bagus.[11] The results are given in Table I. In C we have put $E_{K\text{-}X^2}\cong I_{1s}-I_{np2}$ ($n=2$, 3). This is justified since in the hydrogenic approximation $D_0(\epsilon p,2p)^2\propto\epsilon^{-3}$ for high ϵ and since the s part of the probability is only about 20% of the p part. We note from Table I that the exchange correction $D_0(3l,2l)^2$ is very important for $K\rightarrow L^2$ transitions when $Z\gtrsim 17$. The $P(K\rightarrow L^2)$ and $P(K\rightarrow M^2)$ values vary quite smoothly as a function of Z, as is clear from the hydrogenic approximation. The probability curves in Fig. 1 are based on an interpolation and extrapolation of the B values in Table I.

B. Comparison with Experiments on $K\rightarrow L^2$, $K\rightarrow M^2$, and $L\rightarrow M^2$ Transitions

According to Fig. 1, double-electron transitions of the radiative Auger type should be observable in a wide range of the K x-ray spectrum. Since excitation to bound states is possible and since the final-state configuration has several terms, the

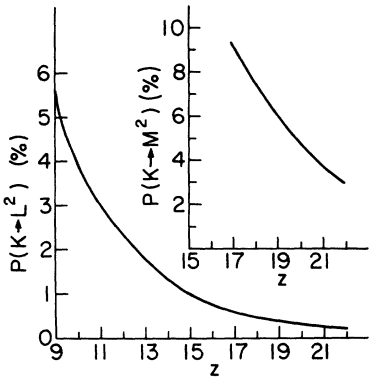

FIG. 1. Relative shake-off probabilities of $K \rightarrow L^2$ and $K \rightarrow M^2$ transitions as a function of the atomic number Z. Values at $Z = 9$ and 17 correspond to the negative ions.

radiative Auger spectrum would look rather complicated. Recent observations indeed show photon distributions with sharp peaks and overlapping continua on the low-energy side of $K\alpha$ and $K\beta$ lines in various elements and compounds.[1-4,19] The asymmetric shape of these distributions is very different from the shape of other possible x-ray satellite lines.

Only preliminary estimates of total integrated intensities are available for $K \rightarrow L^2$ and $K \rightarrow M^2$ transitions. Besides that, most of the measurements are made with solid targets. However, it seems to be clear that the relative integrated $K \rightarrow L^2$ intensities are not as large as predicted by Fig. 1 for low-Z values. In Mg and Al, they are probably less than 1%.[2] The $K \rightarrow L^2$ structure in Ne also seems to be very weak.[20] For $Z \geq 14$, the agreement between the calculated and measured values is better. Preliminary experimental values are 0.7% (Si), 0.8% (S), and 0.6% (Cl^-).[21] In Ti, the intensity is about 0.1%.[22] A study of $K \rightarrow M^2$ transitions indicates that the relative intensity drops from about 6 to 2% when Z goes from 17 to 22.[23] In sulfur the intensity is about 3%.[23] Again the calculations seem to overestimate the intensities at least for low-Z values but not as much as for $K \rightarrow L^2$ transitions in Mg and Al.

Cooper and LaVilla[5] studied the low-energy satellites of the $L_{\eta,t}$ line $[(2p)^{-1}\,{}^2P \rightarrow (3s)^{-1}\,{}^2S]$ in Ar and KCl. They suggested that the main satellite structure is due to $(2p)^{-1}\,{}^2P \rightarrow (3p)^{-2}\epsilon d\,{}^2S$ double-electron transitions. According to Eq. (4), these satellites cannot be due to shake-off even if ϵd is replaced by ϵp. Terms of type $[D_0(3p, 2p)D_1(2p, \epsilon d)]^2$ may contribute slightly to the intensity but do not explain the strong intensity observed. Hence, as Cooper and LaVilla[5] suggested, the final-state interaction between the $(3s)^{-1}$ single-hole configuration and the $(3p)^{-2}\epsilon d$ double-hole configurations probably explains the strong intensity. They also suggested that $(2p)^{-1}\,{}^2P \rightarrow (3s)^{-1}(3p)^{-1}\epsilon p\,{}^2S$ transitions contribute to the tail of the $L_{\eta,t}$ satellites. These transitions occur also as a consequence of shake-off. The dominating intensity term is of the type $[D_0(\epsilon p, 3p)D_1(2p, 3s)]^2$. The relative shake-off probability $P(L_{2,3} \rightarrow M_1 M_{2,3})$ can be calculated from an expression which is analogous to (11) and where $C \simeq \frac{3}{4}$. Bagus's wave functions yield 3.8% (Cl^-), 2.9% (Ar), and 2.3% (K^+). According to these results, the $L_{\eta,t}$ low-energy satellites should indeed have an observable low-energy tail in Ar and KCl.

IV. DISCUSSION

According to Sec. III B, there seems to be a tendency for the $K \rightarrow L^2$ and $K \rightarrow M^2$ shake-off probabilities to overestimate the observed probabilities for low Z. This tendency is stronger for $K \rightarrow L^2$ transitions which correspond to much weaker shake-off probabilities than the $K \rightarrow M^2$ transitions. A possible explanation is that our calculation neglects the interaction between single- and double-hole configurations in the final state. This interaction is more important for low Z than for high Z and could reduce the intensity according to Eq. (7).

In this connection it is important to realize that we have based our calculations on open-shell SCF wave functions[11] which were obtained by the Roothaan analytic-expansion method.[24] Brillouin's theorem is not valid for these wave functions, except in the closed-shell ^1S case.[25] This means that there is an interaction between the unexcited and the singly excited configurations. For example, the matrix elements $\langle (2p)^{-2}\epsilon p\,{}^2P | H | (2p)^{-1}\,{}^2P \rangle$ are not zero if they are constructed from solutions of the $(2p)^{-1}\,{}^2P$ Roothaan SCF equations. It is also important to note that if the excited ϵp orbitals are chosen to be the virtual orbitals of the SCF equation of the $2p$ orbital, they resemble an electron moving in a field of the $(2p)^{-1}$ core rather than of the $(2p)^{-2}$ core. This unphysical situation can be avoided by adding a suitable Hermitian operator to the original SCF operator which changes the virtual orbitals but not the $2p$ orbital.[26] Hence, this procedure would not affect the shake-off probability $1 - D_0(2p, 2p)^2$. However, if both shake-off and configuration interaction have to be taken into account, then their relative strength and also the convergence of the CI expansion would depend on the choice of the virtual orbitals.

The method applied in Sec. III to the calculation of photon-electron emission probabilities is similar to the sudden approximation method[27,28] that has been used in calculations of shake-off following inner-shell ionization by photon and electron impact. In particular, sudden approximation calculations based on Bagus's wave func-

tions have successfully predicted probabilities of simultaneous K ionization and L or M excitation.[28,29] This may be partly due to the fact that Bagus's $(1s)^{-1}\,^2S$ wave function does not interact strongly with the wave functions that can be constructed for the $(1s)^{-1}(np)^{-1}\epsilon p\,^2S$ configuration. In fact, the matrix element $\langle(1s)^{-1}(np)^{-1}\epsilon p\,^2S\,|H|\,(1s)^{-1}\,^2S\rangle$ is proportional to the unimportant exchange integral $R^1(1s, 1s, np, \epsilon p)$.

It is well known that the soft-x-ray emission bands of metals have a low-energy tail that cannot be explained by the one-electron model. Pirenne and Longe[14] attributed the formation of the tail to double-electron transitions where conduction electrons are excited into levels above the Fermi level during the x-ray emission process. They considered the change of the self-consistent field acting on the conduction electrons and derived an intensity formula which is a combination of monopole and dipole matrix elements. Hence, our interpretation of radiative Auger transitions as monopole-dipole shake-off transitions is an application of Pirenne and Longe's idea to isolated atoms. Landsberg,[14] on the other hand, attributed the tailing to an internal Auger effect in the final state. In this effect the positive final-state hole is filled by a conduction electron and another is ejected to a level above the Fermi level. The Landsberg mechanism corresponds to configuration interaction between final single- and double-hole states.

Both mechanisms have been incorporated in a many-body perturbation treatment[30] of electron-interaction effects on the soft-x-ray emission from metals. The many-body perturbation theory also accounts for plasmon emission. It has been found that there is a strong cancellation among contributions that correspond to the Pirenne-Longe and Landsberg mechanisms, at least in the first order.[30] This situation is analogous to what we have suggested to be the case for $K \rightarrow L^2$ and $K \rightarrow M^2$ transitions in free atoms.

V. CONCLUSION

Our treatment of the radiative Auger effect and the theory of electron-interaction effects on the soft-x-ray emission from metals predict that simultaneous photon-electron emission effects should be observable on the low-energy side of many ordinary x-ray lines. They may appear as separate photon distributions like the $K \rightarrow L^2$ and $K \rightarrow M^2$ transitions or merely as tails or asymmetries of the x-ray line. A part of these double-electron transitions can be interpreted as being due to shake-off. In the case of $K \rightarrow L^2$ and $K \rightarrow M^2$ transitions, the calculated shake-off probabilities are of the same order of magnitude as the observed probabilities. However, shake-off is not the only electron-electron interaction mechanism that contributes to the radiative Auger probability. In particular, we suggest that configuration interaction in the final state between single- and double-hole states is important.

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K- and *L*-Shell Electron Shake-off in the Beta Decay of ^{204}Tl†

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Electron shake-off has been investigated in the decay of ^{204}Tl in a measurement of the relative intensities of the Hg and Pb K and L x rays. Values of $(1.0\pm0.1)\times10^{-2}\%$ and $(20.0\pm2.0)\times10^{-2}\%$ have been obtained for the K-shell and total L-shell electron shake-off probabilities, respectively. Approximations of individual L-subshell shake-off probabilities resulted in values of $(2.0\pm0.6)\times10^{-2}\%$, $(9.3\pm0.6)\times10^{-2}\%$, and $(9.0\pm0.6)\times10^{-2}\%$ for the $L_{\rm I}$, $L_{\rm II}$, and $L_{\rm III}$ subshells, respectively. These values, in some cases, differ by as much as a factor of 3 from theoretical predictions.

I. INTRODUCTION

Electron shake-off has been described as a process in which an atomic electron is transferred to an excited state or to the continuum as the result of a sudden change in the central potential of the atom. Such a change in the effective Z of the atom may be caused by any of a number of different processes; however, electron shake-off due to β^- decay has probably been studied most extensively. Recent theoretical calculations by Carlson *et al.*[1] of the electron shake-off probabilities due to β^- decay for all atomic shells and many values of Z have made it desirable to obtain further experimental data concerning K- and L-shell electron shake-off probabilities. In the past few years a number of measurements of the K-shell shake-off probability have been made for the β^- decay of ^{210}Bi. Several of these measurements result in a K-shell shake-off probability which is lower by a factor of nearly 2 than that calculated by Carlson *et al.*[1] Virtually no measurements of the L-shell shake-off probabilities have been made using x-ray techniques for nuclides of large Z. Those L-shell shake-off probabilities which have been calculated were for nuclides of low Z and calculations were made on the basis of the relative abundances of differently charged ions following β^- decay.[2] In view of these facts, it was felt that a critical investigation into the K- and L-shell electron shake-off probabilities for a high-Z nuclide was needed.

II. EXPERIMENT

For this investigation, ^{204}Tl was chosen as the nuclide of study because it has several well-known properties which make it particularly suitable to a relatively direct measurement of the K and L electron shake-off probabilities for $Z=82$. As seen in Fig. 1, ^{204}Tl decays by β^- to Pb and by electron capture to Hg. The branching ratio of electron capture to β^- emission is stated by Klein and Leutz[3] as $(2.29\pm0.06)\%$. Furthermore, Klein and Leutz determined the L/K capture ratio to be 0.55 ± 0.05. Using a high-resolution Si(Li) detector it appeared possible to observe the Pb K and L x rays, 68–85 and 9–15 keV, respectively, resulting from electron shake-off with the Hg K and L x rays from the electron capture branch. Based on the above data and Carlson's values for the shake-off probability, a calculation of the predicted Hg-to-Pb intensity ratio may be made. Calculations of the shake-off probabilities may then be made using the predicted and measured values of the Hg-to-Pb ratio. This method has the advantages that no knowledge of detector efficiency, solid angle, or the absolute β^- count is necessary. Furthermore, the experimental procedure is extremely simple and straightforward.

The experiment utilized a Nuclear Data 4096 channel multichannel analyzer with a Tennelec 202BLR amplifier and a Kevex No. 3000 Si(Li) detector having a resolution of about 260 eV at 5.90 keV. Two sources were used in this experiment. The first source, approximately 10 μCi, was prepared by evaporating the desired quantity of $^{204}TlNO_3$ in solution with HNO_3 from the surface of a NE 111 disk 5.08 cm in diameter and 0.29 cm thick. Calculation indicated that this thickness was adequate to stop the 765-keV β^- particles from the decay; therefore, this unwanted background was