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Double *K*-Hole Creation in the Decay of ^{137m}Ba

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Double *K*-hole cration in the decay of ^{137m}Ba has been studied by means of two Si(Li) detectors for two *K* X rays emitted when the double *K* hole is filled and an anthracene crystal with 4π-detection geometry for electrons from a ^{137}Cs source. The total probability per *K* conversion, that the double *K* hole is formed, has been determined to be less than 10^{-4}. The comparison of the present result with existing theories and also with previous experimental results is given.

In the electromagnetic transition between two nuclear levels, double hole may be formed in the *K* shell with a very small probability. The transition energy is shared between two *K* electrons, which are either transfered to one of unoccupied bound states or ejected to the continuum. From the perturbation theory, many different processes are proposed for the e-e transition. The first-order process, called a *K* shakeoff (SOIC) plus *K* shakeup (SUIC), is caused by the sudden change in effective nuclear charge resulting from the loss of an electron as a consequence of the conversion process.[1] The direct collision (DCIC) between a converted *K* electron and another unconverted electron may be possible.[2] Other higher-order processes were studied by several workers, *i.e.*, double internal conversion (DIC) which proceeds *via* virtual intermediate states of the nucleus[3,4] and internal conversion of internal Compton effect (ICICE) *via* virtual atomic intermediate states.[5]

Ljubičić *et al.*[6] observed simultaneous ejection of two electrons in coincidence with *K* X ray in the decay of ^{137m}Ba. They obtained a value of $(1.8\pm0.5)\times10^{-4}$ for the probability per *K* conversion that two electrons are ejected in the energy range from 115 to 472 keV. In the direct electron spectroscopy measurement using a magnetic β-ray spectrometer with isotope-separator samples, Porter, Freedman and Wagner revealed fine structures just below *K*-conversion line.[7] Although they could not find out a *KK*-satellite due to the SOIC process in the case of ^{137m}Ba, they set an upper limit of 2×10^{-4} for the total SOIC probability per *K* conversion. Briand *et al.*[8,9] have recently performed a series of *K* X-ray–*K* X-ray coincidence measurements to find out the *K*-hypersatellite line which is emitted when an outer shell electron transfers to the completely empty *K* shell. By measuring the intensity of the hypersatellite line, they have determined the double *K*-hole creation probability per *K* conversion to be $(7.1\pm3.5)\times10^{-5}$ for the decay of ^{137m}Ba.[9] This value agrees with the upper limit obtained by Porter, Freedman and Wagner,[7] implying that the main contribution to the e-e transition may be caused by the SOIC plus SUIC process.

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The double K-hole creation probability measured by Briand *et al.* is generally larger than the probability of double K-electron ejection. Nevertheless, their experimental value is considerably smaller than the partial double K-electron ejection probability for electrons with energies from 115 to 472 keV measured by Ljubičić *et al.*[6] This discrepancy between two experimental data has not been explained.

In order to obtain a new datum of the K-hole creation probability of ^{137m}Ba, we have performed a triple-coincidence measurement between two K X-ray photons and electrons emitted in the 662-keV transition. Following the technique by Briand *et al.*,[8,9] we have searched for a K_α-hypersatellite peak in the coincidence photon spectrum. The block diagram of electronics is shown in Fig. 1.

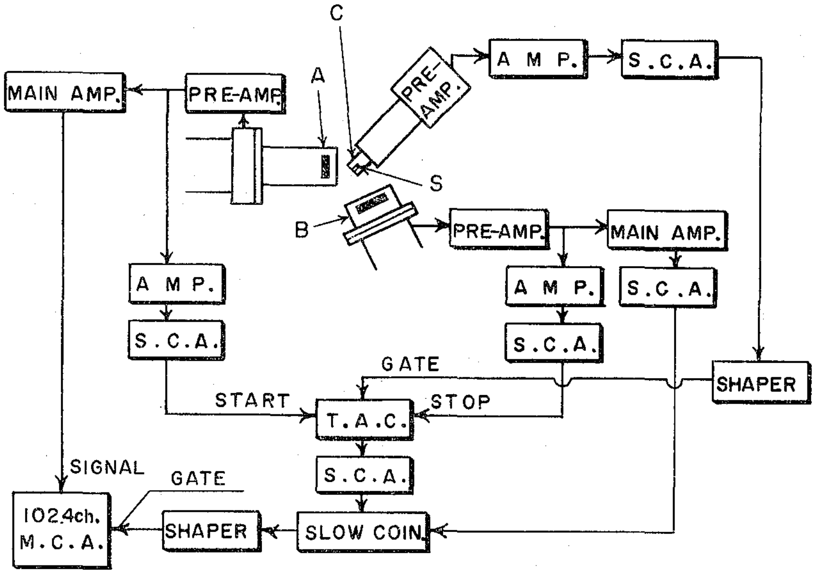

Fig. 1. Block diagram of the electronic system used in the triple-coincidence measurement with ^{137m}Ba: A, Si(Li) detector, 12 mm^2$\times$3 mm; B, Si(Li) detector, 80 mm^2$\times$5 mm; C, anthracene crystal; S, ^{137}Cs source.

An anthracene crystal of 6$\times$6 mm^2 by 8-mm thickness was used to detect electrons from the ^{137}Cs source. To achieve the 4π-detection geometry for electrons, the aqueous solution of ^{137}CsCl was poured into a hole of 0.5-mm diam. by 3-mm depth dug at the center of the crystal. After removing moisture in the source, the crystal was covered with a thin aluminum reflector ($\sim$10 μm) and then mounted on an RCA 4516 photomultiplier. The measured resolving power (FW-HM) was about 15 % for the 624-keV K-conversion line. The intensity of this source was estimated to be about 1.5 μCi from the counting rate of conversion electrons.

The K_α hypersatellite to be studied would be buried in a large background peak of ordinary K_α line caused by random or spurious coincidence events. The detector with better energy resolution is necessary to find out such a small peak due to the hypersatellite line. A Si(Li) detector (A) of EDAX corporation used for this purpose has an energy resolving power (FWHM) of 175 eV for Mn K line. This can resolve an energy difference (~700 eV) between ordinary K_α line and the K_α hypersatellite line of Ba atom. Using the detector A, we have observed the spectrum of photons in coincidence with electrons detected by the anthracene crystal and K X rays by another Si(Li) detector (B) The coincidence counting system with a time-to-amplitude converter (TAC) has a resolving time of about 70 ns with a 100 % coincidence efficiency. Since we achieve the 4π-detection geometry for electrons, two K electrons simultaneously emitted in the e-e transition are always detected by the anthracene crystal and create a signal corresponding to the energy of E_0–$2B_K$, where E_0 is the transition energy and B_K is the K-shell binding energy. Couning rate of each channel and time spectrum from the TAC were monitored to check the performance of the whole system. Gain drifts of linear signals from the Si(Li) detectors were also examined every 12 hours and were verified to be less than 0.2 % during the long-run experiment.

Observed photon spectra are shown in Fig. 2; (a) is the spectrum obtained by a single measurement and (b) is that obtained by the triple coincidence measurement. The measuring period for the coincidence spectrum is 1526 hours. In our coincidence spectrum of Fig. 2b, we could not find out any line at the position (32.95 keV) where Briand *et al.*[9] observed the K_α hypersatellite. The upper limit to the double K-hole creation probability per K conversion, P_{KK}, has been estimated as follows. The total counts of the K_α-hypersatellite line, $N_C^{h\alpha}$, is expressed as

$$N_C^{h\alpha} = n_e \cdot T \cdot P_{KK} \cdot \omega_{KK}^{h\alpha} \cdot D_1^{h\alpha} \cdot (\omega_K^\alpha D_2^\alpha + \omega_K^\beta D_2^\beta)\,, \tag{1}$$

where n_e is total counting rate of K-conversion electrons from the ^{137}Cs source, T total measuring period to obtain the coincidence spectrum, $\omega_{KK}^{h\alpha}$ the fluorescence yield for K_α-hypersatellite line, $D_1^{h\alpha}$ the absolute detection efficiency of the detector A for the K_α-hypersatellite line, D_2^α and D_2^β absolute detection efficiencies of the detector B for K_α and K_β X rays, respectively. Assuming that the fluorescence yield for a doubly ionized atom is the same as that for a singly ionized atom,[10] $\omega_{KK}^{h\alpha}$ is set to ω_K^α. Since the detection efficiency of the anthracene crystal for electrons is approximately 100 %, $D_1^{h\alpha}$ is given by

$$D_1^{h\alpha} = \frac{n_X^\alpha(1)}{n_e \cdot \omega_{KK}^{h\alpha}} \simeq \frac{n_X^\alpha(1)}{n_e \cdot \omega_K^\alpha}\,, \tag{2}$$

where $n_X^\alpha(1)$ is the counting rate of K_α X rays detected by the detector A. Similarly, we obtain

$$D_2^\alpha = \frac{n_X^\alpha(2)}{n_e \cdot \omega_K^\alpha} \quad \text{and} \quad D_2^\beta = \frac{n_X^\beta(2)}{n_e \cdot \omega_K^\beta}\,, \tag{3}$$

where n_X^α (2) and n_X^β (2) are counting rates of K_α and K_β X rays detected by the detector B, respectively. Using Eqs. (2) and (3), the total probability per K conversion, P_{KK}, can be expressed as

$$P_{KK} = \frac{(N_C^{h\alpha}/T) \cdot n_e}{n_X^\alpha(1) \cdot [n_X^\alpha(2) + n_X^\beta(2)]}\,. \tag{4}$$

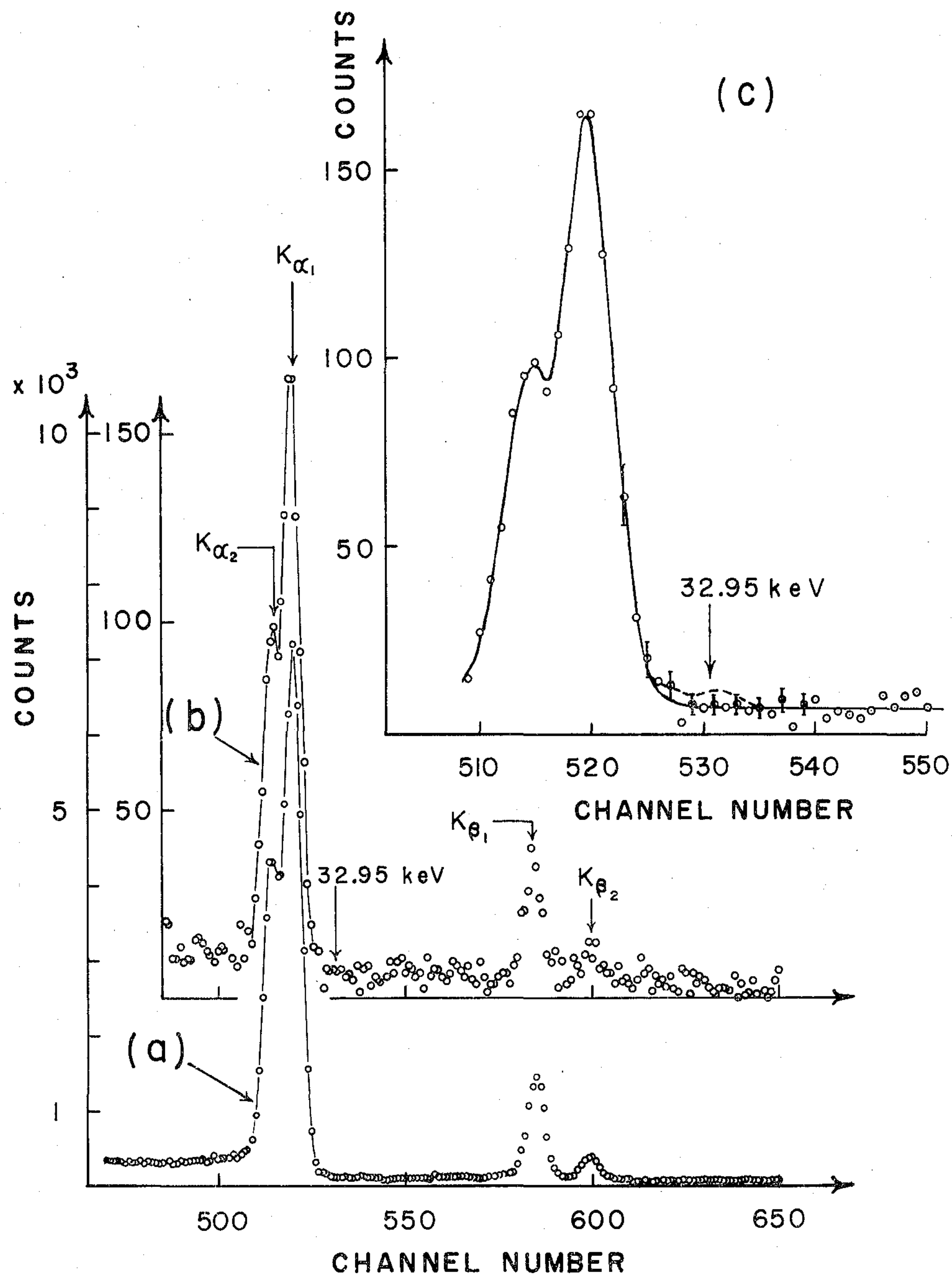

Fig. 2. Experimental spectra of Ba K X rays. Spectrum (a) was obtained by a single measurement, while spectrum (b) was obtained by the coincidence measurement. A part of the coincidence spectrum near K_α X rays is expanded in the inset (c) with a solid curve obtained by the least-χ^2 fit. The area surrounded by the solid line and the dashed one was used to determine the upper limit to the coincidence counts $N_C^{h\alpha}$.

In the present work, values of n_e, $n_X^{\alpha}(1)$, $n_X^{\alpha}(2)$, and $n_X^{\beta}(2)$ are $(5.2\pm0.2)\times10^3$ /sec, (5.5 ± 0.2)/sec, $(4.0\pm0.1)\times10$/sec and (7.2 ± 0.6)/sec, respectively.

The solid line in Fig. 2c was obtained by the non-linear least χ^2 fit, assuming that spectra of $K_{\alpha 1}$ and $K_{\alpha 2}$ lines are given by Gaussian shapes and the background level is given by a straight line. The dashed curve expresses two peaks assumed to be due to the $K^h_{\alpha 1}$- and $K^h_{\alpha 2}$-hypersatellite lines; this curve was drawn so that heights of the Gaussian peaks are not over standard deviations of experimental points near 32.95 keV. We have employed the area of the peaks as an upper limit to the coincidence counts $N_C^{h\alpha}$:

$$N_C^{h\alpha} \leqq 30 \text{ counts.} \tag{5}$$

Substituting this value into Eq. (4), we obtain

$$P_{KK} \leqq 10^{-4}\,. \tag{6}$$

Present result quite disagrees with the measurement by Ljubičić *et al.*;[6] our upper limit to P_{KK} is smaller than their partial probability of double K-electron ejection. Our result rather sides with that by Briand's group.[9] However, it is not clear why we failed to find out the hypersatellite line observed by Briand's group.

Recently, Mukoyama and Shimizu[11] have developed the one-step theory of the SOIC process, taking account of an effect of the multipolarity of the electromagnetic transition. The most reliable value for ^{137m}Ba calculated by them is 3.76×10^{-5}, which is consistent with our result. The experimental value by Briand's group agrees with the theoretical one within the experimental error. These facts suggest that the double K-hole creation in conversion decay may occur mainly through the SOIC plus SUIC process.

We have calculated the total probability for the ICICE process according to the Listengarten's equation[5] of

$$P_{KK}(ICICE) = \frac{4\alpha}{3\pi} E_0 \int_{B_K}^{E_0-B_K} \frac{dk}{k} \frac{1}{2} \alpha_K^1(k, Z)\,, \tag{7}$$

where E_0 and B_K are the transition energy and the K-shell binding energy, respectively, Z is the atomic number, and $\alpha_K^1(k, Z)$ is the K-shell internal conversion coefficient for the electric dipole radiation with energy k. Using $\alpha_K^1(k, Z)$ prepared by Hager and Seltzer,[12] we obtain a value of

$$P_{KK}(ICICE) \simeq 1\times10^{-3}\,, \tag{8}$$

which is much larger than our upper limit. This disagreement is probably due to an approximate nature of the Listengarten's theory.[5] More trustful treatment for the process is hoped.

Although Eichler[3] and Grechukhin[4] developed the theory of the DIC process, it is almost impossible to calculate the total DIC probability mainly because of the difficulty evaluating nuclear matrix elements. From their formulations, the total DIC probability can be assumed to be same order as or less than that for the 2γ-

decay process. Beusch[13] measured coincidence counts between two γ quanta simultaneously emitted in the decay of ^{137m}Ba obtaining a value of $(6.4\pm3.1)\times10^{-6}$ for the total 2γ-emission probability per a single γ emission. With this value, the double K-hole creation probability per K conversion due to the DIC process may become the order of 10^{-5}. This means that we can not reject a possibility that the DIC process may be one of main mechanisms for the e-e transition.

It has recently been pointed out that the theory of K-electron shakeoff in β decay is not sufficient to explain the phenomenon of K-shell internal ionization accompanying β decay and the direct collision process (DC) may also play an important role.[14] The simple theory by Feinberg[2] has often been used to exclude a contribution from the DC process; the relative probability of the DC process vs the shakeoff process in β decay approximately equals to a value of B_K/E_0. However, the relation is not reliable because of the rough approximation. The situation is quite same for the DCIC process; we have no ground to ignore the DCIC process in the internal conversion decay.

In conclusion, our result disagrees with that by Ljubičić's group[6] and sides with that Briand's group, suggesting that one of main mechanism may be the SOIC plus SUIC process. In order to make clear a main mechanism of the e-e transition, at least the order of the probabilities due to the processes of DIC, ICICE, and DCIC should be determined.

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