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Deexcitation of the 662-keV State in ^{137}Ba by the Internal Compton Effect

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A double-decay process in which the transition energy is distributed between one photon and one K conversion electron was investigated in the decay of the 662-keV state in ^{137}Ba. For the energy interval of emitted photons from 50 to 150 keV, the result of $T_{K\gamma}/T_K = (2.2 \pm 0.2) \times 10^{-3}$ was obtained. The measured energy and angular distributions are compared with the Spruch-Goertzel theory in which the process is treated as the internal Compton effect. Some deviations from the theory are observed in both distributions. The contribution of nuclear intermediate states is negligible in the range of energies which was investigated.

INTRODUCTION

Experimental studies of higher-order electromagnetic transitions in nuclei have been performed for only about a dozen transitions. Most of them only provided upper limits on the transition probabilities.[1] The reasons for this are the very low probabilities compared with first-order transitions (γ-ray emission, electron conversion, and internal pair production at higher energies), and low efficiency of detection (due to the use of two or more detection systems in coincidence, and because of the continuous energy spectra of emitted quanta). Most attempts have been made with transitions that are strongly forbidden in first order. The $M0$ transition ($0^+ \rightarrow 0^-$ or the reverse) would be an ideal case. Unfortunately, no such pair of levels has been found in which the higher one has no other transition channels (to intermediate or lower levels). Several $E0$ ($0^+ \rightarrow 0^+$) transitions, for which single γ-ray emission is strictly forbidden, have been studied. The other group of transitions for which the "double decay" has been more extensively studied consists of isomeric transitions of high multipolarity. Only transitions from the first excited state to the ground state have so far been studied, because of the difficulties with crossover transitions.

Two mechanisms are considered to contribute to higher-order electromagnetic transitions with the emission of two quanta (quanta = photons, electrons). In one, nuclear virtual intermediate states are involved; nuclear states above the initial state, through the tails of their resonance lines, give rise to one transition with a simultaneous transition from the intermediate state to the final state. The lowest-order diagrams for these "nuclear" decay modes are shown in Figs. 1(a)–1(c). They are closely related: an $e\gamma$ process is obtained by electron conversion of one of the transitions of the $\gamma\gamma$ process, and an ee process by electron conversion of both transitions. The theory for these processes has been worked out in detail by Eichler and Jacob,[2] by Eichler[3] and by Grechukhin.[4]

The nucleus may also transfer its excitation energy to an orbital electron which either radiates a photon (an $e\gamma$ transition) or gives rise to the emission of another orbital electron (an ee transition). The diagrams involving these "electronic" decay modes are shown in Figs. 1(e) and 1(f). The transition shown in Fig. 1(e) is often called the internal Compton effect. The transition probabilities as well as the energy and angular distributions for this process have been calculated by Spruch and Goertzel[5] and by Melikian[6] in the Born approximation. This mechanism is very successful in accounting for the investigated part of the angular and energy distribution in the $e\gamma$ decay in ^{137}Ba as discussed in the present paper. If the orbital electron radiates two photons and then returns to its initial state, we shall have two photons in the final state ($\gamma\gamma$ transition). This mechanism is the "electronic" analog of the "nuclear" $\gamma\gamma$ transition and is shown in Fig. 1(d). No detailed calculations of the processes shown in Figs. 1(d) and 1(f) have been made, but only very rough estimates.

Nuclear and electronic intermediate states for each of the processes yield kinematically the same final states; hence it is not possible to find out whether a detected two-particle event is due to the one or the other mechanism. However, comparing the measured and calculated transition rates, angular and energy distributions, it is possible to distinguish between them. In the case of ^{137}Ba a calculation of the nuclear part involved in the $e\gamma$ transition would be difficult and rather uncertain, since little is known about the possible intermediate states that would contribute to the process. However, a measurement of the $\gamma\gamma$ emission from the same level at 662 keV in ^{137}Ba has been performed by Beusch.[7] Assuming that this process is entirely due to the transition through the nuclear intermediate state, it is possible to make an

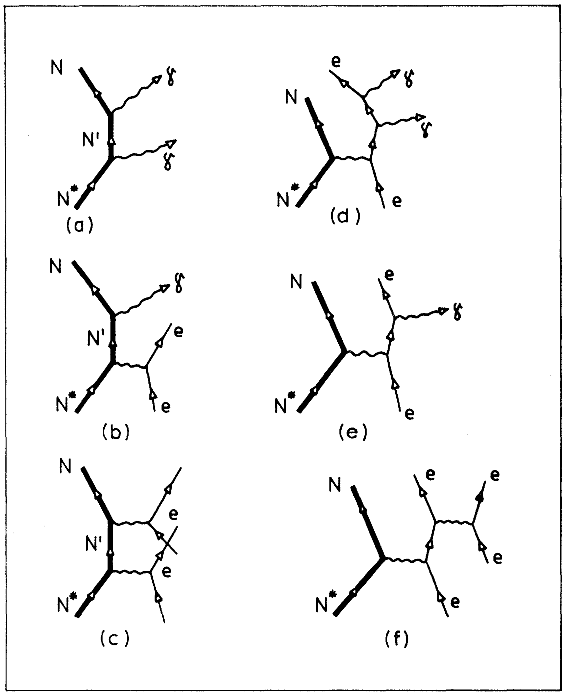

FIG. 1. Feynman diagrams for $\gamma\gamma$, $e\gamma$, and ee double-decay processes in nuclear transitions. Diagrams (a), (b), and (c) show the contributions of nuclear intermediate states, while diagrams (d), (e), and (f) show processes which proceed via electronic intermediate states.

estimate of the transition probability for the nuclear part of the $e\gamma$ emission.

Previous measurements of the angular distribution have shown that the $e\gamma$ process in ^{137}Ba proceeds mostly as the internal Compton effect.[8-10] In our preceding paper[11] the results of measurements of the energy distribution of photons at a relative angle of 27° were presented. In this paper an extension of these measurements is reported together with a more detailed discussion of the measurements, analysis, and results.

EXPERIMENTAL METHOD

Here an outline of the present experimental arrangement is given; for further details we refer the reader to Ref. 11.

The experimental method was based on the two-dimensional analysis of pulses from the electron and photon detectors. In the case of $e\gamma$ emission, the photon and the electron energy satisfy the relation

$$E+E_e=W_0-B\,, \tag{1}$$

where E and E_e are the photon and the conversion-electron energy, respectively, W_0 is the transition energy, and B is the electron binding energy in the corresponding atomic shell. The recoil energy of the emitting nucleus is very small and is neglected. All $e\gamma$ events in which the full energy of quanta is deposited in the corresponding detectors appear in the two-dimensional diagram along straight lines, intersecting the axes at points W_0-B.

The source consisted of 30 μCi of carrier-free ^{137}Cs chloride deposited on a thin Mylar foil as a spot about 4 mm in diam. Electrons were detected in a cooled Si(Li) detector. Its resolution was 11 keV for the 624-keV K conversion electrons of ^{137}Ba. The source and the electron detector were placed in a vacuum chamber. Photons were detected in a NaI(Tl) crystal, $1\frac{1}{2}$ in. diam $\times$ $1\frac{1}{2}$ in. thick. In order to detect double-decay events involving K-shell electrons only (the $K\gamma$ process) and to reduce chance coincidences, another NaI(Tl) counter was used to detect coincident K x rays. The crystal, $1\frac{1}{2}$ in. diam $\times$ 3 mm thick, was placed close above the source. Both scintillation detectors were placed outside the vacuum chamber, at ports made of thin aluminum.

The pulses from the three detectors were analyzed in a triple fast-slow coincidence system. The resolving time of the fast coincidence unit was set at a value of $2\tau=40$ nsec. The slow coincidence ($2\tau=1$ μsec) was activated by coincident pulses from the fast-coincidence and from three single-channel analyzers. These analyzers were used to select the energy range for each detector: 25 to 250 keV for photons, 375 to 600 keV for electrons, and about 20 to 40 keV for the x-ray detector. The fast-slow coincidence system was activating the gates for the photon- and electron-detector pulses that were analyzed in a 128 $\times$ 128-channel analyzer. The output from the analyzer was punched on paper tape and subsequently analyzed in a CAE 9040 computer.

The measurements were performed at nominal angles (angles between the directions from the source to the centers of the electron and photon detectors) $\Theta=0$, 27, 45, 70, 100, 135, and 180°. To minimize the absorption of photons, aluminum windows 0.4 mm thick were mounted on the side of the vacuum chamber in front of corresponding positions of the photon detector.

The only process which can give a contribution of true coincidence events in the same locus where $K\gamma$ events are expected is the external bremsstrahlung of monoenergetic K conversion electrons emitted by the source (i) in atoms of the source and the backing, (ii) in surrounding materials, and (iii) in the electron detector.

The effect (i) was carefully investigated by Lindqvist, Pettersson, and Siegbahn.[8] For a source of ^{137}Cs 30 μg/cm^2 thick deposited on a 270-μg/cm^2 aluminum foil, of dimensions very similar to ours, they showed that the contribution of the external bremsstrahlung is negligible in comparison with the $K\gamma$ process. Although the backing in the present experiment was of a somewhat larger thickness (1 mg/cm^2), it was Mylar, which is composed of low-Z elements.

The effects (ii) and (iii) were eliminated at all angles except at 0 and 180° by using adequate collimators and shields. Therefore, the results obtained in the measurements at 27, 45, 70, 100, and 135°, after the background due to accidental coincidences was subtracted, are considered to be due to the $K\gamma$ double decay. In the present experimental arrangement, in the measurements at 0 and 180°, it was not possible to exclude the scattering processes from the electron into the photon detector. Fuschini, Maroni, and Veronesi[10] used a magnetic spectrometer in their measurements at small angles, yielding more reliable results for angles smaller than 27°. We applied a procedure of correcting for the effects of scattering, which is described further on.

ANALYSIS OF THE RESULTS

The experimental data were analyzed using the relation

$$T(K,\gamma;E,\Theta)/T_K = N_{K\gamma x}(E,\Theta)/N_{Kx}\Omega\Delta E\,, \qquad (2)$$

where $T(K,\gamma;E,\Theta)$ is the differential transition probability of the $K\gamma$ process per unit solid angle and unit energy interval of the photons at a nominal angle of measurement Θ, averaged over the solid angles of the photon and electron detectors; T_K is the K conversion transition probability for the same transition; $N_{K\gamma x}(E,\Theta)$ is the measured number of triple-coincidence events in the photon energy interval ΔE due to the $K\gamma$ process; N_{Kx} is the number of double-coincidence events of K conversion electrons and K x rays, measured in separate runs; and Ω is the effective solid angle of the photon detector. The same factor due to the K fluorescent yield appears in both the numerator and the denominator of Eq. (2), and was canceled out.

The effective solid angle of the photon detector is the product of the geometrical solid angle Ω_0, the total-energy peak efficiency $\epsilon(E)$ of the photon detector, and the attenuation factor $t(E)$ for photons of energy E in the transmission from the source to the NaI(Tl) crystal:

$$\Omega = \Omega_0 \epsilon(E) t(E)\,. \qquad (3)$$

The attenuation factors $t(E)$ for 1.2 mm of aluminum (the vacuum chamber port and the crystal packing) were calculated using the tables of Davisson,[12] and the total-energy peak efficiencies $\epsilon(E)$

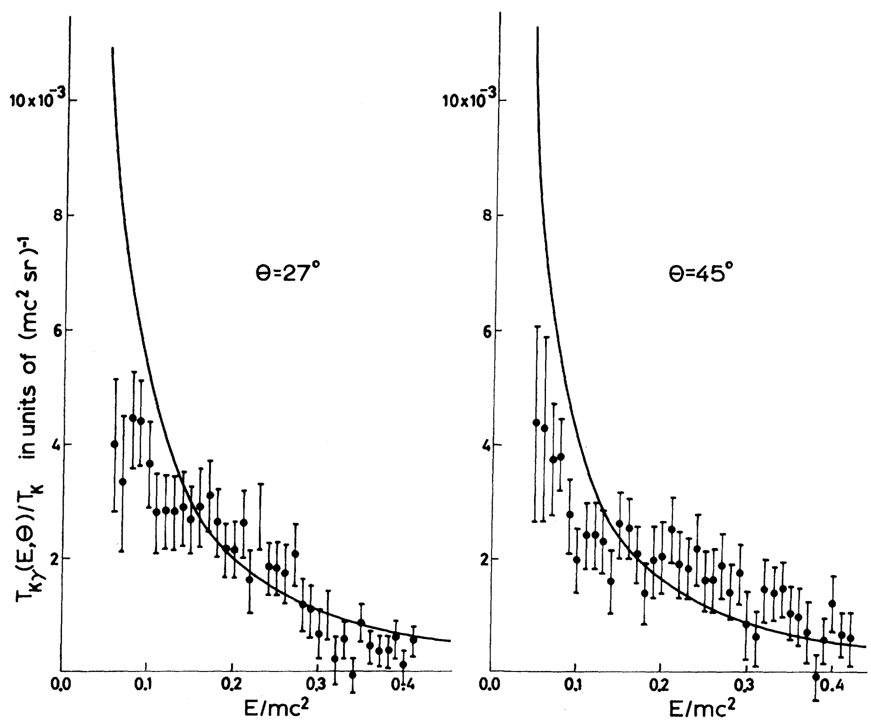

FIG. 2. Energy distributions in the $K\gamma$ process in ^{137}Ba as a function of the photon energy E. The solid lines were calculated from the Spruch-Goertzel theory, which was modified to take into account the finite angular resolution of the system.

were calculated using the tables of Yates.[13]

In our previous paper[11] the corrections for $t(E)\cdot\epsilon(E)$ were applied to the theoretical curve. It seems advantageous to apply these corrections to the experimental data. Therefore in Fig. 2 we again show the reported data obtained at the angle of 27°, together with the new data taken at 45°. In deriving these data two-dimensional spectra of electron- and photon-detector pulses in triple coincidence with K x rays were analyzed. The counting rates per channel were low. In order to improve statistics, counts in pairs of channels in the photon-detector line were summed. Then the energy interval ΔE had a value close to 0.01 mc^2. The background due to accidental coincidences was determined by summing the data in the neighboring channels, four to the right and four to the left from the peaks due to the $K\gamma$-process events. N_{Kx} was measured during the checks of the coincidence efficiency and gain stability of the apparatus, which were performed regularly twice a day. On an average N_{Kx} was 42 coincidences/sec. The geometrical solid angle of the photon detector was 0.065 sr.

To obtain the angular distribution, additional measurements at 0, 70, 100, 135, and 180° were performed. These measurements were not made in order to obtain the energy distribution, but rather to determine the transition probability for the $K\gamma$ process over a wider interval of energies. The data at 27 and 45°, and the data at 70, 100, and 135°, which were obtained as two-dimensional diagrams, were summed along the lines of constant sum-energy over a range of photon energies from 0.1–0.3 mc^2. When the background due to accidental coincidences was subtracted, we obtained the contribution due to the transition probability of the $K\gamma$ process, $T(K,\gamma; E=0.1–0.3\ mc^2, \Theta)$ integrated over energy, which we denote by $T_{K\gamma}(\Theta)$.

The data at 0 and 180° were analyzed in the same manner. At these angles, as mentioned earlier, no shielding could be placed between the electron and photon detector. Monoenergetic K conversion electrons from the source, which are always accompanied by the emission of K x rays, produce bremsstrahlung photons in the Si(Li) detector. If the bremsstrahlung photons can reach the photon detector, real triple coincidences are obtained. At the same time the reduced energy of the K conversion electron and the energy of the bremsstrahlung photon satisfy the energy condition (1). To determine the contribution of this process, which is indistinguishable from the $K\gamma$ double decay, one could calculate the energy and angular distribution of the external bremsstrahlung in the Si(Li) detector. We are presently not in a position to do such a calculation, since it would require a large capacity computer, the application of the Monte Carlo method, and a fair amount of time to obtain statistically satisfactory results.

We therefore estimated the contribution of the bremsstrahlung indirectly, using the reported experimental results of Buechner *et al.*[14] scaled down to the energy of K conversion electrons from ^{137}Ba (624 keV). To check this procedure and to determine the absolute scale, the bremsstrahlung from the Si(Li) detector was measured at scattering angles of 90, 117, and 145°. Buechner *et al.* measured the angular distribution of the emitted photons in the electron bremsstrahlung process for a thick aluminum target. They varied the energy of the incoming monoenergetic electron beam from 1250 to 2350 keV. The target thickness was adjusted to stop completely primary electrons. K conversion electrons from ^{137}Ba are also completely stopped in our Si(Li) detector.

We extrapolated their results to lower electron energies with linear functions that are expected to be valid for electrons above about 500 keV. It is evident from the results of Buechner *et al.*[14] that the contribution of the bremsstrahlung process is practically independent of the incoming electron energy at large angles. At lower energies the angular distribution becomes less forward peaked (elastic multiple scattering becomes predominant at these energies). The angular distribution of the bremsstrahlung photons is shown in Fig. 3 with the solid line.

Measurements of the bremsstrahlung in the Si(Li) detector were performed in the same arrangement as for the $K\gamma$ process, except that the lead shield between the electron and photon detector was removed and was placed between the source and the photon detector. In this manner, only the scattering from the electron into the photon counter was detected. The contributions of the external bremsstrahlung to the measured rates in the double-decay $K\gamma$ experiment at nominal angles $\Theta=0$ and 180° were then determined from Fig. 3.

Our results for the angular distribution in the $K\gamma$ process for photons of energy from 0.1 to 0.3 mc^2 are shown in Fig. 4 (full circles). Also shown are the results of Fuschini, Maroni, and Veronesi (FMV)[9] (open circles). The dotted line represents the theoretical distribution for the $K\gamma$ process for the same energy range, which proceeds as the internal Compton effect. The numerical values for the 662-keV $M4$ transition in ^{137}Ba were calculated from the expression given by Spruch and Goertzel.[5] The finite angular resolution of the detectors was taken into account.

Integrating our experimental results over the

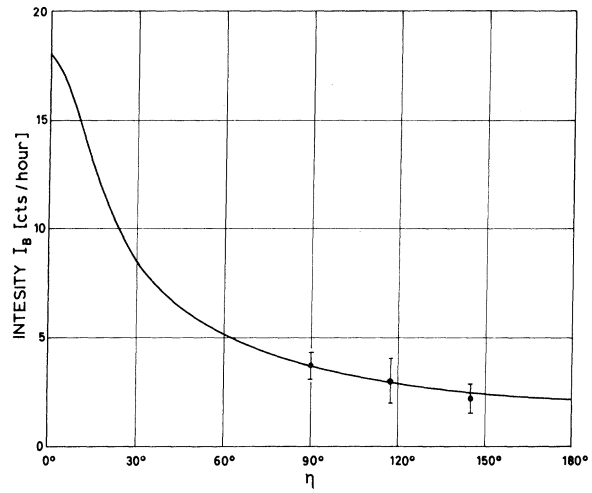

FIG. 3. Angular distribution of emitted photons produced in the bremsstrahlung process for a thick Si(Li) target. The solid line is obtained by extrapolating the experimental results of Buechner *et al.* to the incident electron energy of 624 keV. It was fitted to our experimental data, which are presented with full circles with the statistical errors. The angle η is defined as the angle between the directions of the incident electron beam and the emitted bremsstrahlung photons.

angles of emission, we obtained the total relative transition probability for the $K\gamma$ process for photons in the energy range from 0.1–0.3 mc^2

$$T_{K\gamma}/T_K = (2.2 \pm 0.2)\times 10^{-3}. \qquad (4)$$

This result is somewhat higher than the theoretical value of 1.7×10^{-3} predicted by Spruch and Goertzel and than the experimental value of $(1.7 \pm 0.2)\times 10^{-3}$ derived from the data of FMV.[9]

DISCUSSION

The angular distribution calculated from the Spruch and Goertzel theory is in a fair absolute agreement with the experimental data. At large angles the experimental results are above the theoretical values. Spruch and Goertzel have pointed out that their theory is not quite applicable in the case of ^{137}Ba because of the Born approximation they used.[15] This approximation is valid for low Z and high electron energy, and in this case it is not quite clear whether these requirements are fulfilled. This fact may be the main reason for the discrepancy between the theoretical and the experimental values.

Figure 2 shows that the same theory gives a good absolute fit to the measured energy distributions for energies above approximately 70 keV. At lower energies the experimental values fall below the theoretical predictions. Agreement at low energies is not to be expected, because the transition probabilities of Spruch and Goertzel diverge as the photon energy tends to zero. If this were the case the 662-keV level in ^{137}Ba would decay instantly with the emission of a K conversion electron accompanied by a long-wave photon. The di-

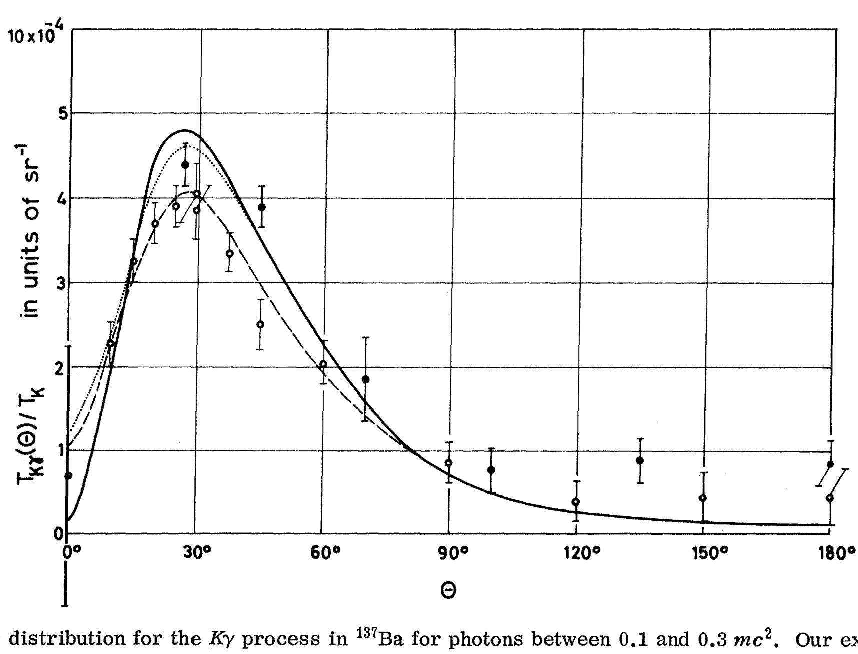

FIG. 4. Angular distribution for the $K\gamma$ process in ^{137}Ba for photons between 0.1 and 0.3 mc^2. Our experimental data are represented with full circles, and the results of Fuschini, Maroni, and Veronesi with open circles. The solid line represents the theoretical distribution for the point geometry calculated from the Spruch and Goertzel theory. The dotted and the dashed line show the same distribution modified to take into account the finite angular resolution of our and Fusčhini's, Maroni's, and Veronesi's system, respectively.

vergence of the theory is due to the fact that the calculations were made with only the lowest-order Feynman diagrams.

From the theory of Eichler one can estimate the contribution of the nuclear second-order mechanism to the $K\gamma$ process. The expressions for the energy spectra of the emitted particles for $\gamma\gamma$ and $K\gamma$ processes can be written in the form[3,4]

$$T_{\gamma\gamma}(E_2) = C_L[(W_0 - E_2)^{2L_1+1}E_2^{2L_2+1} + E_2^{2L_1+1}(W_0 - E_2)^{2L_2+1}], \quad (5)$$

$$T_{K\gamma}(E_2) = C_L[\alpha_K(\tau_1 L_1, E_1)(W_0 - E_2)^{2L_1+1}E_2^{2L_2+1} + \alpha_K(\tau_2 L_2, E_1)(W_0 - E_2)^{2L_2+1}E_2^{2L_1+1}]. \quad (6)$$

E_1 and E_2 are the energies and L_1 and L_2 denote the multipolarities of the two-step transition, $E_1 + E_2 = W_0$ is the total transition energy, and $L_1 + L_2 = L$ denotes the multipolarity of the direct transition. The $\alpha_K(\tau L, E)$ are the K-shell conversion coefficients, where τ designates the type of radiation (electric or magnetic multipole). C_L is a constant factor determined by the nuclear matrix elements of the multipole transition operators, by the transition multipolarity, and by the total transition energy.

In the case of ^{137}Ba, an estimate of the constant C_L can be made, based on the experimental evidence for the double-photon decay. Beusch has obtained a value of $(6.4 \pm 3.1) \times 10^{-6}$ for the ratio of double-photon decay $T_{\gamma\gamma}$ to single-photon decay T_γ, also for the 662-keV transition in ^{137}Ba. We assumed that one double-photon transition occurs per 10^5 single-photon transitions. The results, therefore, represent an upper limit. The energy spectra of photons for several combinations of multipolarities were calculated from the relation (5). When the distributions obtained in this way were integrated over energy and compared with

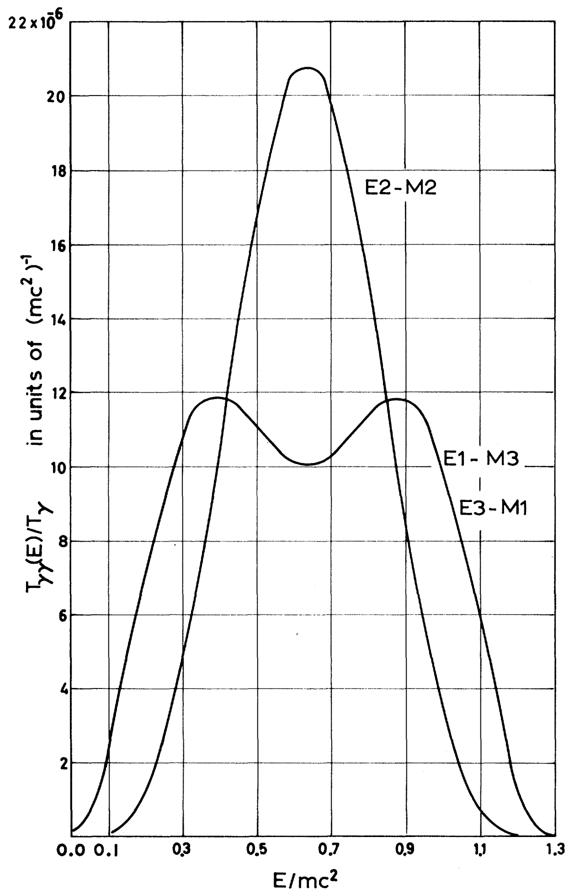

FIG. 5. The relative transition probabilities integrated over the angle for the $\gamma\gamma$ process in ^{137}Ba versus the photon energy E calculated from the theory of Grechukhin. The curves shown represent the most important combinations of multipoles. The ordinate scale was estimated from the experiment of Beusch (the results represent upper limits).

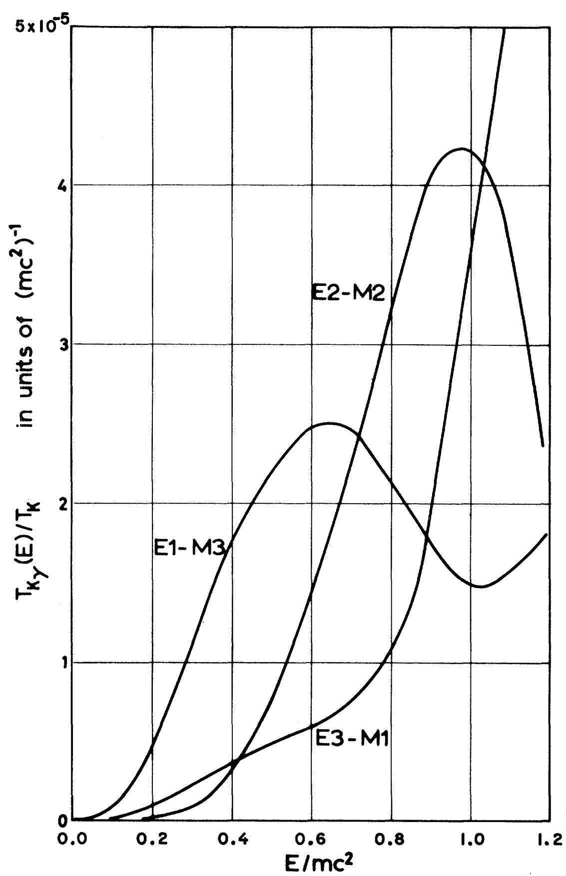

FIG. 6. The relative transition probabilities integrated over the angle for the $K\gamma$ process in ^{137}Ba versus the photon energy E, as calculated from Eichler's theory. The curves shown represent the most important combinations of multipoles of emitted photons and electrons. The ordinate scale was estimated from the experiment of Beusch (the results represent upper limits).

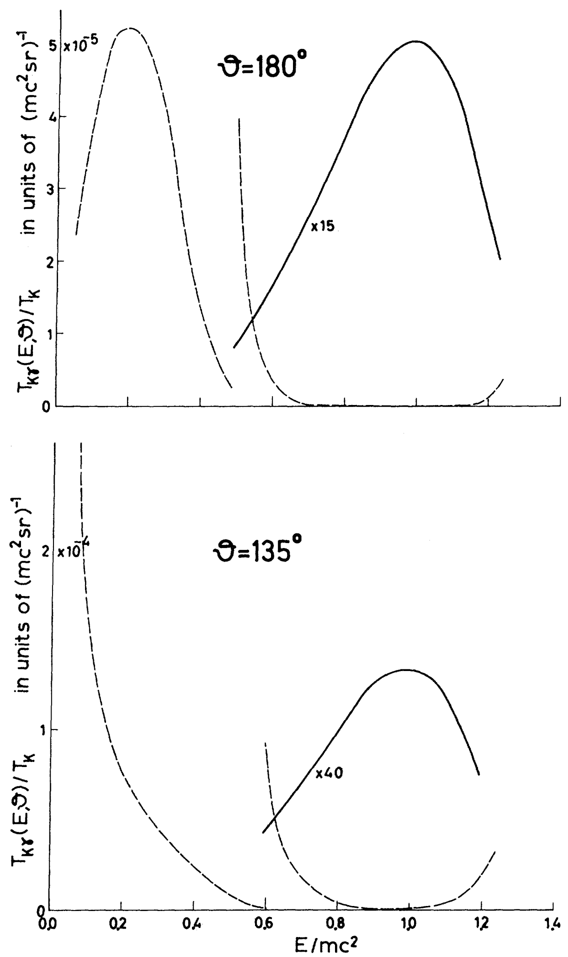

FIG. 7. Theoretical relative transition probabilities per unit solid angle and unit energy interval for the $K\gamma$ processes in ^{137}Ba versus the photon energy E at two angles of relative emission θ. The solid lines represent the upper limits of the contribution of the "nuclear" mechanism assuming the isotropic angular distribution. The $E2$-$M2$ combination of multipoles is taken into account. The theoretical distributions of the "electronic" mechanism, the internal Compton effect, are shown by dashed lines.

the result of Beusch, the constants C_L/T_γ for each combination of multipole orders were determined. With these values the over angle-integrated relative transition probabilities $T_{\gamma\gamma}(E)/T_\gamma$ were calculated. The results are shown in Fig. 5.

After dividing by α_K for the 662-keV state in ^{137}Ba,[16] the constants C_L in units of T_K were obtained: $C_L/T_K \leq 0.877\times 10^{-2}(mc^2)^{-11}$ for $E2$-$M2$ and $C_L/T_K \leq 0.422\times 10^{-2}(mc^2)^{-11}$ for $E1$-$M3$ and $E3$-$M1$. Then, using the values of the K conversion coefficients from the tables of Hager and Seltzer[17] the ratios $T_{K\gamma}(E)/T_K$ were calculated from the relation (6). The results are shown in Fig. 6. The energy distribution for the "nuclear" $K\gamma$ process should be observed for high-energy photons, i.e., low-energy electrons, in contrast with that for internal Compton effect. For the measured photon energy interval from $(0.1-0.3)mc^2$ we obtained the values $T_{K\gamma}/T_K \leq 2.8\times 10^{-6}$, 7.6×10^{-6}, and 10.2×10^{-6} for $E1$-$M3$, $E2$-$M2$, and $E3$-$M1$ combinations, respectively. These results show that the contribution of the "nuclear" part in the measured energy interval is several orders of magnitude smaller than the value predicted by Spruch and Goertzel or the value obtained in the present experiment. This fact strongly suggests that the $K\gamma$ process in the low-energy region of photons proceeds mostly through the internal Compton effect mechanism.

The theoretical energy spectra of the $K\gamma$ process of both mechanisms are compared in Fig. 7. For low photon energy the internal Compton effect plays a predominant role, while at higher energies the main contribution comes from the "nuclear" part of the $K\gamma$ process. Thus the measurement of the energy spectra can in principle distinguish between these two mechanisms. The transition probability for the "nuclear" part is very small, compared with the internal Compton effect, and could be resolved in a measurement of the energy distribution at large angles, where the contribution of the internal Compton effect is relatively small.

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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 $K, L+M+\cdots$ 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 $T_{K,L+M+\cdots}/T_K \leq 4.5 \times 10^{-4}$. 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 *M*4 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