Double K-shell ionization accompanying internal conversion of the 0.662-MeV transition in $^{137}Ba^{m}$

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Double ionization of the atomic K shell accompanying K-shell internal conversion of the 0.662-MeV transition of $^{137}Ba^{m}$ has been studied by recording coincidences between $K\alpha$ hypersatellite x rays and $K\alpha$ satellite 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 $(10.0\pm0.9)\times10^{-5}$ which is in general agreement with less precise earlier experiments, but is a factor of 2.7 ± 0.3 larger than the relativistic, one-step theory of Mukoyama and Shimizu.

I. INTRODUCTION

The principal mechanism for double K-shell ionization accompanying internal conversion is the shakeoff (SO) or shakeup (SU) process which is similar to that found in beta and electron capture decays. It is described in the sudden approximation as resulting from the sudden change in the central atomic charge. A second mechanism is the direct collision (DC) between the converted and unconverted K electrons. Other mechanisms which produce a vacant K-shell are higher-order electromagnetic transitions which can be either nuclear or electronic in nature. In the nuclear case (NC), the decay proceeds through virtual intermediate levels with internal conversion of both transitions. In the electronic case, the nucleus transfers its energy to an orbital electron which radiates a photon as it is ejected from the atom. This effect is called the internal Compton effect (ICE) and double ionization of the K-shell occurs when the internal Compton photon is internally converted. A review article by Freedman[1] gives an excellent overview of shakeoff phenomena.

All of the above processes leave the atom in the same final state, characterized by an empty K shell, where the most common mode of deexcitation for higher Z atoms is the emission of two K x rays. The probability per K internal conversion for double K-shell ionization, P_{KK}(IC), can then be determined experimentally by recording coincidences between the K x rays emitted when the double vacancy is filled. These x rays, a K hypersatellite (K^{H}; $1s^{-2}\rightarrow 1s^{-1}2p^{-1}$) and a secondary K satellite (K^{S}; $1s^{-1}2p^{-1}\rightarrow 2p^{-2}$), are shifted to higher energies with respect to normal K x rays, with the K^{H} shift being about ten times larger than the shift of the K^{S} x ray and leading to a distinct peak in the coincidence spectrum.

In 1985 a summary of experimental investigations of P_{KK}(IC) were presented[2] along with measurements on the 0.145-MeV transition in ^{141}Pr. This probability was found to be a factor of 0.79 ± 0.07 times the theoretical prediction of Mukoyama and Shimizu[3] (MS). A subsequent measurement[4] for the 0.166-MeV transition in ^{139}La gave a value of 1.2 ± 0.3 times the MS prediction. As noted in the 1985 summary, previous measurements of P_{KK}(IC) for the 0.622-MeV transition in $^{137}Ba^{m}$ gave values of $(18\pm5)\times10^{-5}$ (Ref. 5), $<20\times10^{-5}$ (Ref. 6), $(7.1\pm3.5)\times10^{-5}$ (Ref. 7), and $<5\times10^{-5}$ (Ref. 8) compared with the MS theoretical value of 3.76×10^{-5} (Ref. 3). It was thus in this context of fair agreement between experiment and theory for recent experiments and the rather diverse results for $^{137}Ba^{m}$ that the present investigation was undertaken.

II. EXPERIMENTAL PROCEDURES

A. Electronic circuitry and detectors

The schematic diagram of detectors and circuits used in this investigation is shown in Fig. 1. It is essentially identical to the arrangement used in the earlier work on ^{141}Pr[2]. The Ge(1) detector was 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 (full width at half maximum) at 179 eV at 5.9 keV. A Princeton Gamma-Tech model 1G-510 with 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 was the Ge(2) detector. While both $K\alpha^{H}$ and $K\alpha^{S}$ x rays were detected in both detectors, data analyses were performed on the Ba $K\alpha^{H}$ x rays recorded in Ge(1). The prompt single channel analyzer [SCA(P)] window on the time-to-amplitude converter (TAC) output was set at a nominal 400 ns which corresponded to a coincidence efficiency of 0.98 ± 0.02 as determined from an ungated TAC spectrum. For the initial run, an Ortec 7000 series Si(Li) detector with 4-mm diam, 4.2-mm sensitive depth, 0.008-mm Be window, 10-mm window to detector distance, and peak width of

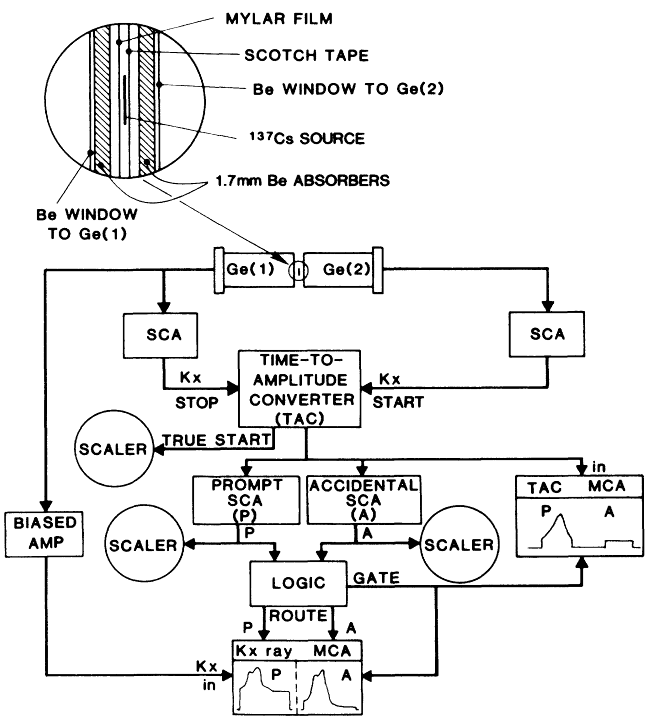

FIG. 1. Schematic diagram of detector and circuits. Preamplifiers, amplifiers, and delay circuits have been omitted for clarity.

160 keV at 5.9 keV, replaced the Ge(2) detector but was used in the Ge(1) position in the circuit.

B. ^{137}Cs source

The ^{137}Cs source material used in this study was purchased from New England Nuclear Corporation as a carrier free fission product. Active solution was evaporated onto a Mylar backing, covered with Scotch brand tape, and measured to be 1.9 ± 0.1 μCi with a well calibrated detector system at the Missouri University Research Reactor facility. No source impurities were observed.

III. DATA ANALYSIS

The decay of 30.2 yr ^{137}Cs is well known and the relatively large K-shell internal conversion coefficient for the 0.662-MeV, $M4$ transition of 0.0916[9] is due to its high multipolarity. Outside of accidental coincidences, the only Ba K x rays which can be in coincidence with each other in this decay are the K^H and K^S x rays described above as the double vacancy is filled. The 2.55-min half-life of the 0.662-MeV level in ^{137}Ba^m eliminates coincidences between K x rays produced by SO+SU accompanying the beta decay to this level and those from simple K-shell internal conversion of the ensuing 0.662-MeV transition.

The number of $K\alpha^H - K\alpha^S$ coincidences recorded in detector Ge(1) during a run, $N(K\alpha^H K\alpha^S)$, is given by

$$\begin{aligned} N(K\alpha^H K\alpha^S) = {} & N_0 f(0.662)\alpha_K(0.662)P_{KK}(\mathrm{IC}) \\ & \times \omega_K^H (K\alpha/K_T)^H (aE)^H_{\mathrm{Ge}(1)} \\ & \times \omega_K^S (K\alpha/K_T)^S (aE)_{\mathrm{Ge}(2)} E_T \;, \end{aligned} \quad (1)$$

where N_0 is the total number of ^{137}Cs decays, $f(0.662)=0.946$[9] is the fraction of decays which go through the 0.662-MeV level, $\alpha_K(0.662)=0.082$[9] is the K-shell internal conversion probability, ω_K^H and ω_K^S are the K-shell fluorescence yields and $(K\alpha/K_T)^H$ and $(K\alpha/K_T)^S$ are the $K\alpha$ to total ratios for Ba K^H and K^S x rays, respectively, $(aE)^H$ and $(aE)^S$ are the absorption factors (a) and photopeak efficiencies (E) for the $K\alpha^H$ and $K\alpha^S$ x rays in the two detectors, and E_T is coincidence efficiency discussed above. The total number of $K\alpha$ x rays detected in Ge(2) during a run, $N(K\alpha)$, is given by

$$N(K\alpha)=N_0 f(0.662)\alpha_K(0.662)\omega_K(K\alpha/K_T)(aE)_{\mathrm{Ge}(2)} \;, \quad (2)$$

where the last three factors refer to normal $K\alpha$ x rays. The very small contributions $(\sim 10^{-4})$ from the various shakeoff mechanisms have been neglected in Eq. (2). Taking $\omega_K^S=\omega_K^H=\omega_K$, where $\omega_K=0.901$[9], $(K\alpha/K_T)^S = (K\alpha/K_T)$, $(aE)^S_{\mathrm{Ge}(2)}=(aE)_{\mathrm{Ge}(2)}$, and substituting in Eq. (1) gives upon rearranging

$$P_{KK}(\mathrm{IC})=\frac{N(K\alpha^H K\alpha^S)}{N(K\alpha)\omega_K(K\alpha/K_T)^H(aE)^H_{\mathrm{Ge}(1)}E_T} \;. \quad (3)$$

The $(aE)^H_{\mathrm{Ge}(1)}$ factor of Eq. (3), taken to be equal to $(aE)_{\mathrm{Ge}(1)}$ for normal Ba $K\alpha$ x rays, was determined from averages of single measurements [using Eq. (2) for Ge(1)] at the beginning and end of each run. These two values for each run were consistent to less than the 5% uncertainty estimated for the source strength.

For normal Ba K x rays, $(K\alpha/K_T)$ is 0.808.[9] Åberg *et al.*[10] have pointed out, however, that the ratio of $K\alpha_1^H$ to $K\alpha_2^H$ is less than $K\alpha_1$ to $K\alpha_2$. For normal Ba K x rays this ratio is 1.82,[9] whereas it is about 1.6 (Ref. 10) for hypersatellite x rays. Assuming that only the $K\alpha_1^H$ is suppressed leads to a $(K\alpha/K_T)^H$ value to 0.79.

In x-ray shakeoff measurements for beta decay, where only one K-shell electron is ejected and the resulting x ray has the normal energy, accidental coincidences must be carefully subtracted from the prompt coincidence spectra to obtain shakeoff probabilities. In double ionization measurements, however, where K^H x rays are observed, which are completely negligible in the singles and hence accidental coincidence spectra, the procedure is to directly determine the number of hypersatellite x-ray coincidences recorded during a run in the most reliable manner. Here the $N(K\alpha^H K\alpha^S)$ factor of Eq. (3) was determined by least-squares computer fits to the $K\alpha - K\alpha^H$ region of the prompt coincidence spectra recorded with the Ge(1) detector using four Voigt profiles modified with low energy tails, as suggested by Gunnick,[11] and a constant continuum. The higher-energy Lorentzian tailing above the peaks is caused by the short lifetime of the atomic states.[12] The same shape was used

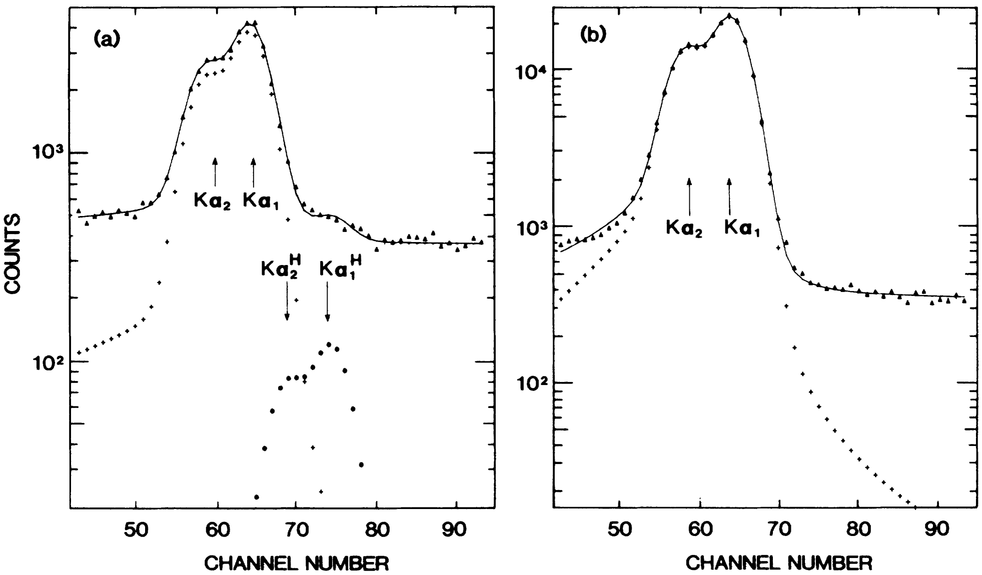

FIG. 2. (a) Prompt coincidence spectrum of the Ba $K\alpha - K\alpha^H$ x-ray region recorded in the Ge(1) detector for run 2. (b) Composite or time-averaged singles spectrum recorded in Ge(1). The solid curves indicate the overall least-squares fit to the data, whereas the pluses and circles give the $K\alpha$ and $K\alpha^H$ components of the fit and the dots give the continua.

for all four of the profiles and was determined from a composite singles spectrum produced by summing singles spectra recorded periodically throughout each run to give a time-averaged spectrum.

In the fitting procedure[11,13] 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[14] at 675 and 669 eV, respectively, above the normal x-ray energies, and the $K\alpha_1^H/K\alpha_2^H$ ratio was set at the value of 1.6 discussed earlier. Since the number of normal $K\alpha$ x rays recorded in the coincidence spectra was some 30 times the number of $K\alpha^H$ and hence $K\alpha^S$ x rays recorded, the very slight perturbation to the position and shape of the normal $K\alpha$ x-ray peaks caused by the $K\alpha^S$ x rays was neglected.

$N(K\alpha)$ was determined from the TAC true starts scalar using a 5% reduction to account for the Compton continuum of the 0.662-MeV gamma rays as well as the external bremsstrahlung caused by adsorption of both beta rays and internal conversion electrons.

IV. RESULTS

Figure 2 shows the (a) prompt and (b) composite singles spectra in the Ba $K\alpha - K\alpha^H$ x-ray region recorded by detector Ge(1) for a 122.6-h run. The solid curves indicate the overall least-squares computer fits to the data and the pluses, circles, and dots give the individual components for the $K\alpha$, $K\alpha^H$, and continuum, respectively.

Table I lists the data necessary to calculate P_{KK}(IC) for the two runs. It is clear from the table that run 2, taken with the two Ge detectors, was statistically superior to run 1 which utilized the lower efficiency Si(Li) detector. Run 1 was taken before it was known that the second Ge detector would be available for use in this investigation and is included in the table to demonstrate the stability of the system.

V. DISCUSSION

Of the four mechanisms discussed earlier (SO+SU, DC, NC, ICE), most of the theoretical effort has been on the first. Mukoyama and Shimizu[3] (MS) have calculated the probability of K-shell electron SO in internal conversion, but not SU, by use of a one-step relativistic overlap theory. Their value for the 0.662-MeV transition in $^{137}Ba^m$ is 3.76×10^{-5} which is a factor of 2.7 ± 0.3 smaller than our experimental value. Any contribution due to SU would decrease the discrepancy.

Although no formal theory has been presented, Feinberg[15] has estimated the ratio of $P_{KK}(\mathrm{DC})/P_{KK}(\mathrm{SO})$ 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 present case, the ratio turns out to be only 0.060 and P_{KK}(DC) is thus relatively unimportant in understanding the large discrepancy between experiment and theory.

While Eichler and Jacob,[16] Eichler,[17] and Grechukin[18]

TABLE I. Data needed to calculate P_{KK}(IC) using Eq. (3).

Run	Time	$N(K\alpha^H K\alpha^S)$	$N(K\alpha)$	$(aE)_{\mathrm{Ge(1)}}$	P_{KK}(IC)
1	1041.5 h	168 ± 39	988.2×10^6	$(2.43\pm0.12)\times10^{-3}$	$(10.0\pm2.3)\times10^{-5}$
2	122.6 h	1125 ± 86	199.2×10^6	$(8.12\pm0.43)\times10^{-2}$	$(10.0\pm0.9)\times10^{-5}$

TABLE II. Summary of experimental results and theoretical calculations for P_{KK}(IC) in the 0.662-MeV transition in $^{137}Ba^m$.

Experimental P_{KK}(IC) ($\times 10^5$)		Theoretical P_{KK}(IC) ($\times 10^5$)[a]		
This work	Previous work	P_{KK}(SO)	P_{KK}(DC)	P_{KK}(ICE)
10.0 ± 0.9	18 ± 5[b]	3.76	0.23	204
	<20[c]			
	7.1 ± 3.5[d]			
	<5[e]			

[a]SO, DC, and ICE refer to shakeoff, direct collision, and internal conversion of the internal Compton radiation, respectively. These values are discussed and referenced in the text.
[b]Reference 5, $(e^- - e^-)$–x-ray coincidences in 115–472-keV energy range.
[c]Reference 6, direct observation of satellite in the e^- spectrum.
[d]Reference 7, x-ray–x-ray coincidences.
[e]Reference 8, x-ray–x-ray coincidences.

have presented formal theories for two quanta transition intensities, it is very difficult to determine P_{KK}(NC) theoretically. It is expected, however, that these values will be much less than P_{KK}(SO) and no specific comparison has been made.

The probability for the internal conversion of the internal Compton radiation has been proposed by Listengarten[19] to be

$$P_{KK}(\mathrm{ICE})=\frac{4\alpha}{3\pi}W\int_{\mathrm{BE}}^{W-\mathrm{BE}}\frac{\alpha(E1,E)}{2}\frac{dE}{E}\ , \tag{4}$$

where W is the energy of the nuclear transition (in electron mass units), 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,[20] evaluation of Eq. (4) yields a value for P_{KK}(ICE) of 204×10^{-5} which is a factor of 20 larger than the experimental P_{KK}(IC) value. It should be noted that the Listengarten suggestion has generally given values 3–5 times larger than experiment.[2]

Table II summarizes experimental and theoretical results for P_{KK}(IC) in the 0.662-MeV transition in $^{137}Ba^m$. For values obtained from x-ray coincidence measurements, the theoretical value would be the sum of all the mechanisms discussed. As stated in the introduction, the context of this experiment was one in which recent P_{KK}(IC) results[2,4] had been in reasonable ($\pm 20\%$) agreement with the MS theory. The present result clearly repudiates the Listengarten suggestion and perhaps calls for new and better estimates for P_{KK}(ICE) as much as it calls for new considerations in $P_{KK}(S0)$ calculations as they apply to high-energy, high-multipolarity transitions.

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