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First observation of the competitive double-gamma (“$\gamma\gamma/\gamma$”) decay process

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Abstract. First observation of the competitive double-γ decay process is presented. It is a second-order electromagnetic decay mode. The 662-keV decay transition from the $11/2^-$ isomer of ^{137}Ba to its ground state proceeds at a fraction of 2×10^{-6} by simultaneous emission of two γ quanta instead of one. The observed angular correlation and energy distribution of coincident γ quanta are well described by a dominant $M2 - E2$ and a minor $E3 - M1$ contribution to the double-γ decay branch. The data were well accounted for by a calculation using the Quasiparticle Phonon Model.

1 Introduction

Progress on nuclear science has been intimately related with our experimental knowledge and theoretical understanding of radioactivity. Radioactive decays occur due to all three unified forces in nature: the strong force, the weak force, and the electromagnetic interactions. First order radioactive decay processes mediated by these interactions are known to us in terms of radioactive α, β, or γ-decay by more then a century thanks to the pioneering works of Becquerel [1], Rutherford [2], Villard [3], and others.

Besides the well-known first-order radioactive decay processes, also second order decays can play important roles in nuclear physics or particle physics, despite the fact that their matrix elements are typically several orders of magnitude weaker than first order processes due to their proportionality to a higher power in the coupling constant. One example is the double-β decay ($\beta\beta$-decay) which corresponds to a simultaneous transformation of two neutrons (two protons) into two protons (two neutrons) and corresponding leptons, i.e., two positrons (two electrons) and two electron-neutrinos (two electron-antineutrinos), for conserving charge and lepton numbers. Double-β decay processes have first been considered theoretically by Maria Göppert some 80 years ago [4] and have later on been found to be responsible for the finite lifetimes of some even-even nuclei at the edge of the valley of stability, such as e.g., ^{76}Ge, ^{100}Mo, ^{116}Cd, ^{124}Sn, ^{128,130}Te, ^{136}Xe, or ^{150}Nd, whose single β-decays are forbidden by energy conservation. The first direct detection of $\beta\beta$-decay reactions was finally achieved in 1987 by Elliott, Hahn and Moe [5].

If neutrinos are their own anti-particles, then the neutrinos from a $\beta\beta$-decay reaction could annihilate themselves and redistribute their total energies to their reaction partners, hence leading to the possibility of neutrinoless double-β decay ($0\nu\beta\beta$-decay) reactions where besides the nuclear residue only two β particles appear in the final state. Searches for the existence of $0\nu\beta\beta$-decay processes are currently considered a promising opportunity to determine the particle character of neutrinos, i.e., to determine if a neutrino is its own anti-particle or not, and to measure its effective mass, which are two of the most pressing problems of contemporary particle physics.

The neutrino effective mass $\langle m_\nu \rangle$ could be extracted from a measurement of the $0\nu\beta\beta$-decay rate

$$\lambda_{0\nu\beta\beta} = G_{0\nu} |M^{(0\nu)}|^2 \left(\frac{\langle m_\nu \rangle}{m_e} \right)^2 \qquad (1)$$

if the nuclear matrix element ($M^{(0\nu)}$) for this process could be calculated sufficiently reliably from nuclear theory. $G_{0\nu}$ represents a known kinematical factor. $M^{(0\nu)}$ is formally a coherent sum of products of two first order transition matrix elements from the initial to the final state through infinitely many virtual intermediate states. While various predictions for the $0\nu\beta\beta$-NMEs are on the market, e.g., from Refs. [6–8], there is no way to test the accuracy of the theoretical calculations without a firm measurement of the $0\nu\beta\beta$-decay rate from Eq. (1) *and* pre-existing knowledge of the neutrino mass.

While $\beta\beta$-decay processes are nuclear reactions of second order in the electroweak interaction it is surprising to find that even less data exist for nuclear decay reactions that proceed in second order in the electromagnetic interaction where two γ-quanta are simultaneously emitted in a single quantum transition from one quantum state to another. In a $\gamma\gamma$-decay reaction both γ quanta with energies E_1 and E_2 share the total transition energy $E_0 = E_1 + E_2$, while the energy spectrum of the individual quanta is continuous, $0 < E_i < E_0$. These double-γ decay ($\gamma\gamma$-decay)

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reactions are formally analogous to $0\nu\beta\beta$-decay processes where in the latter two β-particles and in the former two γ-quanta appear in the final state and share the total transition energy. Indeed, $\gamma\gamma$-decay processes have first theoretically been postulated and studied by Maria Göppert in her PhD thesis [9] with Max Born in Göttingen, even before discussing $\beta\beta$-decay processes.

Up to recently, $\gamma\gamma$-decay reactions in nuclei were known, only, in three particular cases, ^{16}O [10, 11] and ^{40}Ca, ^{90}Zr [12, 13], where the first excited states of these even-even nuclei have spin and parity quantum numbers 0^+ and a single-γ decay is strictly forbidden by helicity conservation. Searches for the more general situation, in which $\gamma\gamma$-decay occurs in a nuclear transition which could proceed by a single-γ decay in competition, the so-called *competitive double-γ decay* ($\gamma\gamma/\gamma$-decay), was not reported in peer-reviewed literature before. The main obstacle in previous unsuccessful attempts for the experimental discovery of the $\gamma\gamma/\gamma$-decay has always been the background stemming from the presence of the typically much more likely and hence much more abundant single-γ decays, e.g. [13–16].

We will report here on the observation of the $\gamma\gamma/\gamma$-decay process. In particular, we have set out to firmly observe $\gamma\gamma/\gamma$-decay events, to study the energy distribution of the simultaneously emitted γ quanta and their angular correlation, to possibly determine their multipole order and radiation character, to measure the total double-γ–to–single-γ decay branching ratio, and thereby finally the partial $\gamma\gamma/\gamma$-decay rate. Our experimental results have initially been published elsewhere [17] and this conference contribution is predominantly based on the previously published material which at the time of the oral presentation has been under review, and on the material presented in the doctoral thesis of Dr. Christopher Walz [18]. The following section provides a brief description of the experimental set-up. Useful formulations with respect to the $\gamma\gamma/\gamma$-decay phenomenon will be compiled in Sec. 3. An account of the data analysis and the results will be given in Sec. 4. Then the data are confronted in Sec. 5 with theory estimates obtained within the Quasiparticle Phonon Model which was initially developed by Professor Dr. Vadim Georgievich Soloviev, to the memory of whom this conference is dedicated. Finally, we will provide a summary and an outlook.

2 Experiment

We have studied the γ radiation from a standard sealed ^{137}Cs radiation source with an activity of 16.3(5) μCi. The radioactive ^{137}Cs nuclei undergo β-decay reactions with a half-life of 30.07(3) years and populate to 5.3(2)% directly the stable $J^\pi = 3/2^+$ ground state of the daughter nucleus ^{137}Ba and to 94.7(2)% its $J^\pi = 11/2^-$ second excited state at an excitation energy of $E_0 = 661.659(3)$ keV [19]. This $11/2^-$ isomer has a half-life of 2.552(1) minutes. It decays to 89.86% by the emission of a 661.657(3) keV γ-ray line with multipolarity $M4$ and a reduced transition strength of $B(M4; 11/2^- \rightarrow 3/2^+) = 2.725(12)$ W.u. to the ground

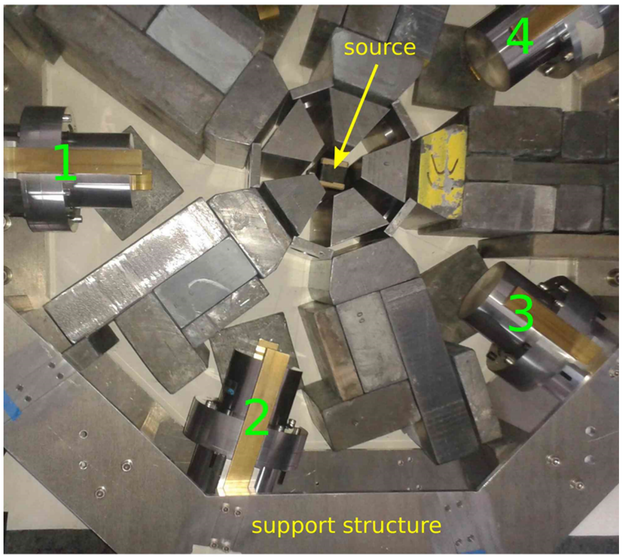

Figure 1. (Color online) Photograph of the experimental set-up with the anti-coincidence scintillator lid removed. Four LaBr detectors are visible and labeled 1 to 4. Heavy lead shielding reduces Compton-cross talk between the LaBr detectors.

state and with a γ-decay branching ratio of $1.12(9)\times10^{-7}$ [20] by an $E5$ transition to the $1/2^+$ first excited state of ^{137}Ba at 283.54 keV. The remaining decay intensity of the 662-keV state is converted with a conversion coefficient of $\alpha = 0.1124$ for the 662-keV $M4$ transition.

This γ-ray source was surrounded by a ring of five large-volume LaBr$_3$:Ce scintillators at a distance of approximately 22 cm to the detector face. Every two LaBr-detectors observed the radiation source at a relative mean angle of either 72° or 144°, respectively. Compton-scattering cross-talk between the detectors was suppressed by lead walls with a typical thickness of 12 cm. A photograph of the set-up is shown in Fig. 1. The entire set-up was covered by scintillator bars operated in anti-coincidence mode for the data acquisition system in order to suppress prompt coincidences between the LaBr detectors originating from shower events induced from energetic cosmic rays.

The absolute photo-peak efficiency of the LaBr-detector array was measured to ϵ_{abs}=1.50(5)% at 662 keV. An energy calibration point was continuously provided by 662-keV γ-ray line from the ^{137}Cs source. Additional data points for the energy calibration and for the definition of coincidence events were obtained from measurement runs with ^{60}Co γ-radiation standards. Single γ-ray spectra and $\gamma\gamma$-coincidence events were recorded for 1,273 hours corresponding to a total of 53 days of continuous data taking.

3 Formulation of the $\gamma\gamma/\gamma$-decay process

The total count rate $n_{\gamma\gamma/\gamma}$ of $\gamma\gamma/\gamma$-coincidence events is given by

$$n_{\gamma\gamma/\gamma} = A \cdot \frac{1}{\Gamma} \int_{4\pi} d\Omega \int_{4\pi} d\Omega' \int_0^{E_0} d\omega \frac{d^5\Gamma_{\gamma\gamma}}{d\omega d\Omega d\Omega'} \times \epsilon_{\text{intr}}(\omega, \Omega)\, \epsilon_{\text{intr}}(E_0 - \omega, \Omega') \quad (2)$$

where A is the activity of the source with respect to the $11/2^-$ state of ^{137}Ba at 662 keV, Γ is its total decay width, ω denotes the energy of one of the two simultaneously emitted γ quanta, $\Omega = (\theta, \phi)$ and Ω' are their angular directions, ϵ is the energy-dependent and angle-dependent absolute detection efficiency, and $\Gamma_{\gamma\gamma}$ denotes the partial decay width for double-γ decay. For an initially unoriented ensemble the quintuple-differential partial decay width in Eq. (2) can depend - besides on ω - only on the relative angle θ_{12} between the two γ quanta.

Following the work of Göppert and starting from Eqs. (A.15,A.34a) from Ref. [13] one can derive [17]

$$\frac{d^5\Gamma_{\gamma\gamma}}{d\omega d\Omega d\Omega'} = \frac{\omega\omega'}{96\pi^3} \sum_{\substack{JS'_1L'_1S_1L_1\\ S'_2L'_2S_2L_2}} P'_J(S'_1L'_1S_1L_1) \times P'_J(S'_2L'_2S_2L_2) \sum_l a_l^{J\xi} P_l(\cos\theta_{12}) \quad (3)$$

where ξ stands for a full set of parameters $\{S'_1L'_1S_1L_1S'_2L'_2S_2L_2\}$ specifying the parities and angular momenta of the two emitted photons with $S = 0$ for electric and $S = 1$ for magnetic transition characters. P_l are Legendre polynomials and $a_l^{J\xi}$ are further coefficients from angular momentum coupling and tabulated in the supplements of Ref. [17]. The functions

$$P'_J(S'L', SL, \omega'\omega) = (-1)^{S+S'} 2\pi(-1)^{I_i+I_f} \omega^L \omega'^{L'} \times \sqrt{2L+1}\sqrt{2L'+1}\sqrt{\frac{L+1}{L}}\sqrt{\frac{L'+1}{L'}} \times \frac{1}{(2L+1)!!(2L'+1)!!} \cdot \sum_n \left[\begin{Bmatrix} L & L' & J \\ I_f & I_i & I_n \end{Bmatrix} \times \frac{\langle I_f \| i^{L'-S'} M(S'L') \| I_n\rangle\langle I_n \| i^{L-S} M(SL) \| I_i\rangle}{E_n - E_0 + \omega} + (-1)^{L+L'+J} \times \begin{Bmatrix} L' & L & J \\ I_f & I_i & I_n \end{Bmatrix} \frac{\langle I_f \| i^{L-S} M(SL) \| I_n\rangle\langle I_n \| i^{L'-S'} M(S'L') \| I_i\rangle}{E_n - E_0 + \omega'} \right] \quad (4)$$

are generalised polarizabilities as given in Eq. (A.19) in Ref. [13]. They involve coherent sums of electromagnetic transition operators connecting the initial state I_i through virtual intermediate states I_n to the final ground state I_f by all combinations of multipoles L and L' that are possible by angular momentum coupling. It is convenient to define the unique-multipolar generalized polarizabilities

$$\alpha_{SLS'L'}(\omega) = \sum_n \frac{\langle I_f \| i^{L'-S'} M(S'L') \| I_n\rangle\langle I_n \| i^{L-S} M(SL) \| I_i\rangle}{E_n - E_0 + \omega} \quad (5)$$

that can be approximated for $E_n \gg E_0$ to a good accuracy by

$$\alpha_{SLS'L'} = \sum_n \frac{\langle I_f \| i^{L'-S'} M(S'L') \| I_n\rangle\langle I_n \| i^{L-S} M(SL) \| I_i\rangle}{E_n - E_0/2} \quad (6)$$

and can be calculated in standard nuclear structure models. One obtains a full account of the partial double-γ decay width, the relative energy distribution, and the angular correlation of the $\gamma\gamma/\gamma$-coincidences as a function of the sizes of the unique-multipolar generalized polarizabilities α. In the case of a uniquely multipolar dominance, the energy distribution of the individual γ-quanta depends on the involved multipolarities L and L' according to

$$P(\omega) \propto \left[\omega^{(2L+1)}(E_0-\omega)^{(2L'+1)} + \omega^{(2L'+1)}(E_0-\omega)^{(2L+1)}\right]. \quad (7)$$

This energy distribution is symmetric around $\omega = E_0/2$ and vanishes for the extreme values of $\omega = 0$ or E_0. For identical multipoles $L' = L$ it maximizes at $E_0/2$ while it can feature either a local minimum or a maximum at $E_0/2$ in case of different multipoles $L' \neq L$. If more than one unique-multipolar generalized polarizability contribute significantly, then the resulting energy distribution becomes a superposition including interferences according to Eqs. (3) and (4). From a combined analysis of the relative energy distribution of the simultaneously emitted γ quanta, their angular correlation, and the absolute value of the partial double-γ decay width $\Gamma_{\gamma\gamma}$ one can determine the unique-multipolar generalized polarizabilities α absolutely, apart from an irrelevant global sign.

4 Data analysis

The detected γ events that have not been vetoed by anti-coincidence with signals from the plastic detectors were analyzed with respect to their time difference. Clear signals of $\gamma\gamma$ coincidences were observed on a considerable background of random coincidences. Fig. 2 shows the time-difference spectrum for all detector combinations that share a mean relative angle of 72°. The prompt peak has a structure. It consists of an about 5 ns broad bump and a narrower peak with a width of about 2 ns. It has been verified that the broad bump is still present even when the ^{137}Cs source was removed. It most likely originates from non-vetoed showers from energetic cosmic radiation. The

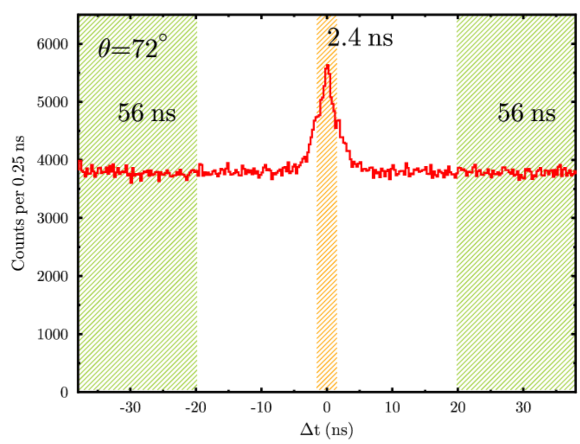

Figure 2. (Color online) Distribution of time differences of events registered in either two LaBr-detectors with a relative mean observation angle of 72°. The shaded areas represent the time gates used to obtain the sum-energy spectra shown in Fig. 3.

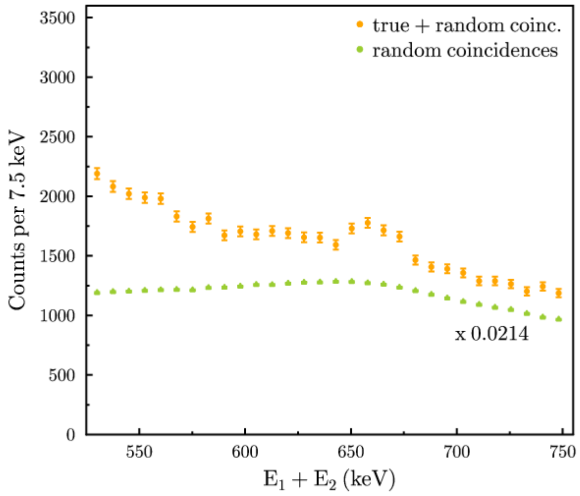

Figure 3. (Color online) Histograms of events gated with the time differences indicated by the shaded areas in Fig. 2 and plotted as a function of the sum of the energies registered in both detectors. The histograms have been scaled by the relative widths of their time gates. The error bars indicate the statistical uncertainties for each channel.

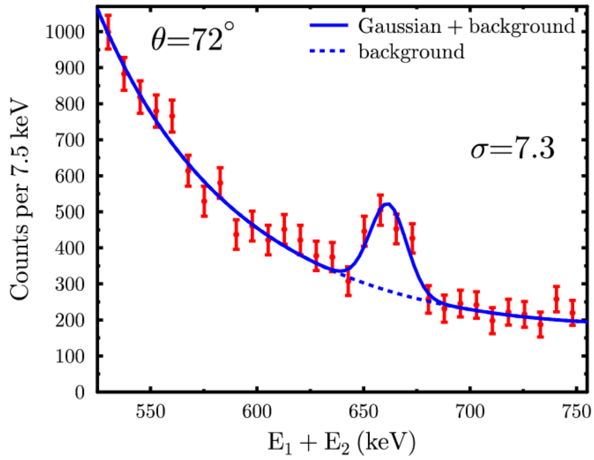

Figure 4. (Color online) Sum-energy histogram for events recorded with LaBr-detectors located at a mean relative angle of 72° after subtraction of random background as indicated in Figs. 2 and 3. For further reduction of background it has been required that neither single-γ energy was smaller than 181 keV (or larger than 481 keV, correspondingly). The error bars indicate the statistical uncertainties for each channel.

narrow peak is expected to contain shower background and background from Compton scattering of 662-keV single-γ decays along with the sought-after $\gamma\gamma/\gamma$ events.

Figure 3 shows the sum-energy spectrum of all pairs of LaBr-detectors with a mean relative angle of 72°, once gated on the narrow prompt time peak as indicated in Fig. 2 and once with an appropriately scaled time gate on random background, only. A peaked excess of counts at a sum energy of about 662 keV is clearly visible.

The random coincidences can be subtracted from the prompt coincidences. The resulting background-subtracted sum-energy spectrum is shown in Fig. 4. A significant sum-energy peak is obtained above a background which otherwise smoothly decreases as a function of energy. When fitted with a Gaussian function consistent with the calibrated detector response above a smooth phenomenological background, as indicated by the solid and dotted curves in Fig. 4, we obtain a peak area of 693(95) counts with a centroid at 661.6 ± 1.6 keV. The latter value is in very good agreement with the $11/2^- \rightarrow 3/2^+_{gs}$ transition energy E_0 = 661.66 keV, albeit obtained here from the sum of two γ-ray energies of less than 481 keV, each. The statistical significance of this signal amounts to 7.3 standard deviations, substantially exceeding the required significance for a three-star discovery in particle physics. From a detailed analysis of the time difference spectrum obtained by gating on the sum-energy events rather then on the time differences, we can firmly exclude [17] that the sum-energy peak at 662 keV results from Compton scattering of a single 662-keV γ quantum from one LaBr-detector to another because of the additional time delay of $\Delta t > 0.8$ ns such a process would induce. This information fully proves that the sum-energy peak must originate from the simultaneous emission of two γ-quanta that share the total transition energy, thereby firmly establishing the discovery of the $\gamma\gamma/\gamma$-decay process.

For the group of detector pairs with relative mean observation angle of 144° a corresponding peak area of 307(78) counts has been observed at a centroid of 664.2 ± 2.8 keV. The strong count rate asymmetry for both detector groups points at a pronounced anisotropy of the $\gamma\gamma/\gamma$-angular correlation function.

For a quantitative analysis of the $\gamma\gamma$-angular distribution and of the distribution of energy amongst the simultaneously emitted γ quanta, we restrict ourselves to the expected two dominant multipole channels: $M2-E2$ and $E3-M1$. Under this restriction Eq. (3) simplifies to

$$\frac{d^5\Gamma_{\gamma\gamma}}{d\omega d\Omega d\Omega'} = A_{qq}(\alpha^2_{M2E2}) + A_{od}(\alpha^2_{E3M1}) + A_x(\alpha_{M2E2}\cdot\alpha_{E3M1}) \quad (8)$$

which only depends on the absolute values of the two unique-multipolar generalized polarizabilities α_{M2E2} and α_{E3M1} and on their relative sign. Other than that the quantities A for the quadrupole-quadrupole, the octupole-dipole and the interference terms are known [17] functions of the correlation angle θ_{12} according to the formulae from Sec. 3.

From a fit of Eq. (8) to our data on the absolute angular distribution (2 absolute data points when having taken into account the known total decay rate of the $11/2^-$ isomer) and on the energy distribution (7 data points) we obtain [17] the following unique-multipolar generalized polarizabilities

$$\alpha_{M2E2} = +33.9 \pm 2.8\ \frac{e^2\mathrm{fm}^4}{\mathrm{MeV}} \quad (9)$$

and

$$\alpha_{E3M1} = +10.1 \pm 4.2\ \frac{e^2\mathrm{fm}^4}{\mathrm{MeV}}\ . \quad (10)$$

Apparently, the $M2-E2$ double-γ decay mode dominates over an alternative $E3-M1$ decay which however contributes constructively to the $\gamma\gamma/\gamma$-decay of the $11/2^-$ isomer of ^{137}Ba. The total branching ratio into the $\gamma\gamma/\gamma$-decay branch amounts to [17]

$$\Gamma_{\gamma\gamma}/\Gamma_\gamma = 2.05(37) \times 10^{-6} \,. \tag{11}$$

5 Comparison to nuclear model calculations

We have applied the Quasiparticle Phonon Model [21] which has been developed by Professor Soloviev. This conference commemorates Professor Soloviev and his numerous scientific achievements in nuclear and particle physics. The QPM provides a rather satisfactory description of the structure of ^{137}Ba and was used here [17] for the first time for calculating unique-multipolar generalized polarizabilities. The calculation is confronted with the data in Table 1.

Table 1. Measured values of the competitive-$\gamma\gamma$ branching ratio and α_{M2E2}, α_{E3M1} coefficients and comparison to theory. The uncertainties include the statistical error from the fit (±1 standard deviation) and systematic contributions.

	exp	QPM
$\Gamma_{\gamma\gamma}/\Gamma_\gamma$ (10^{-6})	2.05(37)	2.69
α_{M2E2} (e^2 fm^4 MeV^{-1})	+33.9(2.8)	+42.60
α_{E3M1} (e^2fm^{4}MeV^{-1})	+10.1(4.2)	+9.50

The structures of the $3/2^+$ ground state and of the $11/2^-$ isomer of ^{137}Ba are dominated by neutron holes in the $\nu(2d_{3/2})$ and $\nu(1h_{11/2})$ orbitals outside the semi-magic core ^{138}Ba. The calculated dominance of the $M2-E2$ contribution to the $\gamma\gamma/\gamma$-branching ratio can be understood microscopically from the dominant contribution of the first excited $7/2^+$ state of ^{137}Ba at an excitation energy of 1252 keV [19]. Its structure is calculated as a superposition of a neutron hole in the $\nu(1g_{7/2})$ orbital outside the semi-magic core ^{138}Ba and the 2^+ core excitation at 1436 keV coupled to the $\nu(2d^{-1}_{3/2})$ ground state configuration of ^{137}Ba. Consequently, there exists a sizeable $M2$ matrix element between the $11/2^-$ isomer and this $7/2^+_1$ state of ^{137}Ba with a strength of the order of a single-particle transition. At the same time there exists a sizeable $E2$ matrix element from the $7/2^+_1$ state to the ground state of ^{137}Ba corresponding to the annihilation of the quadrupole core excitation. Since, in addition, the energy denominator is small the virtual contribution of the $7/2^+_1$ state dominates the α_{M2E2} coefficient and enhances it sufficiently such that it dominates the $\gamma\gamma/\gamma$-decay intensity to about 80%.

6 Summary

We have presented the first observation for the competitive double-γ decay process, which can be considered a new phenomenon of radioactive decay. This discovery was made on the 662-keV decay transition from the $11/2^-$ isomer of ^{137}Ba to its ground state. The $\gamma\gamma/\gamma$-branching ratio was found to be on the order of 10^{-6}. The observation became possible due to the employment of large-volume LaBr-detectors that feature a high detection efficiency, reasonable energy resolution together with good time resolution. Thanks to the time resolution it was possible to demonstrate that the peaks in the coincident background-subtracted sum-energy spectra cannot originate from sequential Compton scattering between the detectors which has been the main experimental obstacle in previous attempts for observing the $\gamma\gamma/\gamma$-branching ratio. The observed angular distribution and energy distribution of coincident γ quanta provide evidence for dominant $M2-E2$ and a minor $E3-M1$ contribution to the double-γ decay branch in competition to the $M4$ single-γ transition. The data were well accounted for by a calculation using the Quasiparticle Phonon Model.

7 Outlook

As discussed above, the phenomenon of double-γ decay gives experimental access to generalized nuclear polarizabilities, nuclear observables that have been studied very little up to now, and only in the dipole-dipole sector. Our observation of the $\gamma\gamma/\gamma$-decay demonstrates the feasability to study generalized nuclear polarizabilities in a broader scope and potentially not limited to the particular decays of even-even nuclei with a first excited 0^+ state. Still, due to the long measurement time and the significant difficulties in reducing the background it remains to be seen how these measurements can develop.

Furthermore, theoretical work is needed for clarifying the significance of the generalized nuclear polarizabilities for the advance of physics. One obvious route of studies may address the relation of the generalized nuclear polarizabilities with the dipole polarizability of the nuclear ground state which itself has been shown to be related to the symmetry energy parameters of the nuclear equation state. It would also be very desirable to clarify if the nuclear models that are used to predict the $M^{(0\nu)}$ NMEs of $0\nu\beta\beta$-decay are capable of quantitatively describe the formally analogous generalized nuclear polarizabilities that in contrast to the $M^{(0\nu)}$ NMEs are experimentally accessible nuclear observables. The recent progress nurtures hope that a new sub-field of γ-ray spectroscopy may open up.

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