Double K-shell ionization accompanying the internal conversion of the 0.145-MeV transition in ^{141}Pr

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Double ionization of the atomic K shell accompanying the K-shell internal conversion of the 0.145-MeV transition of ^{141}Pr has been studied by recording coincidences between $K\alpha$ satellite x rays and $K\alpha$ hypersatellite x rays emitted when the double vacancies are filled. The probability per K-shell internal conversion that a double vacancy is formed, P_{KK}(IC), was found to be $(3.4\pm0.3)\times10^{-5}$, which is a factor of (0.79±0.07) times the theoretical prediction for K-shell electron shakeoff in K-shell internal conversion of Mukoyama and Shimizu. The $K\alpha_1$ to $K\alpha_2$ hypersatellite ratio was found to be (1.50±0.13), a value which is about 6% less than the calculation of Åberg *et al.*

I. INTRODUCTION

In K-shell internal conversion, double ionization of the atomic K shell may occur when the unconverted K electron is either excited to an unoccupied bound state or completely ejected from the atom. This excitation or ejection can be produced by several separate processes. One process is a shakeup (SU) or shakeoff (SO) process similar to that found in electron capture or beta decay and is caused by the sudden disappearance of the electron-electron Coulomb interaction. A second process is the direct collision between the converted and unconverted K electrons. Other processes which can give rise to a doubly ionized K shell are higher-order electromagnetic transitions, both nuclear and "electronic" in nature. In the nuclear higher-order transitions, the decay proceeds through virtual intermediate levels with the internal conversion of both transitions. In the electronic decay mode the nucleus may transfer its energy to an orbital electron, which radiates a photon as it is ejected from the atom. This effect is commonly called the internal Compton effect. Double ionization of the K shell is then produced when the internal Compton photon itself is internally converted.

All of the above processes leave the atom in the same final state, characterized by a doubly ionized K shell. In atoms with large fluorescence yields the most probable mode of atomic deexcitation is the emission of two K x rays. An experimental determination of the probability per K internal conversion (IC) for double K-shell ionization, P_{KK}(IC), can then be made by coincidence measurements between the K x rays emitted when the double vacancy is filled. These x rays, a K hypersatellite x ray (K^H; $1s^{-2}\rightarrow1s^{-1}2p^{-1}$) and a secondary K satellite x ray (K^S; $1s^{-1}2p^{-1}\rightarrow2p^{-2}$) are shifted to higher energy with respect to the normal K x rays, with the K^S x ray shifted only about 10% of the shift of the K^H x ray.

Double K-shell ionization in internal conversion has been experimentally examined in ten isotopes, with approximately one-half of the measurements yielding only an upper limit to P_{KK} (IC). Of these ten isotopes, five have been studied by several groups. Table I shows the results of the measurements performed on these five isotopes, the majority of which have been performed since 1969. A summary of earlier work has been published by Freedman.[12] As seen in Table I, discrepancies exist in the measured values of P_{KK}(IC) in four of the five isotopes. Only in the case of ^{109}Ag (Refs. 1 and 2) is there agreement in the P_{KK}(IC) values. Three other isotopes have also been studied by single groups during this time. Porter *et al.*[8] obtained a P_{KK}(IC) value of $(4-20)\times10^{-5}$ for the 0.122-MeV transition in ^{57}Fe by direct observation of the satellite line in the electron spectrum, Briand *et al.*[4] obtained a value of $\simeq10\times10^{-5}$ for the 0.145-MeV transi-

TABLE I. Previously measured values of P_{KK}(IC) in five isotopes.

Isotope	P_{KK}(IC) ($\times10^5$)	Method[a]	Ref.
^{109}Ag	15.3±2.4	*A*	1
	13.0±1.1	*A*	2
^{14}In	1.7±0.3	*B*	3
	12	*A*	4
^{131}Xe	11±2	*A*	5
	<4	*A*	6
^{137}Ba	18±5[b]	*C*	7
	<20	*D*	8
	7.1±3.5	*A*	9
	<5	*A*	10
^{203}Tl	4.0±1.5	*A*	11
	25	*A*	4

[a]*A* denotes x-ray—x-ray coincidence, *B* denotes e$^-$-e$^-$ coincidence, *C* denotes e$^-$-e$^-$—x-ray coincidence, *D* denotes direct observation of satellite in the e$^-$ spectrum.
[b]Energy range 115—472 keV.

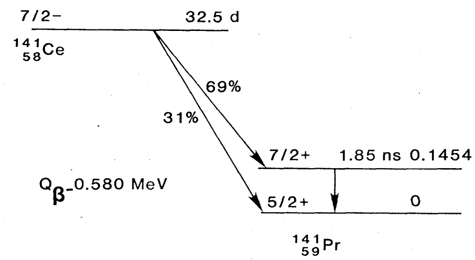

FIG. 1. Decay scheme of ^{141}Ce.

tion of ^{141}Pr by the x-ray—x-ray coincidence method, and Javahery *et al.*[13] obtained a combined value of $(7.63\pm3.8)\times10^{-5}$ for all the transitions in ^{169}Tm by direct observation of the $K\alpha_1^H$ x ray with a curved-crystal spectrometer.

In the present work double ionization of the K shell in the 0.145-MeV transition of ^{141}Pr has been reexamined by the x-ray—x-ray coincidence method to help give a better data base for comparing experiment and theory. Figure 1 shows the decay scheme of ^{141}Ce to the 0.145-MeV level of ^{141}Pr. While K-shell electron SO and SU also occurs in the beta decay of the ^{141}Ce, the x rays produced in this process would be normal K x rays, and not the hypersatellite and satellite x rays produced in the filling of the double K-shell vacancy.

II. EXPERIMENTAL PROCEDURES

A. Electronic circuitry and detectors

The value for $P_{KK}(\mathrm{IC})$ in this study was deduced from the coincidences recorded between the Pr $K\alpha^H$ and $K\alpha^S$ x rays ($K\alpha^H K\alpha^S$ coincidences). The schematic diagram of detector and circuits used for these measurements is shown in Fig. 2. This arrangement is very similar to that used in recent investigations of K-shell electron SO and SU in electron capture.[14–16] The main difference between this and the earlier investigations is associated with the detectors used for the studies. In this investigation two intrinsic Ge low-energy photon spectrometers were used to detect the $K\alpha^H$ and $K\alpha^S$ x rays. The detectors used were Ge(1), a Princeton Gamma-Tech model IG-510 with a 25 mm diam, 10 mm sensitive depth, 0.152 mm Be window, 3 mm window to detector distance, and peak width of 550 eV at 122 keV, and Ge(2); an Ortec model GLP-16195/10 with a 16 mm diam, 13 mm sensitive depth, 0.127 mm Be window, 5 mm window to detector distance, and peak width of 179 eV at 5.9 keV. While both $K\alpha^H$ and $K\alpha^S$ x rays were detected in both detectors, data analysis was performed on Pr $K\alpha^H$ x rays recorded in detector Ge(2). Hence as far as data analysis was concerned, Pr $K\alpha^S$ x rays were detected in Ge(1).

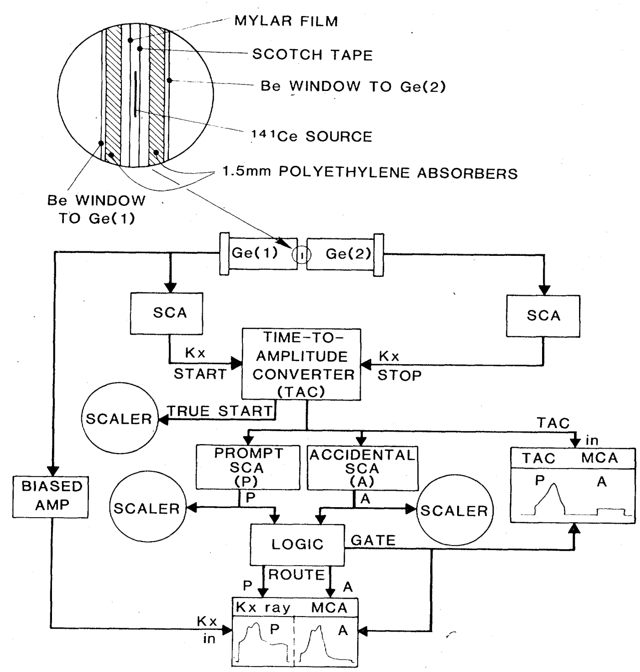

FIG. 2. Schematic diagram of detectors and circuits. Preamplifiers, amplifiers, and delay circuits have been omitted for simplicity.

B. ^{141}Ce source

The ^{141}Ce activity used in this study was purchased from the New England Nuclear Corporation. During the course of the investigation three separate runs, whose durations were 252.8, 308.9, and 161.6 h, were made using a single source. The activity at the beginning of each run was 1.38, 1.04, and 0.74 μCi, respectively. The source was prepared by evaporating drops of active solution onto a 0.07 mm thick Mylar backing. After drying, the source was covered with Scotch brand tape and placed between two 1.5 mm thick polyethylene absorbers. The source was then placed between the two detectors in a close geometry.

A long run with a well-shielded Ge detector at the Missouri University Research Reactor (MURR) facility showed the only source impurity to be 137.2 d ^{139}Ce, with an activity of about 0.15 per cent of the ^{141}Ce activity at the beginning of the investigation. While this activity produced La x rays in the prompt coincidence spectra these x rays were easily resolved from the Pr $K\alpha$ and $K\alpha^H$ x rays in detector Ge(2).

III. DATA ANALYSIS

The total number of $K\alpha^H K\alpha^S$ coincidences recorded during a run, $N(K\alpha^H K\alpha^S)$, is given by

$$N(K\alpha^H K\alpha^S)=N_0 f(0.145)\alpha_K(0.145)P_{KK}(\mathrm{IC})\omega_K^S(K\alpha/K_T)^S(aE)^S_{\mathrm{Ge(1)}}\omega_K^H(K\alpha/K_T)^H(aE)^H_{\mathrm{Ge(2)}}E_T\ , \tag{1}$$

where N_0 is the total number of decays of ^{141}Ce during a run, $f(0.145)=0.69$ (Ref. 17) is the fraction of ^{141}Ce decays which go to the 0.145-MeV level of ^{141}Pr, $\alpha_K(0.145)=0.26$ (Ref. 17) is the K-shell internal conversion probability for the 0.145-MeV transition, $P_{KK}(\mathrm{IC})$ is the probability per K-internal conversion for double K-shell ionization, ω_K^S and ω_K^H are the K-shell fluorescence yields for Pr, K^S and K^H x rays, respectively,

$(aE)^S_{\mathrm{Ge}(1)}$ and $(aE)^H_{\mathrm{Ge}(2)}$ are the products of absorption factors (a) and photopeak detection efficiencies (E) for Pr $K\alpha^S$ and $K\alpha^H$ x rays in the two intrinsic Ge detectors, and E_T is the time-to-amplitude converter (TAC), coincidence efficiency. The total number of Pr $K\alpha$ x rays detected in Ge(1) during a run, $N(K\alpha)$, is given by

$$N(K\alpha)=N_0 f(0.145)\alpha_K(0.145)\omega_K(K\alpha/K_T)(aE)_{\mathrm{Ge}(1)}\ , \quad (2)$$

where ω_K is the K-shell fluorescence yield for normal Pr K x rays, $(K\alpha/K_T)$ is the $K\alpha$ fraction of all Pr K x rays and $(aE)_{\mathrm{Ge}(1)}$ is the product of the absorption factor and total detection efficiency for normal $K\alpha$ x rays in detector Ge(1). The very small contributions from the $K\alpha$ x rays emitted in K-shell SO and SU in the beta decay of ^{141}Ce and the $K\alpha^S$ and $K\alpha^H$ x rays emitted in the internal conversion of the 0.145-MeV transition have been neglected in Eq. (2). Taking $\omega_K^S=\omega_K^H=\omega_K$, where $\omega_K=0.915$,[17] $(K\alpha/K_T)^S=(K\alpha/K_T)$, $(aE)^S_{\mathrm{Ge}(1)}=(aE)_{\mathrm{Ge}(1)}$, $(aE)^H_{\mathrm{Ge}(2)}=(aE)_{\mathrm{Ge}(2)}$, and substituting Eq. (2) into Eq. (1) gives

$$N(K\alpha^H K\alpha^S)=N(K\alpha)P_{KK}(\mathrm{IC})\omega_K(K\alpha/K_T)^H \times(aE)_{\mathrm{Ge}(2)}E_T\ . \quad (3)$$

Rearranging Eq. (3) then yields

$$P_{KK}(\mathrm{IC})=\frac{N(K\alpha^H K\alpha^S)}{N(K\alpha)\omega_K(K\alpha/K_T)^H(aE)_{\mathrm{Ge}(2)}E_T}\ . \quad (4)$$

The $(aE)_{\mathrm{Ge}(2)}$ term of Eq. (4) was determined by singles spectra measurements of the $K\alpha$ x ray count using the known decay rates of the ^{141}Ce source. For each of the $K\alpha^H K\alpha^S$ runs, a singles measurement was taken at both the start and end of the run. The average of the two values of $(aE)_{\mathrm{Ge}(2)}$ acquired in these single runs was then used in Eq. (4). These two values for any one run were consistent to less than 2%.

For normal Pr K x rays $(K\alpha/K_T)$ is 0.803.[17] Åberg *et al.*[18] have pointed out, however, that the ratio of $K\alpha_1^H$ to $K\alpha_2^H$ x rays decreases with respect to the $K\alpha_1$ to $K\alpha_2$ x-ray ratio. For normal Pr K x rays this ratio is 1.82,[17] whereas it is about 1.6 (Ref. 18) for hypersatellite x rays. Assuming that only the $K\alpha_1^H$ transition is suppressed, $(K\alpha/K_T)^H$ can be calculated on the basis of the observed $(K\alpha_1/K\alpha_2)^H$ ratio to be

$$(K\alpha/K_T)^H=\frac{(K\alpha_1/K\alpha_2)^H I(K\alpha_2)+I(K\alpha_2)}{(K\alpha_1/K\alpha_2)^H I(K\alpha_2)+I(K\alpha_2)+I(K\beta_1')+I(K\beta_2')}\ , \quad (5)$$

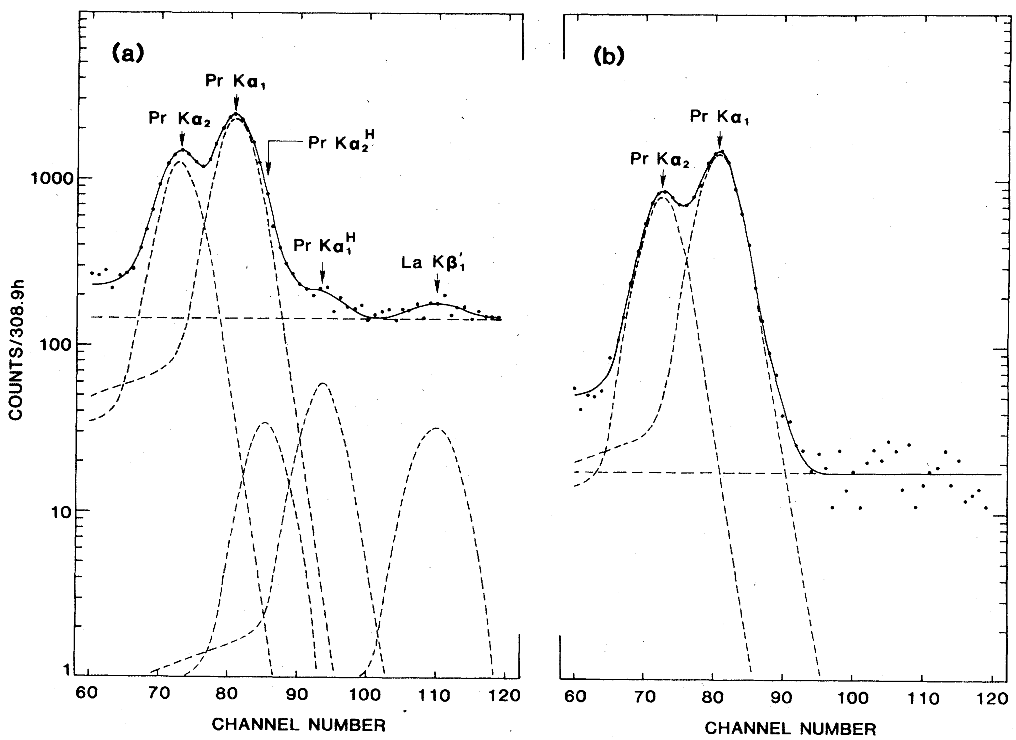

FIG. 3. (a) Prompt coincidence spectrum from the Pr $K\alpha_2$ x ray to La $K\beta_1'$ x-ray region recorded in Ge(2). The solid curve gives the overall computer fit to the data, whereas the dashed curves give the components of the fit, the Pr $K\alpha_2$, $K\alpha_1$, $K\alpha_2^H$, and $K\alpha_1^H$ x rays, the La $K\beta_1'$ x ray, and the constant continuum. (b) Accidental coincidence spectrum for the same region.

where the relative K x ray intensities, $I(K\alpha_2)$, $I(K\beta_1')$, and $I(K\beta_2')$ are taken to be 54.8, 29.9, and 7.9,[17] respectively.

The $N(K\alpha^H K\alpha^S)$ term of Eq. (4) was determined from least-squares computer fits to the $K\alpha_2 - K\alpha_1^H$ region of the prompt coincidence spectra recorded in Ge(2). Each prompt spectrum was fit in this region with the sum of four modified Gaussian distributions, one each representing the Pr $K\alpha_2$, $K\alpha_1$, $K\alpha_2^H$, and $K\alpha_1^H$ x rays, and a constant continuum. The modified Gaussian distributions used had the form suggested by Jorch and Campbell.[19] In the fitting procedure the energies of the $K\alpha_1^H$ and $K\alpha_2^H$ x rays were constrained to values predicted by Chen, Crasemann, and Mark[20] with the $K\alpha_1^H$ x ray located at an energy of 717 eV above the $K\alpha_2$ x ray and the $K\alpha_1^H$ x ray located 726 eV above the $K\alpha_1$ x ray. The shapes of all four of the modified Gaussian distributions were taken to be the same.

$N(K\alpha)$ was determined from the TAC true starts scalar with a 2% background reduction due principally to the external bremsstrahlung photons produced in the absorption of both the internal conversion electrons and the 0.436-MeV beta rays as well as Compton photons from the 0.145-MeV gamma rays. E_T was determined from TAC spectra recorded simultaneously during the course of each run and was equal to 0.95 ± 0.05 for all three runs.

IV. RESULTS

Figure 3(a) shows the region of the gamma-ray spectrum from the Pr $K\alpha_2$ x ray to the La $K\beta_1'$ x ray recorded in Ge(2) in prompt coincidence with the Pr $K\alpha$ x rays selected by Ge(1). The solid curve indicates the overall computer fit to the region, whereas the dashed curves indicate the components of the fit, the Pr $K\alpha_2$, $K\alpha_1$, $K\alpha_2^H$, and $K\alpha_1^H$ x rays, the La $K\beta_1'$ x ray, and the constant continuum. Figure 3(b) shows the accidental coincidence spectrum for the same region. The La $K\beta_1'$ x ray seen in Fig. 3(a) is due to the ^{139}Ce contaminant discussed earlier. The large continuum present in the prompt spectrum is due to external bremsstrahlung photons from the absorption of both the internal conversion electrons and the 0.436-MeV beta rays in Ge(2) in coincidence with Pr $K\alpha$ x rays in Ge(1). The large number of true Pr $K\alpha$ x rays seen in this spectrum are primarily due to the reverse process, external bremsstrahlung photons in Ge(1) in coincidence with Pr $K\alpha$ x rays in Ge(2). However, some of these x rays would be Pr $K\alpha^S$ x rays in coincidence with Pr $K\alpha^H$ x rays in Ge(1). Also, some Pr $K\alpha$ x rays would be due to the SO and SU effect in the beta decay of ^{141}Ce which are in coincidence with external bremsstrahlung photons detected in Ge(1). These x rays would be normal Pr x rays and not hypersatellite x rays. If, however, K-shell internal conversion of the 0.145-MeV transition occurred before a K-shell vacancy produced by the above beta decay processes to this level were filled, hypersatellite x rays could be produced. Since the K-vacancy lifetime in Pr is about 4×10^{-17} s (Ref. 20), the 1.85×10^{-9} s half-life for the 0.145-MeV level noted in Fig. 1 makes any contributions to the hypersatellite peaks due to this effect completely negligible compared to the measured P_{KK}(IC) value.

Table II shows the data necessary for evaluation of P_{KK}(IC) for each run. $N(K\alpha_2^H)$ and $N(K\alpha_1^H)$ represent the total number of $K\alpha_2^H$ and $K\alpha_1^H$ x rays detected during a run, respectively. The uncertainties on the individual $(K\alpha_1/K\alpha_2)^H$ and $(K\alpha/K_T)^H$ ratios are based upon the uncertainties associated with the individual $N(K\alpha_2^H)$ and $N(K\alpha_1^H)$ values. The uncertainties on the P_{KK}(IC) values are based upon the uncertainties associated with $N(K\alpha^H K\alpha^S)$, $(K\alpha/K_T)^H$, $(aE)_{\mathrm{Ge(2)}}$, and E_T. The three P_{KK}(IC) values of Table II give an unweighted average of $(3.4\pm0.3)\times10^{-5}$. The uncertainty on this average P_{KK}(IC) value is the average value of the uncertainties of the individual P_{KK}(IC) values and has not been statistically reduced because of systematic errors which may have existed in computer fitting procedures and detection efficiencies. This average value is approximately a factor of 3 less than an earlier measurement by Briand *et al.*[4]

When the energies of the Pr $K\alpha_1^H$ and $K\alpha_2^H$ x rays were also obtained from the computer fitting procedures, the values for P_{KK}(IC) changed by as much as 10%. While the energy shifts found in this manner for the $K\alpha_1^H$ hypersatellite were in good agreement (±5%) with the predictions of Chen, Crasemann, and Mark, an average 20% deviation found for the $K\alpha_2^H$ shift due to the presence of the intense Pr $K\alpha_1$ peak led to the procedure used whereby the energies of the hypersatellites were fixed at the theoretical values to give the best value for P_{KK}(IC). This procedure was justified by the good agreement found between experiment and theory for these energy shifts in similar investigations.

The average value of $(K\alpha_1/K\alpha_2)^H$ found from the three individual values is (1.50 ± 0.13). The uncertainty on this value is also the average value of the three individual un-

TABLE II. Data necessary to calculate $(K\alpha/K_T)^H$ from Eq. (5) and P_{KK}(IC) from Eq. (4).

Run	I	II	III
Time (h)	252.8	308.9	161.6
$N(K\alpha_2^H)$	247±16	240±15	130±11
$N(K\alpha_1^H)$	375±19	404±20	169±13
$N(K\alpha^H K\alpha^S)$	622±25	644±25	299±17
$N(K\alpha)$ $(\times10^{-6})$	371	334	130
$(aE)_{\mathrm{Ge(2)}}$ $(\times10^{2})$	7.9±0.5	8.7±0.5	8.8±0.5
$(K\alpha_1/K\alpha_2)^H$	1.52±0.12	1.68±0.14	1.30±0.15
$(K\alpha/K_T)^H$	0.79±0.01	0.80±0.01	0.77±0.01
P_{KK}(IC) $(\times10^{5})$	3.1±0.3	3.2±0.3	3.9±0.4

TABLE III. Summary of experimental results and theoretical calculations for P_{KK}(IC) in the 0.145-MeV transition of ^{141}Pr.

Experimental P_{KK}(IC) ($\times 10^5$)		Theoretical P_{KK}(IC) ($\times 10^5$)[b]		
This work	Previous work[a]	P_{KK}(SO)	P_{KK}(DC)	P_{KK}(ICE)
3.4±0.3	≃10	5.74[c]	2.35[c]	23
		4.32[d]	1.77[d]	

[a]From experimental result in Ref. 4.
[b]SO, DC, and ICE refer to shakeofff, direct collision, and internal conversion of the internal Compton radiation, respectively. The manner in which these values were calculated is discussed and referenced in the text.
[c]Two-step method.
[d]One-step method.

certainties, unreduced as in the case of the average P_{KK}(IC) value. This average value is about 6% lower than predicted by Åberg *et al.*[18]

V. DISCUSSION

Of the four processes which can give rise to double K-shell vacancies in internal conversion, shakeoff or shakeup (SO or SU), direct collision (DC), double conversion or intermediate nuclear state (N), and internal conversion of the internal Compton effect (ICE), theoretical comparisons can be made for all but process N. Although Eichler and Jacob,[21] Eichler,[22] and Grechukhin[23] have presented formal theories for two quantum transition intensities, it is very difficult to determine the probability for double conversion from either theory since all the virtual intermediate states which may contribute to the process are not known. Hence, no comparison is made between the experimental P_{KK}(IC) and a theoretical P_{KK}(N).

Mukoyama and Shimizu[24,25] have calculated the probability of K-shell electron SO in internal conversion, but not SU, by use of one- and two-step relativistic overlap theories. They found P_{KK}(SO) to be 5.74×10^{-5} by the two-step and 4.32×10^{-5} for the one-step method. The experimental P_{KK}(IC) value is a factor of (0.79±0.07) times this latter P_{KK}(SO) value. Any contribution to P_{KK}(SO) by a SU process, however, would increase this discrepancy.

No formal theory for a direct collision process in internal conversion has been presented, but Feinberg[26] has estimated the relative probability of direct collision to shakeoff in beta decay to be BE/E_0, where BE is the binding energy of the electron subsequently ejected and E_0 is the kinetic energy of the colliding electron. If this estimate is applied to the K-shell internal conversion of the 0.145-MeV transition in ^{141}Pr, where the ratio of BE/E_0 is 0.41, the above SO predictions yield P_{KK}(DC) values of 2.35×10^{-5} for the two-step method and 1.77×10^{-5} for the one-step method. This latter value is about a factor of 2 less than the experimental P_{KK}(IC) value.

Listengarten[27] has proposed that the probability for the internal conversion of the internal Compton radiation be given by

$$P_{KK}(\mathrm{ICE})=\left[\frac{4\alpha}{3\pi}\right]W\int_{\mathrm{BE}}^{W-\mathrm{BE}}\frac{\alpha(E1,E)}{2}\frac{dE}{E}\,, \qquad (6)$$

where W is the energy of the nuclear transition, BE is the K-shell binding energy, and $\alpha(E1,E)$ is the K-shell internal conversion coefficient for electric dipole radiation of energy E. Using $\alpha(E1,E)$ as given by Hager and Seltzer,[28] evaluation of Eq. (6) for the 0.145-MeV transition of ^{141}Pr gives a P_{KK}(ICE) value of 23×10^{-5}. This theoretical estimate is about a factor of 6 larger than the experimental P_{KK}(IC) value. It is worthwhile to note, however, that the Listengarten suggestion has generally yielded values 3 to 5 times larger than experimental values.[1,6]

Table III summarizes experimental and theoretical results for P_{KK}(IC) in the 0.145-MeV transition of ^{141}Pr. While the P_{KK}(IC) value determined in this investigation is a combination of all processes producing double K-shell vacancies, the measured value of P_{KK}(IC) is in fairly good agreement with the P_{KK}(SO) value of the one-step method of Mukoyama and Shimizu. To better understand the various processes occurring in the decay, more values of P_{KK}(IC) with experimental spectrum shape studies of the ejected electrons are needed along with more formal theoretical calculations.

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