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M-Shell Internal Ionization Accompanying Electron Capture

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The M-shell electron ejection probabilities accompanying electron capture decay have been studied relativistically in the sudden approximation, using screened hydrogenic wave functions. The M-subshell ionization probabilities per electron capture for K, L, and M shells and the energy spectra of the ejected electrons have been calculated. The screening constants have been determined from the relativistic self-consistent-field calculations, taking into account the presence of the vacancy produced in electron capture. The obtained results are compared with the prediction of Carlson *et al.* and with the Hartree-Fock calculations of Crasemann *et al.* The difference between present model and other models is discussed.

KEY WORDS: M-Shell Internal Ionization Probability/ Electron Capture Decay/ Ejected Electron Spectrum/

I. INTRODUCTION

When a radioactive nucleus decays by orbital electron capture, there is a small probability that another orbital electron in the same atom is ejected due to sudden change in the central potential. This process, called *internal ionization accompanying electron capture* (IIEC), has been extensively studied both theoretically and experimentally.[1] However, most investigations are concerned with the K-shell ionization during K-electron capture and only limited number of theoretical results have been published for L-electron ejection.[2-6] For M-shell electron ejection during electron capture, there has been reported no theoretical and experimental work.

Crasemann *et al.*[7] have calculated the atomic electron excitation probability from various atomic states during orbital electron capture. They used the Hartree-Fock wave functions. However, their calculations are nonrelativistic and based on the two-step model, *i.e.* the phase-space factor is neglected. This means that the excitation probability is independent of the transition energy. Furthermore, their results include the contributions from the shake-up process; the electron transition to an unoccupied bound state.

Recently the studies of higher-shell electron capture decay have received special attention. De Rújula[8] proposed that internal bremsstrahlung in electron capture (IBEC) may provide a new and superior approach for assessing the rest mass of electron neutrino. As suitable nuclides for this method, small Q-value nuclides, such as ^{157}Tb,[9] ^{163}Ho,[10,11] and ^{193}Pt,[12] have been studied. In these nuclides, the K-electron ejection in K capture is energetically forbidden because of low-transition

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energy. Although De Rújula claimed that the atomic and molecular problems in IBEC are negligible, his estimation is based on rough assumption.[8] De Rújula and Lusignoli[13] recently proposed to measure the Auger electron spectrum in electron-capture decay of ^{163}Ho. In this case, the main contribution of the background comes from IIEC.

In the present work, the M-shell internal ionization probabilities accompanying K-, L_I-, and M_I-electron capture have been calculated for ^{55}Fe, ^{71}Ge, ^{109}Cd, and ^{131}Cs by the use of screened relativistic hydrogenic (Dirac) wave functions. The calculations are made in the one-step model and the phase-space sharing between the neutrino and the ejected electron is correctly taken into consideration. The energy spectra of the ejected electrons have also been calculated. The present model is similar to that used for K-electron ejection in K capture[14] and for L-electron ejection in L-capture.[5]

II. THEORETICAL MODEL

According to the first-order perturbation theory, the internal ionization probability per electron capture that the electron is ejected with the total energy W is given by[14]

$$P(W)\mathrm{d}W = \frac{1}{2\pi^2}|M_A|^2 \frac{S(W_K-W)}{S(W_0)} \frac{(W_K-W_0)^2}{W_0^2} pW\mathrm{d}W\,, \tag{1}$$

where M_A is the atomic matrix element, $S(W)$ is the energy-dependent shape factor of electron capture decay, W_0 is the transition energy, and p is the momentum of the ejected electron. Throughout the present work relativistic units ($\hbar=m=c=1$) are used.

The transition energy is expressed as

$$W_0 = Q - B_i + 1\,, \tag{2}$$

where Q is the Q value of the transition and B_i is the binding energy of the electron to be captured in the parent atom. The energy W_K denotes

$$W_K = W_0 + 1 - B_j\,, \tag{3}$$

where B_j is the binding energy of the electron to be ejected in the daughter atom. The total internal ionization probability per capture is given by

$$P = \int_1^{W_K} P(W)\mathrm{d}W\,. \tag{4}$$

The atomic matrix element is defined as wave-function overlap:

$$M_A = \langle \psi_f(Z', W) | \psi_i(Z, B) \rangle\,. \tag{5}$$

Here $\psi_i(Z, B)$ is the bound-state wave function of the parent atom with atomic number Z and $\psi_f(Z', W)$ is the wave function for a continuum electron with total energy W in the field of the daughter atom of the nuclear charge Z'.

We use the relativistic hydrogenic wave functions for initial and final states:[15)]

$$\psi(r) = \begin{pmatrix} g_\kappa(r)\chi_\kappa^\mu(\hat{r}) \\ if_\kappa(r)\chi_{-\kappa}^\mu(\hat{r}) \end{pmatrix}, \tag{6}$$

where $g_\kappa(r)$ and $f_\kappa(r)$ are the radial wave functions, $\chi_\kappa^\mu(\hat{r})$ is the spin-angular function, κ and μ are the quantum numbers. The explicit expressions for the bound and continuum wave functions are given in Ref. 15.

Using these wave functions, the atomic matrix element in Eq. (5) can be obtained in the analytical forms. The relevant expressions of the atomic matrix element for the ejection of an arbitrary shell electron have already been derived in Ref. 16 [See Eqs. (5)$\sim$(9)].

In order to take into account the Coulomb interaction between atomic electrons, the concept of the screening constant is used. The nuclear charge in the atomic matrix elements mentioned above is replaced by an appropriate effective nuclear charge:

$$Z_{\text{eff}} = Z - \sigma , \tag{7}$$

where σ is the screening constant.

Following the method used in the previous works,[4,5,14,16)] the screening constant for the initial state is determined from

$$\sigma = Z(1 - \bar{r}_Z/\bar{r}_{SCF}) , \tag{8}$$

where $\bar{r}_Z$ is the mean relativistic hydrogenic radial distance and $\bar{r}_{SCF}$ is the mean radius obtained from the relativistic self-consistent-field calculations.[17)] For the wave functions in Eq. (6), $\bar{r}_Z$ can be obtained analytically [See Eq. (12) in Ref. 16].

The screening constant for the final continuum wave function is chosen to be the same as that for the bound-state electron to be ejected. This choice is justified by the fact that the dominant contribution to the atomic matrix element comes from the region near the mean radius of the bound electron before ejection. In order to take into account the presence of a vacancy created by electron capture, the screening constant is modified as

$$\sigma_c = (\sigma_h/\sigma_s)\sigma , \tag{9}$$

where σ is the screening constant of the bound electron to be ejected in the daughter atom, and σ_h and σ_S are the Slater screening constants[18)] with and without a vacancy, respectively.

III. RESULTS AND DISCUSSION

We have calculated the M-shell internal ionization probabilities and the ejected electron spectra for K, L_1, and M_1 capture. Four radioactive nuclides, ^{55}Fe, ^{71}Ge, ^{109}Cd, and ^{131}Cs, are considered. The nuclear parameters of these nuclides and binding energies of atomic electrons are taken from the tables prepared by Lederer *et al.*[19] All the calculations in the present work have been performed with the FACOM M-382 computer in the Data Processing Centre of Kyoto University.

In Fig. 1, the ejected electron spectra during K-capture decay of ^{55}Fe are plotted

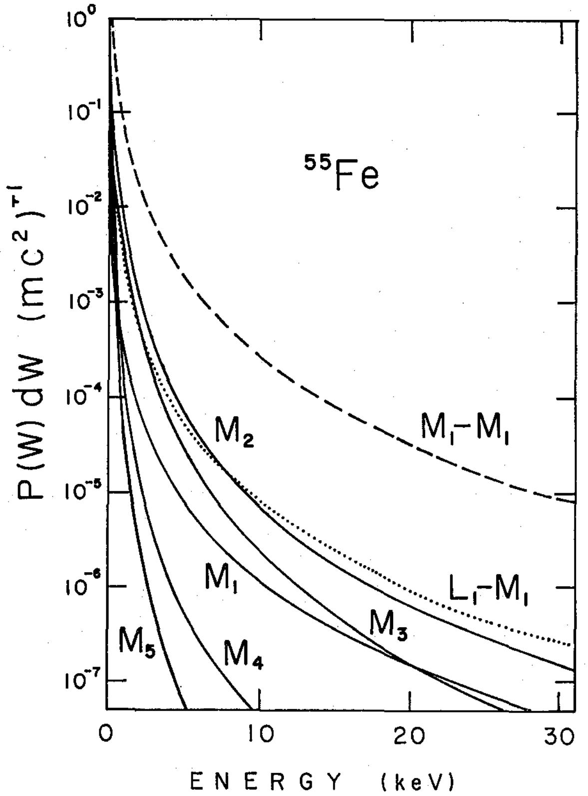

Fig. 1. The ejected electron spectra from M subshells during electron capture decay of ^{55}Fe. The solid curves represent the M_i-shell spectra in K capture. The dotted curve indicates the M_1-shell spectrum in L_1 capture and the dashed curve shows that in M_1 capture.

for various M subshells. In order to demonstrate the difference in spectral shapes, each energy spectrum is normalized per M-subshell electron. The real spectra can be obtained by multiplying the number of electrons in each M subshell. For comparison, the energy spectra of electrons ejected from M_1 shell during L_1 and M_1 capture are also shown.

It can be seen from the figure that the M-shell electron spectra ejected in K-capture decay are concentrated in low-energy region. All the spectra have similar shape at low energies, but the M_i-subshell spectrum with larger i value decreases more rapidly with increasing electron energy. Only exception is the M_1-shell spectrum, which has low intensity in the low-energy region than the M_2- and M_3-shell spectrum. This fact means that for M_1-shell electron the sudden change of nuclear charge in K-capture decay is compensated by the change in atomic potential due to creation of K-shell vacancy and different from the M-subshell ionization probabilities in K conversion,[16] where the change in the potential comes only from the decrease in shielding due to loss of K-shell electron.

For L_1 and M_1 capture, the M_1-shell electron ejection probability is larger than that for K capture in the whole energy region. The probability becomes higher when the captured shell approaches to the shell from which the electron is ejected. The electron ejection probability is sensitive to the loss of electron near to the shell concerned.

In Table I, the M-subshell ionization probabilities per K capture are listed. As has been derived by Migdal,[20] the ionization probability is approximately pro-

Table I. The M-subshell internal ionization probabilities per K capture ($\times 10^{-4}$).

Z	Nuclide	E (keV)	M_1	M_2	M_3	M_4	M_5
26	^{55}Fe	232	0.254	2.64	4.08	1.33	0.418
32	^{71}Ge	235	0.174	1.32	1.44	3.10	3.07
48	^{109}Cd	184	0.0569	0.456	0.518	0.130	0.107
55	^{131}Cs	350	0.126	0.398	0.515	0.193	0.0644

portional to $1/Z_{\rm eff}^2$. However, some values for ^{109}Cd are smaller than those for ^{131}Cs. This can be ascribed to the smallness of the transition energy of the former nuclide. In contrast to the two-step model, such as Crasemann *et al.*,[7] the ionization probability in the present model depends on the transition energy, or more exactly the ratio of sum of the binding energies of the captured and active electrons to the transition energy,

$$(B_K+B_{M_i})/E \fallingdotseq B_K/E\,.$$

The value of B_K/E is 0.145 for ^{109}Cd and 0.103 for ^{131}Cs. When this value is larger, the ionization probability becomes smaller. The small values for the M_4 and M_5 shells in ^{55}Fe comes from the fact that the M_5 shell in Fe is not closed and this leads to reduction of the shielding effect on the M_4 shell.

The M-subshell ionization probabilities per L_1 capture are given in Table II.

Table II. The M-subshell internal ionization probabilities per L_1 capture ($\times 10^{-4}$).

Z	Nuclide	E (keV)	M_1	M_2	M_3	M_4	M_5
26	^{55}Fe	232	0.614	0.687	0.620	1.54	0.376
32	^{71}Ge	235	0.325	0.276	0.574	3.81	3.08
48	^{109}Cd	184	0.159	0.0340	0.0629	0.178	0.140
55	^{131}Cs	350	0.131	0.0347	0.0508	0.124	0.0772

For L_1 capture, the loss of L_1-shell electron strongly affects on $3p$ shell and compensates the change in nuclear charge. Owing to this effect, the M_2- and M_3-electron ejection probabilities decrease. On the other hand, the M_1-shell electron has large radial density near to the nucleus and the change in nuclear charge by electron capture dominates in comparison with the effect of decrease in screening due to the vacancy creation.

The dependence on the transition energy is small for L_1 capture because B_{L_1}/E is small for all elements considered here. The M_4- and M_5-shell electrons feel almost same change in atomic potential as in K capture and their ejection probabilities are nearly equal to the values for K capture.

Table III shows the M-subshell internal ionization probabilities per M_1 capture.

Table III. The M-subshell internal ionization probabilities per M_1 capture ($\times 10^{-3}$).

Z	Nuclide	E (keV)	M_1	M_2	M_3	M_4	M_5
26	^{55}Fe	232	1.14	1.20	1.18	0.201	0.0430
32	^{71}Ge	235	0.901	1.19	1.14	0.400	0.315
48	^{109}Cd	184	0.324	0.393	0.344	0.0188	0.0145
55	^{121}Cs	350	0.251	0.289	0.243	0.0109	0.00792

It is clear from the table that the M_1-, M_2-, and M_3-shell ionization probabilities are about one order of magnitude higher than those in K and L_1 capture. However, the ejection probabilities of M_4- and M_5-shell electrons are almost equal to the values in the case of K- and L_1-shell capture. This can be explained by the fact that the $3d$ electron stays most times outside of the mean radial distance of the $1s$, $2s$, and $3s$ electrons and experiences almost similar change in the potential when the ns electron is lost. The effect of transition energy becomes much more unimportant for M_1-capture decay because of small M-subshell binding energies.

Although there have been reported neither theoretical nor experimental studies on M-shell IIEC, we can compare the present results with the theoretical values of M-electron excitation probabilities in electron capture. In Table IV, comparison of the present results with the values based on the prediction of Carlson *et al.*[21] and with the calculated values of Crasemann *et al.*[7] is shown for ^{71}Ge.

According to Carlson *et al.*[21] the ionization probability of an atomic electron is proportional to the square of the change in effective nuclear charge experienced by that electron during nuclear transition and can be expressed as

$$P = (\Delta Z)^2 P_\beta \,, \tag{10}$$

where ΔZ is the change in the effective nuclear charge and P_β is the ionization probability accomapnying β^- decay, *i.e.* $\Delta Z=1$. This relation has been proved by the present author analytically for K- and L-electron ejection in the nonrelativistic case,[22] and numerically for K-shell internal conversion with relativistic wave function.[16]

Carlson *et al.*[21] calculated the P_β values by the use of the self-consistent-field wave functions.[17] In order to avoid to calculate matrix element with the continuum wave function, they used the two-step model and obtained the expression for the probability of promotion of an electron from an nl state in the parent atom to a higher-bound state or to the continuum state in the daughter atom,

$$P_\beta = 1 - |\langle\psi(Z+1, nl)\,|\,\psi(Z, nl)\rangle|^N - P_F \,, \tag{11}$$

where $\psi(Z,nl)$ is the electron wave function for the nl state with nuclear charge Z, N is the number of electrons in the nl state, and P_F is the transition probability to occupied bound state, which is forbidden by the Pauli principle. Using their P_β values and the effective nuclear charge calculated in the present work, the CA values in Table IV were obtained.

Table IV. Comparison of the IIEC probabilities of ^{71}Ge with the prediction of Carlson *et al.* and with the results of Crasemann *et al.* ($\times 10^{-4}$).

EC[a]	Shell[b]	Present	CA[c]	CR[d]
K	M_1	0.174	0.0239	6.3
	M_2	1.32	2.25	1.1
	M_3	1.44	6.28	
	M_4	3.10	38.9	1.4
	M_5	3.07	60.5	
L_1	M_1	0.325	0.860	25.8
	M_2	0.276	0.0278	40.1
	M_3	0.574	0.251	
	M_4	3.81	38.9	4.4
	M_5	3.08	60.5	
M_1	M_1	9.01	10.0	24.0
	M_2	11.9	19.5	156.6
	M_3	11.4	42.5	
	M_4	4.00	38.9	134.3
	M_5	3.15	60.5	

a) Captured electron.
b) Ejected electron.
c) Prediction of Carlson *et al.*, with P_β value of Ref. 21.
d) Crasemann *et al.* (Ref. 7).

Crasemann *et al.*[7] calculated the electron excitation probabilities during electron capture decay using the same expression as Eq. (11), but took into consideration correctly the change in nuclear charge and presence of a vacancy in the daughter atom due to electron capture. Their wave functions are the nonrelativistic Hartree-Fock ones and the calculations were not extended above $Z=54$. Their results are listed in Table IV as CR.

It is seen from Table IV that for K and L_1 capture the present results are satisfactory agreement with the values of Crasemann *et al.*, except for the case of M_1-shell ejection. The M_1-electron excitation probabilities of Crasemann *et al.* are much larger than the present values. This may be partially ascirbed to the effect of the transition energy. For M_1 capture, the model of Crasemann *et al.* also gives much larger values than the present model. It should be noted, however, that their values include the contributions from the shake-up process to unoccupied bound states. In the case of Ge, the $4p$ shell is an open shell and the $3d$ shell is the outermost d shell. Considering the fact that the shake-off and shake-up processes are the monopole transition, there are large probabilities for the $3p$—$4p$ and $3d$—$4d$ transitions. On the other hand, the contributions of these transitions are not included in the present calculations.

The results based on the prediction of Carlson *et al.* are also calculated in the two-step model and include the contributions from the shake-up process. The values based on this model are strongly dependent on the change in effective nuclear charge and very small values are obtained when ΔZ is small, such as K-M_1, L_1-M_2, and L_1-M_3 cases. Except for these cases, the prediction of Carlson *et al.* gives larger probabilities than the present model. Similar tendency has already been pointed out by us in the case of K- and L-shell internal ionization during K conversion.[16] We found that when we use the P_β values of Carlson *et al.*,[21] the $(\Delta Z)^2 P_\beta$ values are larger than the screened hydrogenic model and this is attributed to the largeness of the P_β values of Carlson *et al.*[21] This can be explained by the fact that in the P_β values of Carlson *et al.* the presence of the vacancy in the daughter atom is not taken into account and there is no cancellation effect between change in nuclear charge and decrease in sihelding. For transitions in which an inner-shell vacancy is produced, such as electron capture, internal conversion, and photoelectric effect, it is inadequate to use the P_β values of Ref. 21 in the prediction Carlson *et al.*

IV. CONCLUSION

The probabilities and ejected electron spectra of M-shell IIEC have been calculated using the screened relativistic hydrogenic wave functions. The effect of phase-space sharing factor between two leptons and existence of the vacancy created in the daughter atom by electron capture are taken into consideration. The present results indicate that the probabilities for the M-shell IIEC is $10^{-6} \sim 10^{-3}$ per K, L_1, and M_1 cpature. The experimental study on this phenomenon would be difficult because of small probability, low M-shell X-ray energies, and presence of high background due to IBEC and inner-shell IIEC.

For the case of M_1-shell IIEC, it is impossible to distinguish experimentally between inner-shell IIEC in M capture and M-shell IIEC in innre-shell capture and the real probability is given as the sum of the probabilities for two processes including the interference effect.[3,4] However, the M_1-shell capture probability is usually less than K- and L_1-shell capture probabilities. In the present work, the effect of indistinguishability of these processes is neglected and the IIEC probabilities are calculated separately.

The calculated values are in satisfactory agreement with the values of Crasemann *et al.*, except for the case of M_1-electron ejection, but different from the values based on the prediction of Carlson *et al.* The discrepancy in the latter case comes from neglection of presence of the vacancy in the daughter atom in their transition probability.

It is interesting to estimtae the M-shell IIEC probabilities for nuclides with low transition energy, where K- and L-shell IIEC is energetically surpressed. The experimental studies may be possible for these nuclides because the contributions of the background due to the inner-shell IIEC are missing. However, in general, the nuclides with low transition energy are high-Z elements and the IIEC probabilities for such elements are very small.

The most interesting nuclide with low transition energy is ^{163}Ho. As has been described already, this nuclide is the most favorable candidate to measure the rest mass of electron neutrino. Recently we have determined the transition energy to be 2.56 keV.[23] This result means that K- and L-shell electron capture decay is impossible and M-shell IIEC in M capture is also forbidden energetically. It is needed to calculate N- and O-shell IIEC probabilities in M- and higher-shell electron capture. For this nuclide, the calculation should be in the one-step approach, because the transition energy is so small that the effect of phase-space sharing plays an important role.

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