from self-consistent calculations and are not necessarily identical with the actual energy spectra.

C. Correction Factors

The relative number of photon events recorded by the detector for the early and late spectra shown in Fig. 9 is given by

$$C_T = \sum_I \epsilon_I N_I,$$

where ϵ_I is the detection efficiency for the Ith energy bin. The correction factor was obtained by taking the ratio of the totals for the two spectra. The result is 1.142, comparing the early-time spectrum with the late. In applying this correction, it was assumed that for the time region 0.35–4.0 sec, there is no change in the γ-energy spectrum and therefore no correction. For the time domain 0.01–0.35 sec it was assumed that the detection efficiency changed linearly with time. From the above calculation, the resulting correction coefficient was -0.053%/msec. Thus, the correction applied to the 0.01-sec data was 18% and that applied to the 0.35-sec data was zero.

The error assigned to the correction was assumed to be $\pm 50\%$. This error was folded in with the other errors to obtain the error flags shown in Figs. 5 and 6.

PHYSICAL REVIEW VOLUME 187, NUMBER 4 20 NOVEMBER 1969

Energy-Distribution Measurement in the Double Decay of ^{137}Ba

A. Ljubičić, B. Hrastnik, K. Ilakovac, V. Knapp, and B. Vojnović
Institute "Ruer Bošković," Zagreb, Yugoslavia

(Received 28 January 1969)

The energy spectrum in the K-electron–photon double decay of the 662-keV state of ^{137}Ba has been measured at an angle of 27°. The experimental data for photon energies above 70 keV are in good agreement with the theory of Spruch and Goertzel, in which the process is treated in an approximation as the internal Compton effect. At lower energies a disagreement has been found. The contribution of the K-electron–photon double decay proceeding via nuclear intermediate states has been estimated on the basis of previous results on double γ decay, and has been found to be negligible.

INTRODUCTION

In addition to single-decay processes (γ-ray emission, electron conversion, and emission of an electron-positron pair), nuclear transitions can also proceed via double-decay processes.[1] These involve a simultaneous emission of a photon–conversion-electron pair, of two photons, or of two conversion electrons (at higher energies three other processes involving emission of electron-positron pairs are possible). In this paper we present a measurement of the double decay of the former type of the 622-keV state in ^{137}Ba. This type of double decay has previously been studied by several groups.[2–4] It can proceed via two mechanisms, which differ in whether the real photon is emitted by an intermediate virtual state of the electron or of the nucleus. Spruch and Goertzel[5] and Melikian[6] have developed a theory of the former mechanism, often called the internal Compton effect. Also, calculations of the latter mechanism have been made.[7] The measurements of the electron-photon angular correlation are in agreement with the theory of Spruch and Goertzel.[3,4,8] The present measurements of the photon energy distribution represent an independent check of the theory.

EXPERIMENT

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 approximately 1 mg/cm^2 thick. The external bremsstrahlung of K conversion electrons would yield triple coincidence events, which would be indistinguishable from the electron-photon double-decay events. Lindquist *et al.*[3] have investigated the contribution of this effect and have shown it to be negligible for a source of a similar thickness on an aluminium backing.

The experimental arrangement is shown in Fig. 1. The source was mounted in the center of a 22-cm-diam vacuum chamber. A Si(Li) detector 12 mm diam×0.9

[1] H. J. Leisi, in Proceedings of the Conference on the Electron Capture and Higher Order Processes in Nuclear Decay, Debrecen, 1968 (unpublished); E. L. Church and T. R. Gerholm, Phys. Rev. **143**, 879 (1966).
[2] H. B. Brown and R. Stump, Phys. Rev. **90**, 1061 (1953); P. Kleinheinz, L. Samuelsson, R. Vukanović, and K. Siegbahn, Nucl. Phys. **59**, 673 (1964).
[3] T. Lindquist, B. G. Pettersson, and K. Siegbahn, Nucl. Phys. **5**, 47 (1958).
[4] E. Fuschini, C. Maroni, and P. Veronesi, Nuovo Cimento **26**, 831 (1962); **41**, B252 (1966).
[5] L. Spruch and G. Goertzel, Phys. Rev. **94**, 1671 (1953).
[6] E. G. Melikian, Zh. Eksperim. i Teor. Fiz. **31**, 1088 (1956) [English transl.: Soviet Phys.—JETP **4**, 930 (1957)].
[7] J. Eichler, Z. Physik **160**, 333 (1960).
[8] B. G. Pettersson, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Co., Amsterdam, 1965), p. 1569.

mm thick, placed in the chamber at a distance of 5 cm from the source, was used to detect electrons. Its resolution for ^{137}Ba conversion electrons was 11 keV. Photons and K x rays were detected in NaI(Tl) scintillation crystals $\frac{3}{2}$ in. diam$\times\frac{3}{2}$ in. thick and $\frac{3}{2}$ in. diam$\times$3 mm thick, respectively, mounted on XP 1020 photomultipliers. The x-ray detector was placed close above the source in a recessed aluminium window. The photon detector was placed close to one of the thin aluminium windows that were mounted on the cylindrical wall of the vacuum chamber. To prevent the scattering from the electron into the photon detector, and reversely, lead shields were displaced in the vacuum chamber and outside it. The photomultiplier feeding voltages were supplied through bases made according to Dolan *et al.*[9] In a previous experiment the voltages were adjusted for best time and energy resolution.[10] The energy resolution of the photon detector was 7% for the 662-keV γ rays, and the resolution of the x-ray detector was about 12 keV for the 32-keV x rays of Ba.

A standard triple fast-slow coincidence system with a resolving time $2\tau=40$ nsec was used for gating the electron and photon pylses. These were analyzed in a 128$\times$128-channel analyzer. Pairs of channel numbers, corresponding to the amplitudes of the gated electron and photon detector pulses, were recorded on punched

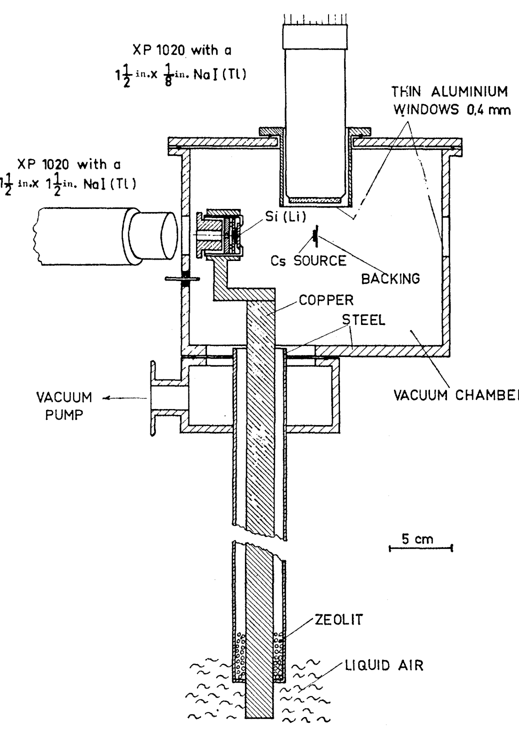

FIG. 1. Sketch of the experimental arrangement.

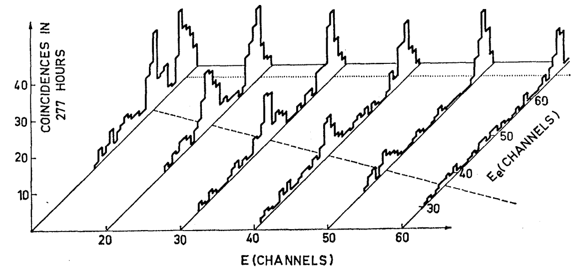

FIG. 2. Two-dimensional display of experimental data at 27°. The sections show numbers of counts obtained by adding counts in two E channels at channel numbers 10, 20, etc.

paper tape. The rate was approximately 60 triple coincidences per hour. The data on tape were analyzed in the CAE 9040 computer. The transition energy less the K-electron binding energy is shared between the emitted electron and the photon. The double-decay events, therefore, appear in a two-dimensional diagram as a continuous distribution along a straight line. The expected locus of double-decay events, based on the energy calibration of the detectors, is shown by the dashed line in Fig. 2. The counting rates obtained in the measurements are displayed for some sections of the two-dimensional diagrams as histograms. The peaks above the dashed line indicate the presence of the electron-photon double decay in ^{137}Ba. The peaks along the dotted line are due to coincident K conversion electrons and K x rays of Ba in accidental coincidence with pulses in the photon detector. This line was used to recheck the energy calibration of the electron detector.

The double coincidence rates of K conversion electrons and K x rays for the same transition were determined with the same experimental arrangement and adjustment, except that triple coincidence was changed to double coincidence, and the electron detector pulses, gated by the pulses from Ba K x rays, were analyzed in a 256-channel analyzer. This counting of K conversion-electron–x-ray coincidences was performed durthe daily checks of the coincidence efficiency and gain stability of the apparatus.

ANALYSIS OF RESULTS

Calculation of the absolute transition probabilities from the observed rates of double decay would require a determination of the source strength and efficiency of the detectors and of the solid angles. It is simpler to determine the ratio of the transition probability of double decay to the K conversion transition probability, which follows from the triple- and double-coincidence rates. This ratio is also best determined in theory.

We denote by $T_0(K, \gamma; E, \theta)$ the transition probability for the K-electron–photon double decay for point geometry. This probability is defined per unit energy interval of photons of energy E and per unit solid angle, at a relative angle θ between the emitted K electron and the photon. Similarly, we denote by $T(K, \gamma; E, \Theta)$

[9] K. W. Dolan, J. P. Hurley, and J. M. Mathiesen, Nucl. Instr. Methods **39**, 232 (1966).

[10] B. Hrastnik, A. Ljubičić, B. Vojnović, K. Ilakovac, and M. Jurčević, Fizika **1**, 127 (1969).

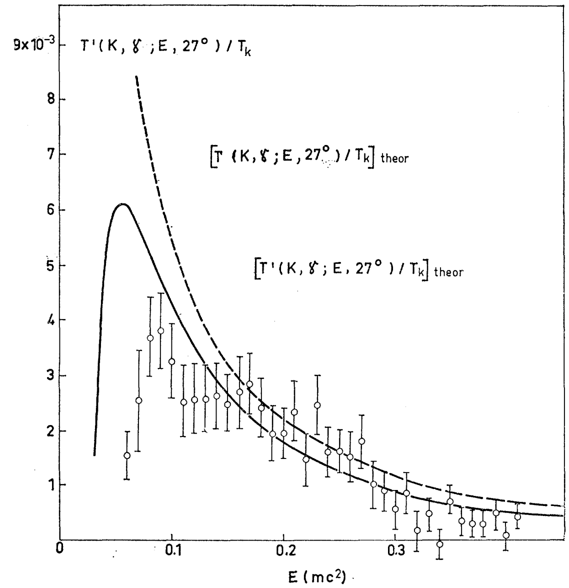

FIG. 3. Experimental data on the K-electron–photon double decay in ^{137}Ba as a function of the photon energy E. The dashed line represents the theoretical distribution for point geometry as given by Spruch and Goertzel. The solid line shows the same distribution, which was modified to take into account the experimental conditions.

the transition probability averaged over the solid angles Ω_e and Ω of the electron and photon detectors, respectively (efficiencies of the detection and requirements set by electronics and in the analysis are included). Θ is the nominal angle between the detectors. The triple coincidence counting rate is given by

$$N_{K\gamma\mathrm{x}}(E,\theta)=N(4\pi)^{-2}\Omega_e\Omega_\mathrm{x}\Omega T(K,\gamma;\,E,\theta)\Delta E,$$

where Ω_x is the x-ray–detector solid angle, and N is the number of radioactive atoms in the source. The averaging over the direction of emission of x rays does not appear, because the emission of K x rays is isotropic.

The double-coincidence rate of K conversion electrons and K x rays is given by

$$N_{K\mathrm{x}}=(4\pi)^{-1}\Omega_e(4\pi)^{-1}\Omega_\mathrm{x}T_KN,$$

where T_K is the K conversion transition probability. Therefore, we have

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

It is assumed that the triple- and double-coincidence efficiencies are equal. In the experiment they were both very close to 1.

Since the solid angles also include the efficiencies, one can write

$$\Omega=\Omega_0\epsilon(E)t(E),$$

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

The experimental data are presented without the corrections for ϵ and t as the ratio

$$T'(K,\gamma;\,E,\Theta)/T_K=N_{K\gamma\mathrm{x}}(E,\Theta)/N_{K\mathrm{x}}\Delta E\Omega_0.$$

Therefore, for comparison with theory, the theoretical results were modified to include the factors (in addition to the averaging over angles)

$$[T'(K,\gamma;\,E,\Theta)/T_K]_{\mathrm{theor}}=\epsilon t[T(K,\gamma;\,E,\Theta)/T_K]_{\mathrm{theor}}.$$

The photon and electron detectors were placed at a relative angle $\Theta=27°$, where the theoretical predictions of Spruch and Goertzel give a maximum transition probability.

The results of the measurements are shown in Fig. 3. The error bars represent the statistical errors of the measurement. Owing to the application of two-dimensional analysis, no distortions of the spectrum are encountered. The Compton-continuum part of the photon-detector response does not contribute to the spectrum under the dashed line in Fig. 2.

When the relative transition probabilities obtained in this experiment, corrected for the attenuation factor and the total-energy peak efficiency, were integrated over the energy interval 50–150 keV, we obtained a value of $(4.40\pm0.24)\times10^{-4}$ electron-photon double decays per K conversion electron per steradian at 27°. This is in agreement with the value of $(3.8\pm0.4)\times10^{-4}$ obtained by Fuschini *et al.*[4] at an angle of 30° between the electron and photon emission.

DISCUSSION

The electron-photon double decay proceeding via the virtual state of the electron can be most simply visualized as the internal bremsstrahlung of the K conversion electron. Measurements of the angular distribution[4] have shown that the theory of internal bremsstrahlung does not fit well the experimental data on the electron-photon double decay.

Spruch and Goertzel[5] developed a theory of the electron-photon double decay for magnetic transitions, regarding it as a two-step process in which a virtual photon is absorbed from the nucleus by a K electron and the real photon is emitted by the electron prior to or after that interaction. They calculated the "magnetic internal-Compton coefficients"—the ratio of the transition probability of the electron-photon double decay and of the K conversion transition probability. Because the same Born approximation was used for either process, a much simplified expression was obtained, which is independent of nuclear matrix elements. The inaccuracies due to the use of the Born approximation were also expected to cancel partially. The theory was expected to be valid for low Z and high transition energies. A similar theory, but for electric transitions, was developed by Melikian.[6]

Experiments of Lindquist *et al.*[3] and Fuschini *et al.*[4] on the angular distribution in the electron-photon double decay in ^{137}Ba have shown that the theory of Spruch and Goertzel fits the data very well.

The probabilities for the electron-photon double decay per K conversion electron and per unit solid angle and unit energy interval of the photon, obtained in the present measurement, are shown in Fig. 3. The theoretical probabilities as given by Spruch and Goertzel for point geometry are shown by the dashed line. The solid line represents the theoretical probabilities, in which the finite spread of angle was taken into account, and also the photon absorption (in 1.2 mm of Al, the vacuum chamber port and the NaI packing) and the total-energy peak-efficiency factors. Good absolute agreement is seen for photon energies in a range from approximately 70 to about 200 keV. The corresponding range of electron energies is 560–430 keV, which seems to be sufficiently high to justify the application of the Born approximation.

At energies below about 70 keV, a disagreement was found. The distribution of Spruch and Goertzel diverges as the photon energy is reduced to zero. This "infrared catastrophe" is a result of their calculation with only the first Feynman diagrams. The effect can be seen as a deviation from the measured spectrum up to about 70 keV. In the theory of electron bremsstrahlung, a similar "infrared catastrophe" is eliminated by taking into account the radiative correction terms to Coulomb scattering.[11] An improved theory which would include equivalent terms would most likely yield a distribution in accord with the experimental data.

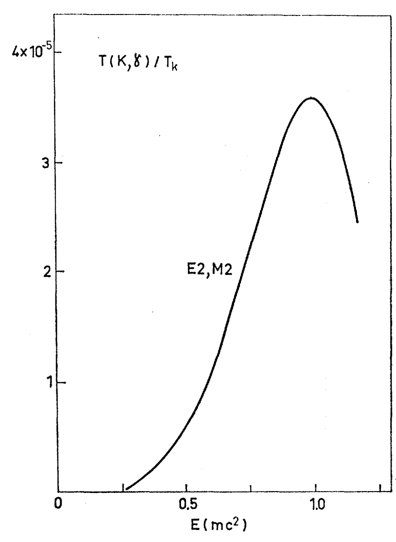

FIG. 4. Upper limit of the contribution of the intermediate nuclear states to the electron-photon double decay in ^{137}Ba.

[11] J. D. Bjorken and S. D. Drell, *Relativistic Quantum Mechanics* (McGraw-Hill Book Co., New York, 1964).

The electron-photon double decay can also proceed via nuclear intermediate states. If the double-γ-decay transition probabilities were known, a good estimate could be made,[7] since the conversion coefficients are well known. Theoretical estimates of the double-γ decay can only roughly be given.[12] An estimate of the upper limit for this process was made, however, based on the experimental evidence for the double-photon decay. Beusch[13] has obtained a value of $(6.4\pm3.1)\times10^{-6}$ for the ratio of double-photon decay to single-photon decay for the same transition in ^{137}Ba.

We made an estimate of the contribution of this mechanism by assuming that one double-photon decay occurs per 10^5 single-γ-ray decays. The results, therefore, represent an upper limit. The most likely combination of multipolarities was taken into account: the $E2$-$M2$ double decay. The energy spectrum of photons was calculated according to Grechukhin.[12] The electron-photon double-decay probabilities (integrated ever solid angle) per single-γ-ray decay and per unit energy interval of the photon were calculated by assuming that one of the photons was K converted. The Hager and Seltzer[14] tables of conversion coefficients were used in the calculation. After dividing by the K conversion coefficient of ^{137}Ba [$\alpha_K=0.0916$ (Ref. 15)], the angle-integrated transition probabilities per K conversion electron and per unit energy interval of the photon, shown in Fig. 4, were obtained. To compare these curves with the results of Fig. 3, one should divide the probabilities by 4π, if the electron-photon angular distribution is assumed to be isotropic.

The contribution of double-decay processes proceeding via nuclear intermediate states seems to be negligible in the case of the 662-keV $M4$ transition in ^{137}Ba. The process proceeds primarily via electronic intermediate states, i.e., as the internal Compton effect.

ACKNOWLEDGMENTS

We wish to thank Dr. M. Martinis and K. Pisk for helpful discussions. We also thank D. Protić and Mrs. Anica Hrastnik for the preparation of the Si(Li) counters used in the experiment.

[12] D. P. Grechukhin, Nucl. Phys. **35**, 98 (1962); **47**, 273 (1963); **62**, 273 (1965).

[13] W. Beusch, Helv. Phys. Acta **33**, 363 (1960).

[14] R. S. Hager and E. C. Seltzer, Nucl. Data **A4**, 1 (1968).

[15] H. H. Hansen, G. Lowenthal, A. Spernol, W. Van der Eijk, and R. Vaninbroukx, Z. Physik **218**, 25 (1969).