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Double Electron Ejection in the Decay of ^{137}Ba

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Simultaneous ejection of two electrons in the decay of the 662-keV state of ^{137}Ba has been investigated by means of two Si(Li) detectors and a two-dimensional pulse-height analyzer. The KK process has been observed, while for the $K, L+M+\ldots$ processes only an upper limit has been determined. The results obtained are $T_{KK}/T_K = (1.8 \pm 0.5) \times 10^{-4}$ and $T_{K, L+M+\ldots}/T_K \leq 4.5 \times 10^{-4}$. The comparison of the experimental results with the existing theories indicates that the process proceeds primarily via electronic intermediate states.

INTRODUCTION

A considerable number of experiments have been performed in the search for higher-order electromagnetic transitions in nuclear decay.[1] Investigations of the $M4$ transition from the excited state at 662 keV to the ground state in ^{137}Ba have been particularly successful. Double-photon ($\gamma\gamma$) emission has been observed only in the decay of that state.[2] Also, the only measurements of the angular distribution and of the energy distribution in the $e\gamma$ process have been made for the same transition.[3,4] The third possible two-quantum transition needed to complete the scheme, namely double electron ejection (ee process), was a missing piece of evidence. In the present experiment this process has been observed.

Two types of experiments were previously performed with the aim of discovering the ee process: (i) systems of β spectrometers were used to search for coincident emission of the two electrons,[5] and (ii) coincident emission of x rays that follow the ejection of the two electrons from the atom were searched for with systems of coincident x-ray detectors.[6] In the former case, energy selection takes place before detection, and therefore it is possible to observe the double electron ejection without interference from the direct radiation. Vukanović *et al.*[7] have recently published the results of investigations of the ee processes in ^{114}In using a three-lens electron spectrometer at Uppsala. They find evidence for the KK process with a relative transition probability of $T_{KK}/T_K = (1.7 \pm 0.3) \times 10^{-5}$. The main difficulties in the work with magnetic spectrometers come from the small transmission of the apparatus and the relatively narrow energy interval accepted. Identification of the ee process events by observing coincident x rays is rather difficult and uncertain because of other processes that may produce equivalent coincidences. For example, the two K x rays emitted from the same atom in the case of a KK process may be masked by coincident emission of K x rays from different atoms in the source. These events may arise if a K conversion electron (always accompanied by the emission of a K x ray) ejects a K electron from another atom in the source, giving rise to the emission of the other K x ray.

The method of two-dimensional energy analysis combined with Si(Li) detectors of good resolution, which was applied in the present experiment, has some advantages. It allows a simultaneous measurement of the whole energy spectrum of emitted particles. The sum of energies of the two electrons, E_1 and E_2, ejected from an atom in the ee process is equal to the total transition energy W_0

minus the sum of the corresponding binding energies B_1 and B_2,

$$E_1+E_2=W_0-(B_1+B_2)\,. \tag{1}$$

This corresponds to lines of constant sum energy (depending on the electron shells) in a two-dimensional plot of E_1 versus E_2. By studying the patterns in such diagrams one can distinguish events due to the *ee* process from other electron-electron coincidences, such as scattering of conversion electrons on electrons in other atoms, real *e-e* and β^--*e* cascades.

The *ee* process can proceed via two different mechanisms, involving either nuclear or electronic intermediate states. A detailed theory of the former mechanism was first developed by Eichler.[8] No specific calculations of the other mechanism have been made. However, Listengarten[9] has shown that an estimate of the order of magnitude of the transition probability for this process can be made, considering it as the "internal conversion of the internal Compton effect." The "nuclear" *ee* process is usually called "double internal conversion."

EXPERIMENTAL ARRANGEMENT

The experimental arrangement is shown in Fig. 1. The radioactive source was an approximately 40-μg/cm^2-thick film of ^{137}Cs of about 30 μCi. It was deposited on a Mylar foil 1 mg/cm^2 thick. Electrons were detected in two cooled Si(Li) detectors 15 mm diam$\times$1.2 mm thick, mounted inside a vacuum chamber. Their resolution for 624-keV K conversion electrons from ^{137}Ba was 14 keV. To prevent the scattering of electrons from one detector into another, an aluminum shield was placed between them. A NaI(Tl) scintillation crystal $1\frac{1}{2}$ in. diam$\times$3 mm thick was placed close above the source to detect K x rays. The electronic equipment consisted of a standard fast-slow coincidence system, which is described in the preceding paper.[4] The resolving time of the triple fast-coincidence circuit was set at a value of 45 nsec in order to achieve a good efficiency over a wide energy interval. The windows of the single-channel analyzers were set to accept electrons from 115 to 472 keV and K x rays in the energy interval from 17 to 41 keV, respectively.

According to Eq. (1) several parallel lines corresponding to the KK, KL, KM, etc., processes are expected in the E_1 versus E_2 plane. Due to the finite energy resolution of the apparatus we could not distinguish the processes on loosely bound electrons. For that reason we were only able to separate the KK events from the $K, L+M+\cdots$ events.

MEASUREMENTS AND RESULTS

Figure 2 shows the results of measurements at three settings of the electron detectors, for which the angles between the emitted electrons, θ, were around 35, 45, and 90°. Since too few events were recorded to determine the energy distribution of electrons, the events in the two-dimensional diagram were summed along the straight lines of constant sum energy of the two electrons. At the sum energy $E_1+E_2=587$ keV one can see peaks that are interpreted as events due to the KK process. The background is due to accidental coincidences.

Careful energy calibration of the experimental arrangement was performed to make the energy scale accurate. First, the system was calibrated with one counter at a time. Then, with the same arrangement as in the measurements of the two-electron processes, except that the aluminum shield between the electron counters was removed, triple-coincidence events between K x rays and K conversion electrons (from direct transitions) scattered from one detector into another were recorded. The results of that measurement are

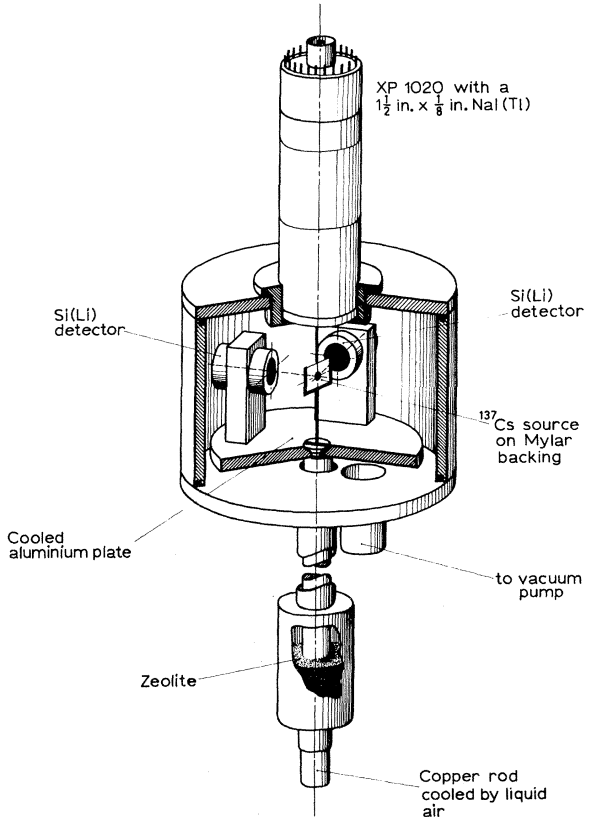

FIG. 1. Sketch of the vacuum chamber with electron and x-ray detectors. The aluminum shield between the detectors is not shown.

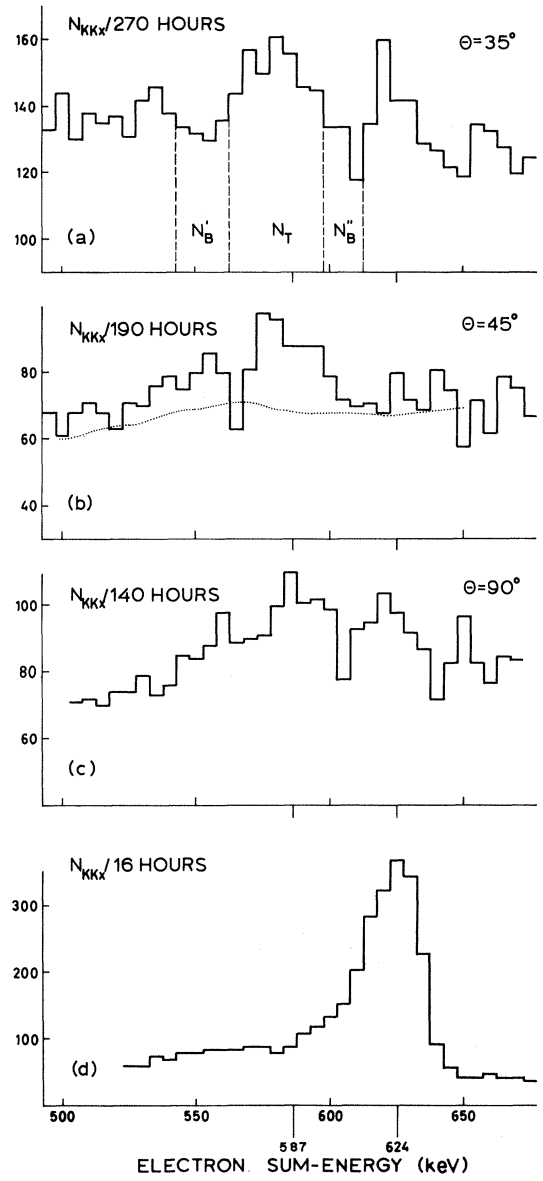

FIG. 2. Number of triple coincidence events as a function of the electron sum energy at (a) 35°, (b) 45°, (c) 90°. N_T represents the total number of coincidences under the peak. N'_B and N''_B are used to determine the background under the peak. The dashed lines determine the interval in which N_T, N'_B, and N''_B were integrated. The dotted line in (b) represents the calculated number of accidental coincidences. The energy calibration line, obtained by removing the shield between the two electron detectors, is shown in (d).

shown in Fig. 2(d). In this way, in the two-dimensional diagram the calibration line $E_1+E_2=624$ keV was obtained. To recheck this line, a thin aluminum foil was placed between the source and the electron detectors to observe the triple-coincidence events of K x rays and of K conversion electrons scattered on loosely bound electrons in the foil.

An event due to the scattering of a K conversion electron on K-shell electrons in other barium or cesium atoms (external scattering) is indistinguishable from an internal KK event. However, if the external process contributed considerably to the observed peak at 587 keV, a more intense peak at an electron sum energy of 624 keV would be expected, which would be due to the external scattering of K conversion electrons on L,M, etc. electrons in barium and cesium atoms, and on other loosely bound electrons in atoms in the source. In fact, the ratio of the total probability for the external scattering of K conversion electrons on K electrons, T^{ext}_{KK}, and on loosely bound electrons, $T^{\text{ext}}_{K,L+M+\ldots}$, in the surrounding atoms should be

$$T^{\text{ext}}_{KK}/T^{\text{ext}}_{K,L+M+\ldots} \leq 2/(Z-2) = 1/27 . \qquad (2)$$

The relation (2) is not valid for the ratio of differential probabilities because the binding of electrons in atomic shells may cause different distortions of angular distributions.

A smaller peak appears at the energy of 624 keV. Although one cannot decide whether it is due to external scattering or to the internal $K, L+M+\ldots$ process, one can derive a limit for either process using an equation analogous to Eq. (3) below. The results are shown in Table I (first column). Due to the electron-electron scattering a ratio much smaller than $\frac{1}{27}$ is expected at the angle of 90° between the two electron detectors. The statistics of the 90° data are not so good as those of the 35 and 45° data (experiments are rather time consuming). However, the absence of a stronger peak at 624 keV in the 90° data shows that the external scattering in the source was very weak.

In order to determine the ratio of the transition probability for the two-electron process $T_{KK}(\theta)$ to the single-electron conversion probability T_K, the number of double coincidences N_{Kx} between the direct K conversion electrons detected in the electron detector No. 1 and K x rays was recorded during the daily checks of the apparatus. The ra-

TABLE I. The relative transition probabilties of double electron ejection processes per unit solid angle.

θ (deg)	$T_{K,L+M+\ldots}(\theta)/T_K$	$T_{KK}(\theta)/T_K$
35	$\leq 1.9\times10^{-5}$	$(1.2\pm0.4)\times10^{-5}$
45	$\leq 1.7\times10^{-5}$	$(1.5\pm0.5)\times10^{-5}$
67	$\leq 1.4\times10^{-5}$	$(1.1\pm0.5)\times10^{-5}$
90	$\leq 3.0\times10^{-5}$	$(1.2\pm0.6)\times10^{-5}$
135	$\leq 4.8\times10^{-5}$	$(1.3\pm0.6)\times10^{-5}$

tio can be obtained from the expression

$$T_{KK}(\theta)/T_K = N_{KKx}/(4\Omega_2 N_{Kx}), \quad (3)$$

where N_{KKx} is the number of triple-coincidence counts attributed to the KK process, and Ω_2 is the solid angle of the electron detector No. 2. The factor 4 in the denominator is due to (i) the emission of two K x rays per KK process (their energy is only slightly different), and (ii) the factor $\Omega_1/(\Omega_1+\Omega_2)$ with Ω_1 approximately equal to Ω_2, which arises because one of the electrons has the option being detected in either electron detector.

The background N_B under the peaks at 587 keV was first estimated from the calculated accidental coincidence rates. To obtain these rates, we measured direct spectra in each electron detector and also spectra from each electron detector in coincidence with the K-x-ray detector. These spectra show a continuous distribution, except at 624 and 656 keV in the former case, and at 624 keV in the latter case. We calculated accidental triple coincidence rates between the two electron detectors and the x-ray detector, and the accidental coincidence rates between the measured true coincidences in one electron detector and the x-ray detector with the direct spectrum from the other electron detector. The calculations were performed in the energy interval from 115 to 472 keV for each electron detector, so that the peaks at 624 and 656 keV were not taken into account. The spectrum of total accidental events was then obtained summing both spectra of accidental coincidences. The result is shown in Fig. 2(b) by the dotted line. The number of accidental coincidences obtained is somewhat smaller than the background seen in the measured data. It varies, as expected, smoothly as a function of the sum energy. Therefore, a more reliable procedure to determine N_B was adopted. It consisted in determining the number of counts in seven channels, four to the left (N_B') and three to the right (N_B'') from the seven channels at the position of the peak [see Fig. 2(a)].

The number of triple coincidences N_{KKx} was obtained from the relation

$$N_{KKx} = N_T - (N_B' + N_B''), \quad (4)$$

where N_T is the total number of counts in seven channels at the position of the peaks at 587 keV. From the number of coincidences in the peaks at 587 keV, the ratio of the KK process to the internal K conversion was determined using Eq. (3). The results are shown in the table (second column) and in Fig. 3. Integrating over all angles of emission, we obtained the experimental result for the relative total transition probability of the KK process in the decay of ^{137}Ba,

$$T_{KK}/T_K = (1.8 \pm 0.5) \times 10^{-4} \quad (5)$$

for electrons between 115 and 472 keV.

In a previous experiment[1] we obtained the result $T_{KK}/T_K = (1.6 \pm 0.9) \times 10^{-4}$ by the fourfold coincidence technique. We detected two electrons and two K x rays emitted in the KK process. Since we were not able to estimate the contribution of the external scattering effects, we reported only an upper limit of 2.5×10^{-4}. This value is in agreement with the result obtained in the present experiment.

From the data in the first column we also obtained an upper limit for the $K, L+M+\cdots$ process in ^{137}Ba,

$$T_{K,L+M+\ldots}/T_K \leq 4.5 \times 10^{-4} \quad (6)$$

also for electrons between 115 and 472 keV.

The upper limit of the contribution of the external scattering of K conversion electrons on K-shell electrons in barium and cesium atoms in the source can be derived from the latter result. Assuming that the result for the $K, L+M+\ldots$ processes is entirely due to the external scattering effects, and taking into account the relation (2), we obtained an upper limit of $T_{KK}^{\text{ext}}/T_K \leq 1.7 \times 10^{-5}$, which is small compared with our experimental value for the internal KK process, and was neglected in the analysis of the data at 587 keV.

An excited state below the 662-keV state in ^{137}Ba, at an energy of 281 keV, was reported.[10] A cascade transition through such a state would also yield true coincidences, which satisfy the same energy condition (1) as the ee processes. From the experiment of Beusch[2] it is evident that the contribution of such a real cascade is very small, and it was also neglected in the analysis of the present experimental data.

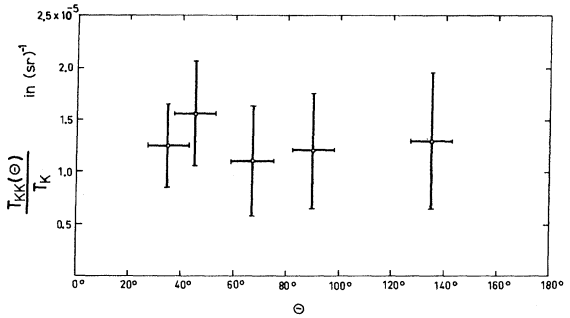

FIG. 3. The experimental angular distribution of the KK process per K conversion in ^{137}Ba. The vertical error bars represent the statistical errors of the measurements. The horizontal bars represent the total region of angles accepted in each measurement.

DISCUSSION

The relative contribution of the mechanism involving nuclear intermediate states and of the mechanism involving electronic intermediate states can be determined by comparing the experimental data with the theoretical predictions. For a second-order nuclear process, in the case of one predominant combination of multipoles, Eichler[8] gives the energy spectra and transition probabilities for the two-quantum decay involving two-conversion electrons. He has shown that the interaction with K-shell electrons gives the main contribution. For the nuclear KK process the energy distribution can be written in the form

$$T_{KK}(E) = G(\tau_1 L_1, \tau_2 L_2) C_L [\alpha_K(\tau_1 L_1, \omega_1)\alpha_K(\tau_2 L_2, \omega_2) \times \omega_1^{2L_1+1}(W_0-\omega_1)^{2L_2+1} + \alpha_K(\tau_2 L_2, \omega_1) \times \alpha_K(\tau_1 L_1, \omega_2)\omega_1^{2L_2+1}(W_0-\omega_1)^{2L_1+1}] . \quad (7)$$

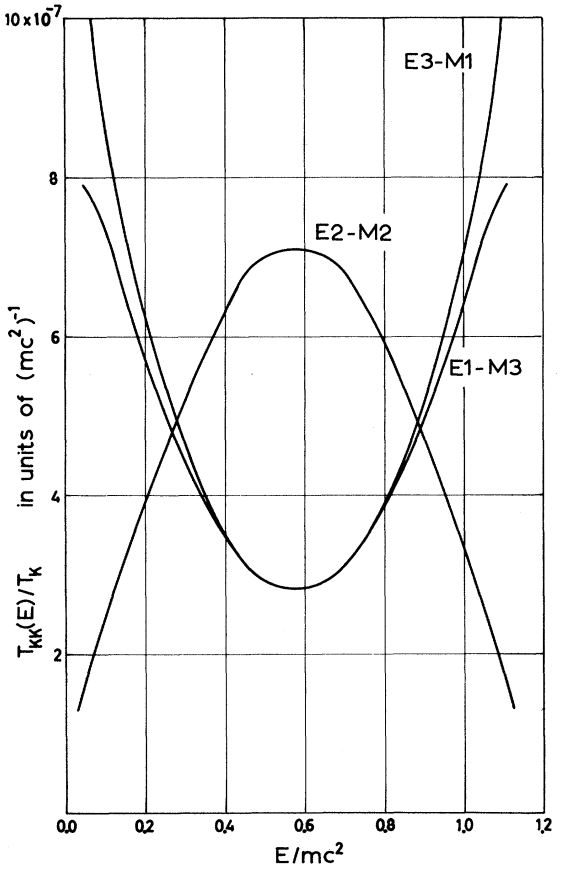

FIG. 4. The transition probabilities integrated over the angle for the nuclear KK processes in ^{137}Ba versus electron energy E calculated from Eichler's theory. The curves shown represent the most important combinations of multipolarities of emitted electrons. The ordinate scale was estimated from the experiment of Beusch. The results represent upper limits.

ω_1 and ω_2 are the energies of the two converted photons which satisfy the relation $W_0 = \omega_1 + \omega_2$. The corresponding energies of the emitted electrons, $E = \omega - B$, are smaller because of the binding energies B of the electrons in the atom. L_1 and L_2 denote the multipolarities of each transition separately, and $L_1 + L_2 = L$ denotes the multipolarity of the direct transition. The $\alpha_K(\tau L, \omega)$ are the K-shell conversion coefficients, where τ designates the type of radiation (electric or magnetic multipole). The factor G is given by

$$G(EL_1, EL_2) = (L_1 + L_2 + 1)/2(L_1 + 1)(L_2 + 1); \quad (8)$$
$$G(ML_1, EL_2) = G(EL_2, ML_1) = (2L_2 + 1)/(2L_2 + 2) .$$

The constant C_L must be determined from experimental data or evaluated from nuclear models. It includes all of the properties of nuclear states involved in the double decay of the nucleus. In this way it would be difficult to calculate C_L since the intermediate nuclear states of ^{137}Ba, which may contribute to the process, are not known. We assumed that in the nuclear KK process the same intermediate states are involved as in the double-photon decay. Then the constant C_L is the same for both processes and it can be evaluated from

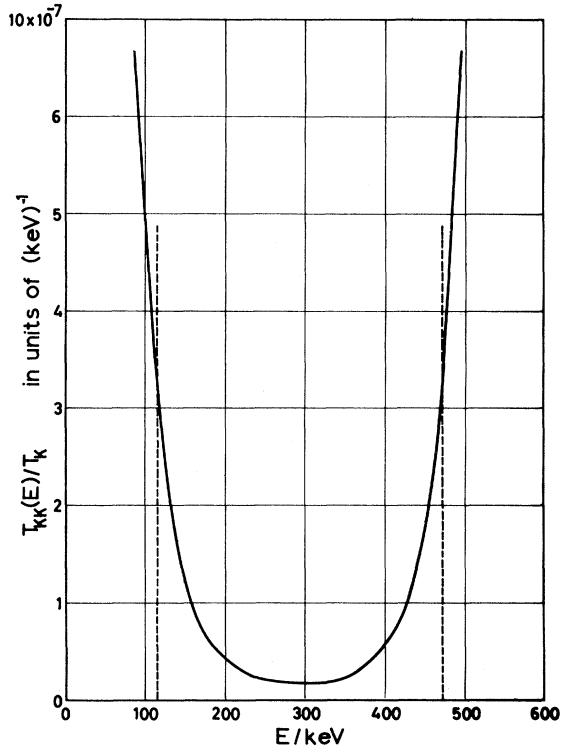

FIG. 5. Relative transition probability for the KK process versus electron energy E as calculated from Listengarten's theory. The dashed vertical lines denote the energy interval of electrons that were detected in the experiments.

the results of the experiment of Beusch. In the previous paper[4] we obtained the following values for C_L/T_K: $\leq 8.77\times 10^{-3}$ for the $E2$-$M2$ transition, and $\leq 4.22\times 10^{-3}$ for the $E1$-$M3$ and $E2$-$M1$ transitions. The constants C_L/T_K are given in units of $(mc^2)^{-11}$.

Using these values of C_L/T_K and Eqs. (7) and (8) we calculated the energy spectra for the nuclear KK processes for different multipole combinations. The results are shown in Fig. 4. The numerical values for α_K were obtained from the tables of Hager and Seltzer.[11] Integrating over the electron energy interval from 115 to 472 keV, we calculated the contributions of the nuclear mechanism in the KK process. For the $E1$-$M3$, $E2$-$M2$, and $E3$-$M1$ transitions, we obtained limits for T_{KK}/T_K of $\leq 2.6\times 10^{-7}$, $\leq 4.3\times 10^{-7}$, and $\leq 2.6\times 10^{-7}$, respectively. We therefore conclude that nuclear intermediate states give a negligible contribution to the observed process.

We also made an estimate of the transition probability of the KK process proceeding via electronic intermediate states by following Listengarten's simplified theory.[9] The transition probability for the KK process in that theory can be calculated by multiplying the transition probability for the $K\gamma$ process by $\alpha_K(E1,\omega)/2$ to account for the conversion of the remaining K electron by the internal Compton photon. This procedure can be written as

$$T_{KK}(E)/T_K=[T_{K\gamma}(\omega)/T_K][\alpha_K(E1,\omega)/2], \qquad (9)$$

where $T_{K\gamma}/T_K$ are the coefficients for the internal Compton effect as a function of the photon energy ω and α_K are the internal-conversion coefficients for the $E1$ type transition in the K shell.[11] From the expression (9) we calculated the relative transition probability for the KK process as a function of the electron energy. The energy spectrum obtained is shown in Fig. 5. By integrating the relation (9) from 115 to 472 keV we obtained a value of $T_{KK}/T_K=2.5\times 10^{-5}$. This theoretical value is too small by a factor of about 7, when compared with the result of the present experiment given in Eq. (5). This disagreement is probably due to the approximate nature of Listengarten's estimate of the KK process. The value calculated from Eichler's theory (which is much more reliable, but takes account of nuclear intermediate states only) is lower by almost two further orders of magnitude. This shows that the main contribution to the KK process in ^{137}Ba comes from the processes which proceed via virtual intermediate states of electrons.

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