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Electron shakeoff accompanying internal conversion

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The electron shakeoff probabilities accompanying the internal conversion process have been calculated in the sudden approximation, using screened relativistic hydrogenic wave functions. The screening constants have been determined from the relativistic self-consistent–field calculations and the presence of a vacancy resulting from internal conversion has been taken into account. The present results indicate that relativistic effects cause an appreciable increase in the probabilities. It is also shown that the prediction of Carlson *et al.* is a good approximation to the shakeoff probability accompanying internal conversion. The calculated results have been compared with available experimental data. There is fairly good agreement between the calculated and the recently measured values. The need for new data is emphasized.

[RADIOACTIVITY ^{57}Fe, ^{109}Ag, ^{113,114}In, ^{131}Xe, ^{137}Ba, ^{141}Pr, ^{150}Sm, ^{203}Tl; calculated shakeoff probability accompanying internal conversion.]

I. INTRODUCTION

In the electromagnetic transition between two nuclear levels, the simultaneous emission of two atomic electrons is possible as a second-order process. The transition energy is shared between two electrons, and the electrons emitted have a continuous energy distribution. Such a transition takes place via three different processes, involving either nuclear or atomic intermediate state: (1) double internal conversion (DIC), (2) internal conversion of internal Compton effect (ICICE), and (3) electron shakeoff accompanying internal conversion (SOIC). The first process, suggested by Sachs,[1] is a kind of two-quantum transition of the atomic nucleus. Theoretical studies of this process have been first worked out by Eichler,[2] and in more elegant form by Grechukhin.[3] Listengarten[4] estimated the probability of the second mechanism, which is the secondary effect of the two-quantum nuclear transition. On the other hand, for the third mechanism no theoretical study has so far been made, except for an order-of-magnitude estimation by Seykora and Waltner.[5]

The process where an atomic electron is ejected owing to a sudden change in nuclear charge during radioactive transitions, such as α decay, β decay, and electron capture, has been studied theoretically and experimentally.[6,7] This phenomenon, called internal ionization or electron shakeoff (SO), is also possible in the case of internal-conversion processes where the nuclear charge does not change. In this case, the change in effective nuclear charge resulting from the loss of an electron as a consequence of the internal-conversion process causes ejection of another atomic electron. This process is analogous to the electron shakeoff accompanying the photoelectric effect (SOPE), which has been extensively investigated theoretically and experimentally.[8]

Although the spectral distributions of the ejected electrons are different, it is rather difficult to distinguish experimentally among three processes described above because of very low probabilities. Most experiments for the double-electron ejection process have determined only the upper limit of the probability, and cannot explain clearly from which of the three processes the observed phenomenon comes.

Recent observations of the existence of the satellite lines in the conversion-electron spectra in ^{109}Ag by Briançon, Valadares, and Walen[9] and in ^{57}Fe and ^{137}Ba by Porter, Freedman, and Wagner[10] have established the evidence that the double-electron ejection process in these nuclides is mainly ascribed to the SOIC process.

On the other hand, Carlson *et al.*[11] predicted that the probability of the SOIC or SOPE process must be proportional to the square of the difference in effective nuclear charges experienced by the electron concerned.

In the present paper, we estimate the SOIC probability within a framework of the sudden approximation using the method similar to previous works for internal ionization probability accompanying electron capture.[12-14] Calculations have been performed using relativistic hydrogenic wave functions with screening constants determined from relativistic self-consistent-field (SCF) calculations. The effect of the presence of a hole resulting from internal conversion has been taken into consideration.

II. THEORETICAL MODEL

For simplicity, the sudden approximation is used in the present work. In this approximation the SOIC process is considered as a two-step process; a sudden change in the central potential resulting from internal conversion causes ionization of an orbital electron. This treatment corresponds to the case where the kinetic energy of the conversion electron is extremely high. This implies that the observed electron spectrum is easy to separate into the one concerning the conversion electrons which form a satellite line near to the normal conversion line, and the other concerning the shakeoff electrons which are mainly concentrated in the very low-energy region. The probability for the SO process is independent of the kinetic energy of the conversion electron and the effect of the electron exchange in the final state can be neglected.

In the experiment of the SOPE process by Carlson and Krause,[15] it was found that the SO probability is independent of the kinetic energy of the photoelectron when its energy is approximately three times or more than the SOPE threshold. Sachenko and Burtsev[16] estimated theoretically that the sudden approximation is valid in the SOPE process when the photon energy is 1.2–1.3 times or more than the energy required for double photoionization. These facts suggest that the sudden approximation gives good estimates for the SOIC probabilities when the nuclear transition energy is larger than the threshold for double-electron ejection.

It should be noted that the above statement is concerned only with the case where the two electrons do not come from the same atomic shell. When both electrons are ejected from a shell with the same principal quantum number, the treatment explicitly including the electron correlation is required.[17] However, in the K-shell internal-ionization process accompanying K capture, where the correlation effect between two K electrons is important, we have shown[12] that our theoretical model gives probabilities which are in good agreement with the experiments, and also with the calculations including electron correlation.[18] Therefore, we apply our model to the case of the SOIC process where the two electrons come from the same principal shell.

In the present work, only the SO process is treated and contribution from the direct-collision process is neglected. The direct collision refers to ionization of an orbital electron by the conversion electron through the Coulomb interaction. The relative probability of this process vs the SO process in β^- decay has been estimated by Feinberg[19] to be approximately equal to the ratio of the binding energy of the atomic electron to be ejected to the maximum energy of the β^- particle. This prediction suggests that the direct collision is unimportant in the present case unless the transition energy is very low.

Furthermore, we neglect the inner-shell SO process accompanying internal conversion of the electron with larger principal quantum number than that of the SO electron for the following reason. For example, it is impossible to distinguish experimentally between L shakeoff with K conversion and K shakeoff with L conversion, because the sum of the kinetic energies of the ejected electrons is the same and final vacancies occur in the K and L shells. The ejected-electron spectrum accompanying internal conversion should include both contributions. However, the change in the effective nuclear charge seen by the inner-shell electron during outer-shell conversion should be considerably smaller than that experienced by the outer-shell electron accompanying inner-shell conversion.

Based on the approximations described above, the probability that an orbital electron, initially in the state $\psi_i(Z, n)$, makes a transition to a final continuum state $\psi_f(Z', W)$ following the internal conversion of another orbital electron can be written in relativistic units ($\hbar = m = c = 1$) as

$$P(W)dW = \frac{1}{2\pi^2} |\langle\psi_f(Z', W)|\psi_i(Z, n)\rangle|^2 pW dW, \quad (1)$$

where W is the total energy (including the rest mass) of the ejected electron and $W^2 = p^2 + 1$.

The total SOIC probability per conversion is

$$P = \int_1^\infty P(W)dW. \quad (2)$$

III. ATOMIC MATRIX ELEMENT AND SCREENING CONSTANTS

A. Atomic matrix element

The atomic matrix element in Eq. (1) is given as wave-function overlap:

$$M_A = \langle\psi_f(Z', W)|\psi_i(Z, n)\rangle. \quad (3)$$

Here $\psi_i(Z, n)$ is the wave function of an orbital electron of the initial atom with a set of quantum numbers n, $\psi_f(Z', W)$ is the wave function of a continuum electron with total energy W, and Z and Z' denote the effective nuclear charges seen by the electrons in the initial and final atoms, respectively.

The expression for the electron wave functions in the Coulomb field is given by[20]

$$\psi_\kappa^\mu = \begin{pmatrix} g_\kappa(r)\chi_\kappa^\mu(\hat{r}) \\ i f_\kappa(r)\chi_{-\kappa}^\mu(\hat{r}) \end{pmatrix}, \quad (4)$$

where $\chi_\kappa^\mu(\hat{r})$ is the spin-angular function, and $\kappa = \mp(j+\frac{1}{2})$ for $j = l \pm \frac{1}{2}$. The radial functions for $\psi_i(Z, n)$ are designated by f_κ and g_κ, while $f_{\kappa'}$ and $g_{\kappa'}$ refer to those for $\psi_f(Z', W)$. Then M_A can be written in terms of radial integrals as follows:

$$M_A = R_1 + R_2 . \quad (5)$$

Here the radial integrals are defined as

$$R_1 = \int_0^\infty g_\kappa(r) g_{\kappa'}(r) r^2\, dr , \quad (6a)$$

$$R_2 = \int_0^\infty f_\kappa(r) f_{\kappa'}(r) r^2\, dr . \quad (6b)$$

The bound-state radial wave functions are given by[20]

$$f_\kappa(r) = -\frac{2^{1/2}\lambda^{5/2}}{\Gamma(2\gamma+1)} \left[\frac{\Gamma(2\gamma+n'+1)(1-W_B)}{n'!\,\zeta(\zeta-\lambda\kappa)}\right]^{1/2} \times (2\lambda r)^{\gamma-1} e^{-\lambda r} I_- , \quad (7a)$$

$$g_\kappa(r) = -\frac{2^{1/2}\lambda^{5/2}}{\Gamma(2\gamma+1)} \left[\frac{\Gamma(2\gamma+n'+1)(1+W_B)}{n'!\,\zeta(\zeta-\lambda\kappa)}\right]^{1/2} \times (2\lambda r)^{\gamma-1} e^{-\lambda r} I_+ , \quad (7b)$$

where $I_\pm = n' F(-n'+1, 2\gamma+1; 2\lambda r) \pm (\kappa - \zeta/\lambda) \times F(-n', 2\gamma+1; 2\lambda r)$.

On the other hand, the radial parts of the continuum wave function normalized such that it is asymptotically a plane wave plus outgoing waves are expressed as[20]

$$f_{\kappa'}(r) = i\left(\frac{W-1}{2W}\right)^{1/2} \frac{2\pi^{1/2}(2pr)^{\gamma'-1} e^{\pi y/2} |\Gamma(\gamma'+iy)|}{\Gamma(2\gamma'+1)} J_- , \quad (8a)$$

$$g_{\kappa'}(r) = \left(\frac{W+1}{2W}\right)^{1/2} \frac{2\pi^{1/2}(2pr)^{\gamma'-1} e^{\pi y/2} |\Gamma(\gamma'+iy)|}{\Gamma(2\gamma'+1)} J_+ , \quad (8b)$$

where $J_\pm = e^{-ipr+i\eta}(\gamma'+iy) F(\gamma'+1+iy, 2\gamma'+1; 2ipr) \pm$c.c., and c.c. denotes complex conjugate. Other parameters in Eqs. (7) and (8) are given in Ref. 20.

Writing

$$u = 2p/(p-i\lambda), \quad v = 2\lambda/(p-i\lambda), \quad w = 1/(p-i\lambda),$$

the radial integrals reduce to

$$R_1 = C[(1+W_B)(W+1)]^{1/2} L_+ , \quad (9a)$$

$$R_2 = iC[(1-W_B)(W-1)]^{1/2} L_- , \quad (9b)$$

where

$$C = (-i)^{\gamma+\gamma'+1} \frac{2^{3/2}\pi^{1/2}\lambda^{5/2}\Gamma(\gamma+\gamma'+1)}{\Gamma(2\gamma+1)\Gamma(2\gamma'+1)} \times e^{\pi y/2 + i\eta} |\Gamma(\gamma'+iy)| \left[\frac{\Gamma(2\gamma+n'+1)}{2W_B n'!\,\zeta(\zeta-\lambda\kappa)}\right]^{1/2} \times u^{\gamma'-1} v^{\gamma-1} w^3 ,$$

$$L_\pm = \sum_{m=0}^\infty \frac{(-i)^m v^m (\gamma+\gamma'+1)_m}{(2\gamma+1)_m m!} \times [(-n')_m(\kappa-\zeta/\lambda) \pm (-n'+1)_m n'] \times [(\kappa + iy/W) K_2^{(m)} \mp (\gamma'+iy) K_1^{(m)}] ,$$

$$K_1^{(m)} = F(\gamma'+1+iy, \gamma+\gamma'+1+m; 2\gamma'+1; u),$$

$$K_2^{(m)} = F(\gamma'+iy, \gamma+\gamma'+1+m; 2\gamma'+1; u),$$

and $(j)_m$ denotes $j(j+1)\cdots(j+m-1)$.

For K or L shell, the matrix element M_A reduces to Eq. (24) in Ref. 12 or Eq. (14) in Ref. 13, by letting $N = [\Gamma(2\gamma+n'+1)/n'!\,\zeta(\zeta-\lambda\kappa)]^{1/2}(2\lambda^5)^{1/2}/\Gamma(2\gamma+1)$, $a_0 = n' - \kappa + \zeta/\lambda$, $c_0 = -n' - \kappa + \zeta/\lambda$, $a_1 = c_1 = 2n'\lambda(\kappa - \zeta/\lambda)/(2\gamma+1)$, and $n' = n - |\kappa|$.

In the small-momentum limit, M_A is expressed as

$$\lim_{p\to 0} M_A \sim -\frac{2^{\gamma+\gamma'}\pi\Gamma(\gamma+\gamma'+1)}{\Gamma(2\gamma+1)\Gamma(2\gamma'+1)} \left(\frac{\zeta'}{\lambda}\right)^{\gamma'-1/2} \left[\frac{\Gamma(2\gamma+n'+1)}{n'!\,\zeta(\zeta-\lambda\kappa)}\right]^{1/2} p^{-1/2} \times \sum_{m=0}^\infty \frac{2^m(\gamma+\gamma'+1)_m}{(2\gamma+1)_m m!} \{A_+^{(m)}[(\gamma'-\gamma-m)K_0^{(m-1)} + (\gamma+m-\kappa-2\zeta'/\lambda)K_0^{(m)}] + A_-^{(m)}\zeta' K_0^{(m)}\} , \quad (10)$$

where

$$A_\pm^{(m)} = (1 \pm W_B)^{1/2}[(-n')_m(\kappa-\zeta/\lambda) \pm (-n'+1)_m n']$$

and

$$K_0^{(m)} = F(\gamma+\gamma'+1+m, 2\gamma'+1; -2\zeta'/\lambda) .$$

This expression implies that the SO probability [Eq. (1)] converges to a finite value in the small-momentum limit.

B. Screening constants

In order to take account of the effect of Coulomb interaction between electrons, the screening method is used as in the previous works.[12-14] In this method, the nuclear charge in the hydrogenic wave functions is replaced by an appropriate effective nuclear charge $Z_{\text{eff}} = Z - \sigma$, where σ is well known as the screening constant. Although this approach is not as accurate as SCF and Thomas-Fermi methods, we have shown in the previous work[12] that the proper choice of the screening constants leads to the K-shell probabilities accompanying K-electron capture in good agreement with the experimental results by the use of simple analytical wave functions.

In the initial bound state, σ is calculated from[11]

$$\sigma = Z(1 - \bar{r}_Z/\bar{r}_{\text{SCF}}), \quad (11)$$

where $\bar{r}_Z$ is the mean relativistic hydrogenic radial distance and $\bar{r}_{\text{SCF}}$ is the mean radius determined from the relativistic SCF wave functions.[21]

Using the radial wave functions in Eq. (7), $\bar{r}_Z$ can be expressed as

$$\bar{r}_Z = \frac{\lambda}{4\Gamma(2\gamma+1)} \frac{\Gamma(2\gamma+n'+1)}{n'!\,\zeta(\zeta-\lambda\kappa)} \times[n'^2G(n'-1,n'-1)+2Wn'(\kappa-\zeta/\lambda) \times G(n'-1,n')+(\kappa-\zeta/\lambda)^2G(n',n')], \quad (12)$$

where

$$G(m,m') = \sum_{j=0}^{m} \frac{(-m)_j\,(2\gamma+1+j)}{j!} \times F(-m', 2\gamma+2+j; 2\gamma+1; 1).$$

It is more difficult to select an appropriate screening constant for the continuum electron. The effective charge seen by the ejected electron is steadily decreasing as the electron moves away from the nucleus. In the present work, the screening constant for the continuum state is taken to be the same as that for the bound electron before ejection, because the main contribution to the atomic matrix element comes from the distances of order of the Bohr radius of the bound electron. This choice was first pointed out by Bethe and Salpeter[22] in their discussion of the photoelectric effect and has been used to calculate internal-conversion coefficient.[23,24]

Taking into account the presence of a hole in the inner shell resulting from internal conversion, the screening constant for the final state is determined by

$$\sigma_c = (\sigma_h/\sigma_S)\sigma. \quad (13)$$

Here σ is the screening constant determined from Eq. (11), σ_h is the Slater screening constant for the atom with a vacancy in an inner shell, and σ_S is that for the ordinary atom.[25]

IV. PREDICTION OF CARLSON *et al.* AND NONRELATIVISTIC CALCULATIONS

In order to compare with the present relativistic theory, two kinds of calculations have been made: the prediction of Carlson *et al.* and nonrelativistic theory. These calculations have been used previously to estimate the probability and the energy spectrum of the ejected electrons for the SOIC and SOPE processes.

According to Carlson *et al.*,[11] the SO probability of an electron in a certain atomic shell is proportional to the square of the change of effective nuclear charge ΔZ experienced by that electron as a result of the internal-conversion process, and thus can be expressed as

$$P = (\Delta Z)^2 P_\beta, \quad (14)$$

where P_β is the SO probability accompanying β^- decay, which represents $\Delta Z = 1$.

To demonstrate the importance of the relativistic effect on the SOIC probability, the nonrelativistic calculations have been performed using the modified Levinger theory. In his theory of the SO process in β^- decay, the initial-state wave function is expanded in terms of final-state wave functions, using the nonrelativistic hydrogenic wave functions.[26] To derive the analytical expressions of the SO probabilities for K-, L_1-, and $L_{2,3}$-shell electrons, Levinger made approximations as $Z \gg 1$ and $\Delta Z = 1$. Owing to these approximations, his expressions have so far been used only in combination with the prediction of Carlson *et al.* [Eq. (14)] by introducing a factor ΔZ.

Starting with the wave functions used by Levinger, we derived the exact expressions for the SO probabilities without any approximation. The SO probabilities for K-, L_1-, and $L_{2,3}$-shell electrons per conversion are written by

$$P_K(W)dW = \frac{2^6\zeta^3\zeta'(\zeta-\zeta')^2}{1-e^{-2\pi y}} \frac{e^{-4y\tan^{-1}(p/\zeta)}}{(\zeta^2+p^2)^4}\,dW, \quad (15)$$

$$P_1(W)dW = \frac{2^{11}\zeta^3\zeta'}{1-e^{-2\pi y}} \times\frac{(5\zeta'\zeta^2-4p^2\zeta'-\zeta^3-4\zeta'^2\zeta+4\zeta p^2)^2}{(\zeta^2+4p^2)^6} \times e^{-4y\tan^{-1}(2p/\zeta)}\,dW, \quad (16)$$

$$P_{2,3}(W)dW = \frac{2^{15}\zeta^5\zeta'}{3} \frac{\zeta'^2+p^2}{1-e^{-2\pi y}} \frac{(\zeta'-\zeta)^2}{(\zeta^2+4p^2)^6} \times e^{-4y\tan^{-1}(2p/\zeta)}\,dW, \quad (17)$$

where $\zeta = \alpha Z$, $\zeta' = \alpha Z'$, $y = \zeta' W/p$, and α is the fine structure constant.

Letting $Z' = Z \pm 1$ and neglecting terms $O(\alpha^3)$ in

Eqs. (15)–(17), we obtain

$$P_K(W)dW = \frac{2^6\alpha^2\zeta^4}{1-e^{-2\pi y}}\,\frac{1}{(\zeta^2+p^2)^4}\,e^{-4y\tan^{-1}(p/\zeta)}\,dW\,, \tag{15'}$$

$$P_1(W)dW = \frac{2^{11}\alpha^2\zeta^4}{1-e^{-2\pi y}}\,\frac{(3\zeta^2+4p^2)^2}{(\zeta^2+4p^2)^6} \times e^{-4y\tan^{-1}(2p/\zeta)}\,dW\,, \tag{16'}$$

$$P_{2,3}(W)dW = \frac{2^{15}\alpha^2\zeta^6}{3(1-e^{-2\pi y})}\,\frac{\zeta^2+p^2}{(\zeta^2+4p^2)^6} \times e^{-4y\tan^{-1}(2p/\zeta)}\,dW\,. \tag{17'}$$

These expressions are same as those obtained by Levinger.[26]

V. NUMERICAL RESULTS

We have calculated the electron SO probabilities per internal conversion for nine nuclides experimentally studied. The nuclear transition energies for these nuclides are taken from Lederer, Hollander, and Perlman,[27] and for the binding energies of atomic electrons we used the table prepared by Bearden and Burr.[28] The numerical works have been performed using the FACOM 230-75 computer in the Data Processing Center of Kyoto University.

For K- and L-electron shakeoff accompanying K conversion, we have performed the relativistic calculations using Eq. (1) and the nonrelativistic ones according to Eqs. (15)–(17). To demonstrate the relativistic effects and the effect of the screening constants on the SOIC probability, the values of $P(W)$ are plotted in Figs. 1–3 for ^{137}Ba in three different cases: (1) relativistic theory with relativistic screening constants [Eqs. (11) and (13)], (2) relativistic theory with Slater screening constants, and (3) modified Levinger theory with Slater screening constants. As has been seen in the internal ionization accompanying electron capture,[12] the relativistic probability is larger than the nonrelativistic one when the same choice of screening constants is made. Furthermore, it is clear from the figures that the curve obtained from the relativistic theory with relativistic screening

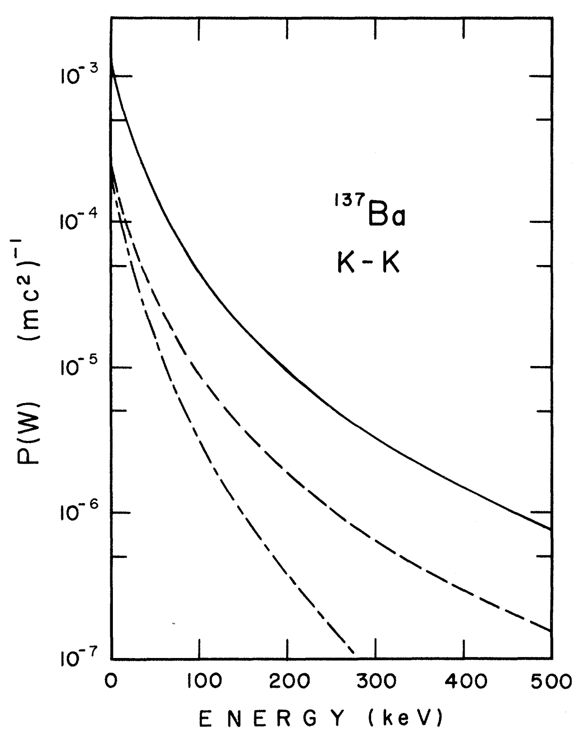

FIG. 1. Transition probability $P(W)$ for K-electron ejection accompanying K conversion of ^{137}Ba. The solid curve has been calculated from relativistic theory with relativistic screening constants; the dashed curve from relativistic theory with Slater screening constants; the dot-dashed curve from modified Levinger theory with Slater screening constants.

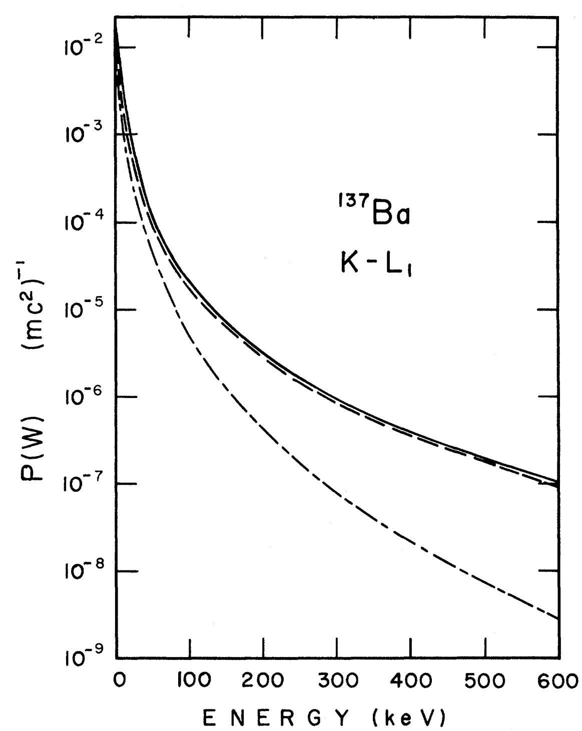

FIG. 2. Transition probability $P(W)$ for L_1-electron ejection accompanying K conversion of ^{137}Ba. The solid curve has been calculated from relativistic theory with relativistic screening constants; the dashed curve from relativistic theory with Slater screening constants; the dot-dashed curve from modified Levinger theory with Slater screening constants.

constants is above the curves with Slater screening constants for all energies. For the case of the L-shell SO process the relativistic screening constants do not play as important a role as in the case of the K-shell SO process.

The total SO probability for K shell per K conversion, P_{KK}, and that for L_i shell, P_i ($i = 1, 2, 3$), obtained in the present numerical calculations are listed in Tables I and II. The columns labeled REL, SL, LEV, and CAR correspond to relativistic theory with Slater screening constants, modified Levinger theory [Eqs. (15)–(17)] with Slater screening constants, and prediction of Carlson *et al.* [Eq. (14)] with relativistic screening constants, respectively. The values in the column CAR are estimated using the P_β values taken from the SCF calculations of Carlson *et al.*[11] It should be noted that the CAR values for the K shell are obtained taking into account the presence of only one K electron in the present case, whereas in the case of the SO process accompanying β^- decay, two K-shell electrons are available. The values for ^{113}In are not included in Table II, because they are equal to those for ^{114}In.

In Table I comparison of the P_{KK} values in the column REL with those in the columns SL and LEV indicates that the relativistic effects do substantially increase the K-shell SOIC probability. This is in contrast to internal ionization in electron capture,[12] where the relativistic effects reduce the probability. The values in the column CAR are larger than those in the column REL.

Inspection of Table II shows that for low-Z nuclides the P_i values in the columns REL, SL, and LEV are almost equal, while for high-Z elements the values in the column SL become about $\frac{1}{2}$ of those in the column REL, and the LEV values further smaller than the SL values.

TABLE I. Comparison of calculated probabilities per K conversion of the K-electron ejection with measured ones.

Nuclide	Energy (keV)	Theoretical $P_{KK}\times 10^5$ REL[a]	SL[b]	LEV[c]	CAR[d]	Experimental $P_{KK}\times 10^5$	Method[e]	Ref.
^{57}Fe	122	16.2	4.51	4.06	20.8	4 ~ 20	F	10
^{109}Ag	88	7.36	1.56	1.11	9.40	68^{+5}_{-14} [f]	A	30
						<100	F	9
^{113}In	393	7.07	1.46	1.01	8.98	<2 [g]	C	31
^{114}In	192	7.07	1.46	1.01	8.98	<22 [h]	D	32
						<13 [h]	D	33
						1.7 ± 0.3 [i]	D	34
						12	B	35
^{131}Xe	164	6.19	1.25	0.808	7.81	≃73 [f]	A	36
						11 ± 2 [f]	A	37
						<4	A	38
^{137}Ba	662	6.03	1.19	0.742	7.58	18 ± 5 [j]	E	39
						<20	F	10
						7.1 ± 3.5	B	40
^{141}Pr	145	5.74	1.09	0.654	7.24	≃10	B	35
^{150}Sm	334	5.46	0.989	0.578	7.05			
^{203}Tl	279	4.58	0.573	0.271	7.53	25	B	35
						4.0 ± 1.5	B	41

[a] Relativistic theory with relativistic screening constants.
[b] Relativistic theory with Slater screening constants.
[c] Modified Levinger theory with Slater screening constants.
[d] Prediction of Carlson *et al.* with the P_β value of Ref. 11 and with relativistic screening constants.
[e] A, B, C, D, E, and F denote the K-x-ray–K-x-ray coincidence experiment, K-x-ray–K-hypersatellite-line coincidence experiment, e^--e^- coincidence experiment with solid-state detectors, e^--e^- coincidence experiment using β-ray spectrometers, e^--e^--K-x-ray triple coincidence experiment, and direct observation of the satellite line in the conversion-electron spectrum, respectively.
[f] Calculated from the ratio of the probability for double K-electron emission to the single γ-ray transition probability.
[g] Energy range 68–276 keV.
[h] Energy and angular distributions are assumed to be isotropic.
[i] Energy spectrum is assumed to be that of DIC.
[j] Energy range 115–472 keV.

In Table III the sum of L-electron SO probabilities per K conversion from all three L subshells, P_{KL}, are listed.

In a manner similar to the P_i values, the probability of L shakeoff accompanying L conversion has been calculated for various combinations of L subshells in two nuclides; ^{109}Ag and ^{114}In. The numerical values of L_j-shell SO probability per L_i conversion, P_{ij} $(i,j=1,2,3)$, are listed in Table IV and compared with the prediction of Carlson *et al.* When two electrons are ejected from the same subshell, the number of available electrons is taken into consideration. It is clear that the prediction of Carlson *et al.* yields about 2 times larger values than the present work.

The L-shell SO probability per L conversion is defined as

$$P_{LL}=\frac{\sum_{i,j}\alpha_i P_{ij}}{\sum_i \alpha_i}\ , \qquad (18)$$

where α_i is the L_i-shell conversion coefficient. The numerical values of α_i are estimated by interpolation from the table prepared by Hager and Seltzer.[29] The P_{LL} values thus obtained are also listed in Table IV.

The relativistic calculations of M-shell SO probability per K conversion have been carried out for ^{109}Ag. The values of $P(W)$ for various M subshells are plotted in Fig. 4. For comparison, the curve for L_1-shell SO probability per K conversion in this nuclide is shown in the figure. The numerical results are listed in Table V and compared with the values according to the prediction of Carlson *et al.* with the P_β values in Ref. 11 and with relativistic screening constants. The values estimated from Eq. (14) are larger than the present values for all M subshells. The sum of all subshells, P_{KM}, is about $\frac{1}{3}$ of the prediction of Carlson *et al.*

VI. COMPARISON WITH EXPERIMENTS

Many experimental studies have been performed on the double-electron ejection process accompa-

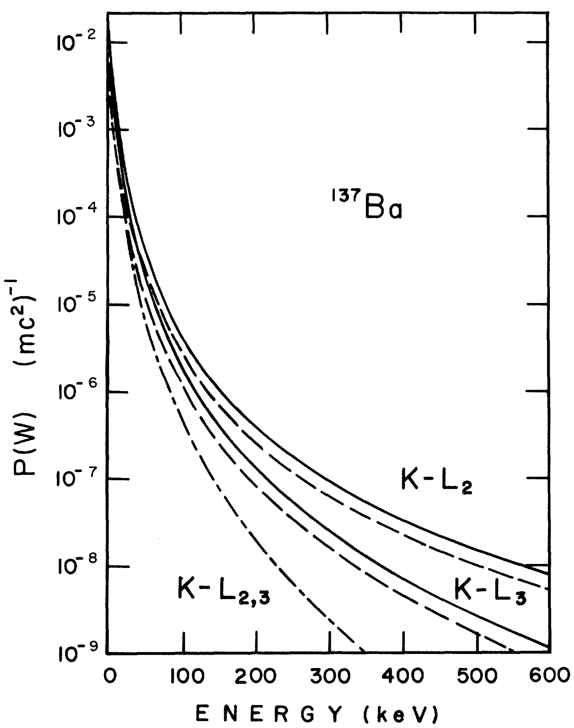

FIG. 3. Transition probabilities $P(W)$ for L_i-electron ejection accompanying K conversion of ^{137}Ba $(i=2,3)$. The solid curves have been calculated from relativistic theory with relativistic screening constants; the dashed curves from relativistic theory with Slater screening constants; the dot-dashed curve from modified Levinger theory with Slater screening constants.

TABLE II. Comparison of calculated probabilities per K conversion of the L_i-shell electron ejection ($\times 10^4$).

Nuclide	Shell	REL[a]	SL[b]	LEV[c]	CAR[d]
^{57}Fe	L_1	9.20	12.4	11.6	20.1
	L_2	7.10	5.56	5.20	24.6
	L_3	13.9	10.6	10.4	50.4
^{109}Ag	L_1	3.97	3.96	3.05	7.59
	L_2	2.95	1.83	1.41	7.65
	L_3	5.17	3.09	2.81	13.9
^{114}In	L_1	3.87	3.69	2.78	7.27
	L_2	2.80	1.71	1.28	7.08
	L_3	4.84	2.82	2.57	12.8
^{131}Xe	L_1	3.39	3.16	2.22	6.13
	L_2	2.54	1.47	1.03	6.09
	L_3	4.20	2.32	2.07	10.5
^{137}Ba	L_1	3.28	2.98	2.05	5.86
	L_2	2.46	1.40	0.953	5.76
	L_3	3.91	2.16	1.91	9.73
^{141}Pr	L_1	3.11	2.77	1.81	5.48
	L_2	2.33	1.30	0.847	5.31
	L_3	3.71	1.94	1.69	8.93
^{150}Sm	L_1	2.96	2.58	1.62	5.16
	L_2	2.22	1.22	0.757	4.94
	L_3	3.42	1.76	1.51	8.13
^{203}Tl	L_1	2.54	1.94	0.850	4.16
	L_2	2.02	0.972	0.408	4.08
	L_3	2.41	1.06	0.816	5.27

[a] Relativistic theory with relativistic screening constants.

[b] Relativistic theory with Slater screening constants.

[c] Modified Levinger theory with Slater screening constants.

[d] Prediction of Carlson *et al.* with the P_β value of Ref. 11 and with relativistic screening constants.

TABLE III. Comparison of calculated probabilities per K conversion of the L-electron ejection with measured ones ($\times 10^4$).

Nuclide	Theoretical $\sum_i P_i$ REL[a]	SL[b]	CAR[c]	Experimental	Method[d]	Ref.
^{57}Fe	30.2	28.6	95.1	90	F	10
^{109}Ag	12.1	8.88	29.1	20	F	9
^{113}In	11.5	8.22	27.2	<1.8[e]	C	31
^{114}In	11.5	8.22	27.2	~3	D	42
				<2.6[f]	D	32
				<10[f]	D	33
^{131}Xe	10.1	6.95	22.7			
^{137}Ba	9.65	6.54	21.4	10	F	10
^{141}Pr	9.15	6.01	19.7			
^{150}Sm	8.60	5.56	18.2	≃4[g]	F	43
^{203}Tl	6.97	3.97	13.5			

[a] Relativistic theory with relativistic screening constants.
[b] Relativistic theory with Slater screening constants.
[c] Prediction of Carlson *et al.* with the P_β value of Ref. 11 and with relativistic screening constants.
[d] Symbols are defined in Table I.
[e] Energy range 79–280 keV.
[f] Energy and angular distributions are assumed to be isotropic.
[g] Quoted from Ref. 44.

nying internal conversion.[7,44] However, some of these experimental results were treated as the DIC or ICICE process and analyzed using the energy and angular distributions of ejected electrons different from the SOIC process. For this reason, in order to compare the calculated values with the experimental results clarification of the adopted experimental conditions is needed.

The experiments of the double-electron ejection accompanying internal conversion may be divided into six categories: (A) x-ray–x-ray coincidence, (B) x-ray–x′(hypersatellite)-ray coincidence, (C) e^--e^- coincidence with solid-state detectors, (D) e^--e^- coincidence using β-ray spectrometers, (E) e^--e^-–x-ray triple coincidence, and (F) direct observation of the satellite line in the conversion-electron spectrum.

Experimental data for double K-electron ejection are listed in Table I. Many experiments in the table give only the upper limits of the probability, which are larger than all the theoretical predictions. The Heidelberg group[30,37] obtained positive evidence of the double K-electron ejection process. However, their large values may be due to the poor resolving power of the detectors employed. In the (A)-type experiments with poor-resolution

TABLE IV. Comparison of calculated probabilities per L conversion of the L-electron ejection with measured ones.

Nuclide	CE[a]	SE[b]	Theoretical, $10^5 P_{ij}$ REL[c]	CAR[d]	Theoretical, $10^4 P_{LL}$ REL[c]	CAR[d]	Experimental, $P_{LL}\times 10^4$	Ref.
^{109}Ag	L_1	L_1	3.47	6.43				
	L_1	L_2	5.29	12.9				
	L_1	L_3	9.41	23.7				
	L_2	L_1	6.94	12.9				
	L_2	L_2	2.65	6.47	1.91	4.33	≃8	9
	L_2	L_3	9.41	23.7				
	L_3	L_1	6.94	12.9				
	L_3	L_2	5.30	12.9				
	L_3	L_3	7.06	17.8				
^{113}In[e]					1.83	4.08	<0.29[f]	31
^{114}In	L_1	L_1	3.44	6.22				
	L_1	L_2	4.94	11.8				
	L_1	L_3	9.02	22.4				
	L_2	L_1	6.88	12.4				
	L_2	L_2	2.47	5.88	1.83	4.08	<2.8[g]	32
	L_2	L_3	9.02	22.4				
	L_3	L_1	6.88	12.4				
	L_3	L_2	4.94	11.8				
	L_3	L_3	6.77	16.8				

[a] Conversion electron.
[b] Shakeoff electron.
[c] Relativistic theory with relativistic screening constants.
[d] Prediction of Carlson *et al.* with the P_β value of Ref. 11 and with relativistic screening constants.
[e] Theoretical values are the same as ^{114}In.
[f] Energy range 91–292 keV.
[g] Energy and angular distributions are assumed to be isotropic.

detectors such as NaI(Tl) crystals, the continuum and spurious x rays would not be resolved from the K x-ray peak and would, therefore, tend to raise the P_{KK} value. On the other hand, little may be concluded from direct comparison of the (C)-, (D)-, and (E)-type experiments with the calculated P_{KK} values, because the measured energy range of the ejected electrons is limited and the total probabilities are obtained assuming an energy spectrum different from the SOIC process.

The (F)-type experiment has the advantage that observation of the complementary SO satellites is direct evidence of the SOIC process. However, experimental errors are large and the probabilities obtained are strongly dependent on the shape of the background curve. This is because the complementary SO satellites are, in general, located on the tail of the high-intensity normal conversion line and extend down to zero energy. Owing to large uncertainty, the P_{KK} value of Porter, Freedman, and Wagner[10] for ^{57}Fe is in agreement with all the theoretical values.

Briand *et al.*[35] deduced the P_{KK} values for three nuclides by the (B)-type experiment which are larger than all the calculated values. For ^{203}Tl, Desclaux *et al.*[41] claimed that their large value

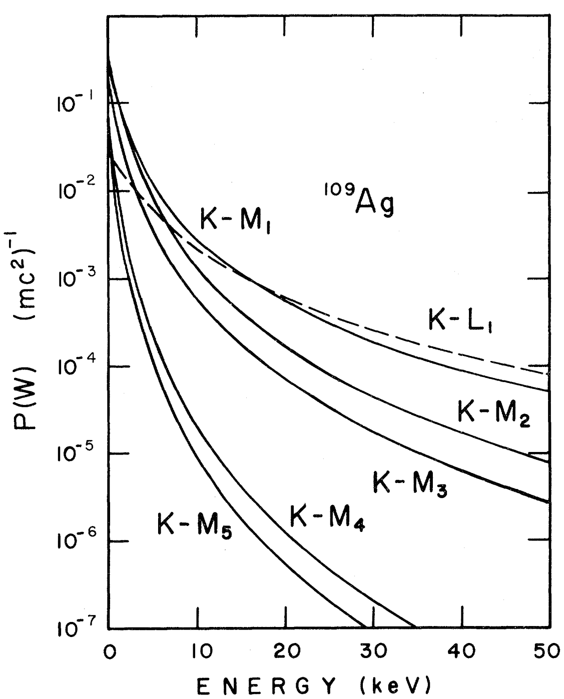

FIG. 4. Transition probabilities $P(W)$ for the SOIC process of ^{109}Ag. The solid curve K-M_i represents M_i-shell electron ejection during K conversion ($i = 1 \sim 5$), while the dashed curve K-L_1 indicates L_1-shell electron ejection accompanying K conversion.

is due to misinterpretation of the hypersatellite line, and estimated the P_{KK} value which is 5 times as small as the value of Briand *et al.* Recently, Briand *et al.*[40] reinvestigated the double K-shell ionization process for several nuclides by the (B)-type experiment. They observed the energy shift of the K hypersatellite line in good agreement with that of Desclaux *et al.*,[41] but the P_{KK} value is reported only for ^{137}Ba.

Of the experimental values in Table I, the most reliable ones are the recent measurements of Briand *et al.*[40] for ^{137}Ba $(7.1 \pm 3.5) \times 10^{-5}$ and of Desclaux *et al.*[41] for ^{203}Tl $(4.0 \pm 1.5) \times 10^{-5}$. These values are in satisfactory agreement with the present numerical values, 6.03×10^{-5} and 4.58×10^{-5}, but do not agree with other theoretical estimations.

Eight P_{KL} measurements are reported and listed in Table III. As has been described, direct comparison of the (C)- and (D)-type experiments with the calculated values is not possible and three of them give only upper limits. The results of comparisons with the four experimental values obtained by the (F)-type experiments are not as simple. The P_{KL} value of Briançon, Valadares, and Walen[9] for ^{109}Ag lies between the present value and the value calculated from the prediction of Carlson *et al.* The experimental value of Porter, Freedman, and Wagner[10] for ^{57}Fe agrees well with the prediction of Carlson *et al.*, while that for ^{137}Ba is in good agreement with the present calculation. The experimental result of Prokofiev[43] for ^{150}Sm is quoted from Ref. 44 and the details of the experiment are not known.

Three experiments for the P_{LL} values are listed in Table IV. The value of Briançon, Valadares, and Walen[9] for ^{109}Ag, obtained by the (F)-type experiment, is larger than both theoretical estimates. The other two P_{LL} values do not allow direct comparison. The upper limit for ^{113}In determined from the (C)-type experiment of Sommer, Knauf, and Klewe-Nebenius[31] is limited in the

TABLE V. Comparison of calculated probabilities per K conversion of the M-electron ejection with measured ones ($\times 10^3$).

Nuclide	Shell	Theoretical REL[a]		Theoretical CAR[b]		Experimental	Ref.
^{109}Ag	M_1	1.61		2.36			
	M_2	1.50		2.98			
	M_3	1.46	5.04	5.78	16.48	10	9
	M_4	0.269		2.17			
	M_5	0.197		3.19			

[a] Relativistic theory with relativistic screening constants.

[b] Prediction of Carlson *et al.* with the P_β value of Ref. 11 and with relativistic screening constants.

TABLE VI. Comparison of the SOIC probabilities with the prediction of Carlson *et al.* ($\times 10^4$).

Nuclide	Shell	Relativistic screening P [a]	$(\Delta Z)^2\overline{P}_\beta$ [b]	$(\Delta Z)^2 P_\beta$ [c]	Slater screening P [d]	$(\Delta Z)^2\overline{P}_\beta$ [e]	$(\Delta Z)^2 P_\beta$ [f]
^{57}Fe	K	1.62	1.47	2.08	0.451	0.417	0.594
	L_1	9.20	6.80	20.1	12.4	8.98	26.5
	L_2	7.10	5.03	24.6	5.57	4.03	19.7
	L_3	13.9	9.63	50.4	10.6	7.55	39.5
^{109}Ag	K	0.736	0.708	0.940	0.156	0.149	0.198
	L_1	3.97	3.40	7.59	3.96	3.40	7.95
	L_2	2.95	2.50	7.65	1.83	1.58	4.83
	L_3	5.17	4.19	13.9	3.09	2.55	8.45
^{114}In	K	0.707	0.681	0.898	0.146	0.140	0.185
	L_1	3.87	3.35	7.27	3.69	3.20	6.94
	L_2	2.80	2.40	7.08	1.71	1.49	4.39
	L_3	4.84	3.95	12.8	2.82	2.36	7.66
^{137}Ba	K	0.603	0.584	0.758	0.119	0.114	0.149
	L_1	3.28	2.93	5.86	2.99	2.67	5.35
	L_2	2.46	2.18	5.76	1.40	1.26	3.32
	L_3	3.91	3.32	9.73	2.15	1.85	5.41

[a] Present theory with relativistic screening constants.
[b] $\overline{P}_\beta$ value from Eq. (19) and ΔZ from relativistic definition.
[c] P_β value from Ref. 11 and ΔZ from relativistic definition.
[d] Present theory with Slater screening constants.
[e] $\overline{P}_\beta$ value from Eq. (19) and ΔZ from Slater's recipe.
[f] P_β value from Ref. 11 and ΔZ from Slater's recipe.

measured energy range. In the case of ^{114}In, the upper limit is obtained from the (C)-type experiment of Kleinheinz *et al.*,[32] assuming isotropic energy and angular distributions.

There has been only one experiment for the M-shell SO process. The P_{KM} value obtained by Briançon, Valadares, and Walen[9] from the (F)-type experiment is between the present value and the prediction of Carlson *et al.*

VII. DISCUSSION

The comparison of the experimental results with the calculated values is not conclusive because of the limited amount of reliable experimental data and large experimental errors. Furthermore, some experimental values cannot be directly compared with the present calculations, since the energy spectrum of the ejected electrons was measured in the limited energy region and the total probability was estimated based on some assumptions. However, the agreement between the present relativistic values and the recently reported P_{KK} values by Briand *et al.*[40] for ^{137}Ba and by Desclaux *et al.*[41] for ^{203}Tl indicates that the model presented here is fairly satisfactory.

As can be seen from Tables I–V, the prediction of Carlson *et al.* yields larger probability than the present theory. It should be noted, however, that the CAR values in the tables are estimated using the P_β values calculated by Carlson *et al.*,[11] which are based on a quite different model from the present calculations.

In order to examine the validity of the prediction of Carlson *et al.* in the SOIC process, we must use the SO probability during β^- decay in the present model. This probability, $\overline{P}_\beta$, is defined as

$$\overline{P}_\beta = \int_1^\infty P_\beta(W)dW\,, \qquad (19)$$

where $P_\beta(W)dW$ is estimated from Eq. (1) by letting $\sigma=0$ for the initial state and $\sigma=-1$ for the final state.

Estimation of the SOIC probability according to the prediction of Carlson *et al.* [Eq. (14)] has been performed for ^{57}Fe, ^{109}Ag, ^{114}In, and ^{137}Ba, using the $\overline{P}_\beta$ values and the ΔZ values determined from relativistic screening constants and from Slater's screening constants. The calculated $(\Delta Z)^2\overline{P}_\beta$ values are listed in Table VI and compared with the P (P_{KK} or P_i) values and the $(\Delta Z)^2 P_\beta$ values using the P_β values in Ref. 11.

Table VI indicates that the prediction of Carlson *et al.* in the present model, $(\Delta Z)^2\overline{P}_\beta$, is slightly smaller than the P values, but is a good approximation to the latter. On the other hand, for the same choice of screening constants the $(\Delta Z)^2 P_\beta$

values are always larger than the P and $(\Delta Z)^2\overline{P}_\beta$ values. This means that the largeness of the prediction of Carlson *et al.* in Tables I–V is attributed to the largeness of the P_β values of Carlson *et al.*[11] It should be noted that their values include the contribution from the shakeup process to an unoccupied bound state.

In conclusion, we have presented a model to estimate the SOIC probability. It is also shown that the prediction of Carlson *et al.* gives an approximate value of the probability. The calculated values are in good agreement with the most reliable experimental results, but more high precision experimental data are needed. Further theoretical and experimental studies on the ejected-electron spectrum of the SOIC process are anticipated.

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