Experiments on the Competitive Double-Gamma ($\gamma\gamma/\gamma$) Decay

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Abstract. The recent 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 has been studied. The emission of a single 662-keV γ quantum is a factor of 5×10^5 more likely than the simultaneous emission of two γ quanta instead of one. The observed angular correlation and energy distribution of these rare coincident double-γ quanta are well described by a dominant $M2 - E2$ and a sub-dominant $E3 - M1$ contribution to the double-γ decay branch.

1. Introduction

The Conference on Neutrino and Nuclear Physics addresses progress and challenges at the common frontier of these scientific sub-disciplines. Several contributions address the rare nuclear double-β decay process because of the paramount importance of the pending observation of its neutrinoless variant for our understanding of lepto-genesis and of the particle character and the mass of the neutrino.

Double-β decay processes (with the emission of two neutrinos) have first been considered theoretically by Maria Göppert some 80 years ago [1]. They have later on been found to be responsible for the finite lifetimes of some even-even nuclei at the edges 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 $2\nu\beta\beta$-decay reactions was finally achieved by Elliott, Hahn and Moe [2] not earlier than 1987.

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 decays 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 (speak: '*double-gamma decay*') 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 $\gamma\gamma$-decays 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 [3] 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 [4, 5, 6] and ^{40}Ca, ^{90}Zr [5, 7], 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 $\gamma\gamma$-decays occur 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 our recent observation [11]. The main obstacle in previous unsuccessful attempts [5, 8, 9, 10] for experimental discovery of the $\gamma\gamma/\gamma$-decay has been the low statistics and the background from much more abundant single-γ decays.

It is the purpose of this contribution to bring our observation to the attention of this cross-disciplinary audience. It is based on our previous publications [11, 12, 13] and on the material presented in the doctoral thesis of Dr. Christopher Walz [14].

2. Experiment

The γ radiation from a standard sealed ^{137}Cs radiation source with an activity of 16.3(5) μCi has been studied. 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 [15]. This $11/2^-$ isomer has a halflife 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 $B(M4; 11/2^- \to 3/2^+) = 2.725(12)$ W.u. to the ground state and with a γ-decay branching ratio of $1.12(9)\times10^{-7}$ [16] 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°. Compton-scattering cross-talk between the detectors was suppressed by lead walls with a typical thickness of 12 cm. 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 $\epsilon_{\text{abs}} = 1.50(5)\%$ at 662 keV. An energy calibration point was continuously provided by the 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.

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 prompt $\gamma\gamma$ coincidences were observed on a considerable background of random coincidences. The latter were subtracted from the prompt coincidences by appropriate time gates. The resulting background-subtracted sum-energy spectrum obtained with the detectors at a relative observation angle of 72° is shown in figure 1. A significant sum-energy peak was obtained above a background which otherwise smoothly decreases as a function of energy. A Gaussian function consistent with the calibrated detector response was fitted to the peak above a smooth phenomenological background, as indicated by the solid and dotted curves in figure 1. A peak area of 693(95) counts with a centroid at 661.6 ± 1.6 keV was obtained from the fit. The position is in very good agreement with the $11/2^- \to 3/2^+_{\text{gs}}$ transition energy of $E_0 = 661.66$ 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.

A detailed analysis of the time difference spectrum obtained by gating on the sum-energy events rather than on the time differences, has firmly excluded [11] that the sum-energy peak at
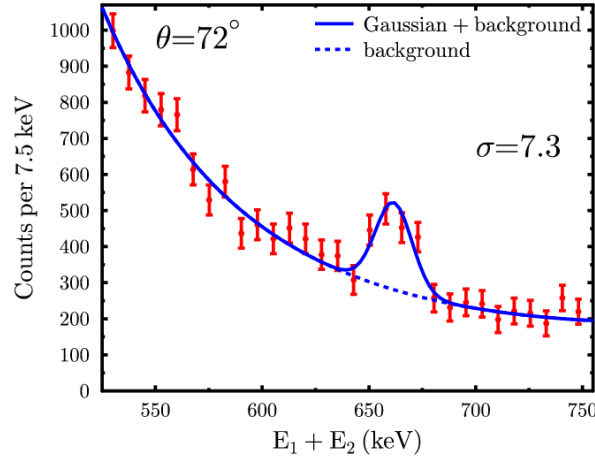
Figure 1. Sum-energy histogram for events recorded with LaBr-detectors located at a mean relative angle of $72°$ after subtraction of random background. For further reduction of background it has been required that neither single-γ energy was smaller than 181 keV (or larger than 481 keV, correspondingly). The error bars indicate the statistical uncertainties for each channel.

662 keV resulted 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 had originated 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.

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

$$n_{\gamma\gamma/\gamma} = A \cdot \frac{1}{\Gamma} \int_{4\pi} d\Omega \int_{4\pi} d\Omega' \int_0^{E_0} d\omega \frac{d^5\Gamma_{\gamma\gamma}}{d\omega d\Omega d\Omega'} \ \epsilon(\omega, \Omega) \ \epsilon(E_0 - \omega, \Omega') \tag{1}$$

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 equation (1) 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 equations (A.15, A.34a) from Ref. [5] one can derive [11]

$$\frac{d^5\Gamma_{\gamma\gamma}}{d\omega d\Omega d\Omega'} = \frac{\omega\omega'}{96\pi^3} \sum_{\substack{JS_1'L_1'S_1L_1 \\ S_2'L_2'S_2L_2}} P_J'(S_1'L_1'S_1L_1)P_J'(S_2'L_2'S_2L_2) \sum_l a_l^{J\xi} P_l(\cos\theta_{12}) \tag{2}$$

where ξ stands for a full set of parameters $\{S_1' L_1' S_1 L_1 S_2' L_2' S_2 L_2\}$ 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. [11]. The functions

$$
\begin{aligned}
P_J'(S'L', SL, \omega'\omega) &= (-1)^{S+S'} 2\pi (-1)^{I_i + I_f} \omega^L \omega'^{L'} \cdot \frac{\sqrt{(2L+1)(2L'+1)}}{(2L+1)!!(2L'+1)!!} \sqrt{\frac{L+1}{L}} \sqrt{\frac{L'+1}{L'}} \cdot \\
&\sum_n \left[\begin{Bmatrix} L & L' & J \\ I_f & I_i & I_n \end{Bmatrix} \frac{\langle I_f || i^{L'-S'} M(S'L') || I_n \rangle \langle I_n || i^{L-S} M(SL) || I_i \rangle}{E_n - E_0 + \omega} \right. \\
&\left. + (-1)^{L+L'+J} \cdot \begin{Bmatrix} L' & L & J \\ I_f & I_i & I_n \end{Bmatrix} \frac{\langle I_f || i^{L-S} M(SL) || I_n \rangle \langle I_n || i^{L'-S'} M(S'L') || I_i \rangle}{E_n - E_0 + \omega'} \right] .
\end{aligned}
\tag{3}
$$

are generalised polarizabilities as given in equation (A.19) in Ref. [5]. They involve coherent sums of electromagnetic transition operators connecting the initial state I_i through virtual intermediate states I_n to the final 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

$$
\alpha_{SLS'L'}(\omega) = \sum_n \frac{\langle I_f || i^{L'-S'} M(S'L') || I_n \rangle \langle I_n || i^{L-S} M(SL) || I_i \rangle}{E_n - E_i + \omega} .
\tag{4}
$$

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

$$
\alpha_{SLS'L'} = \sum_n \frac{\langle I_f || i^{L'-S'} M(S'L') || I_n \rangle \langle I_n || i^{L-S} M(SL) || I_i \rangle}{E_n - E_0/2}.
\tag{5}
$$

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 unique multipolar dominance, the energy distribution of the individual γ-quanta depends on the involved multipolarities L and L' according to

$$
P(\omega) \propto \left[\omega^{(2L+1)}(E_0 - \omega)^{(2L'+1)} + \omega^{(2L'+1)}(E_0 - \omega)^{(2L+1)} \right] .
\tag{6}
$$

This energy distribution is symmetric around $\omega = E_0/2$ and vanishes for the extreme values of $\omega = 0$ or E_0. If more than one unique-multipolar generalized polarizability contribute significantly, then the resulting energy distribution becomes a superposition including interferences according to equations (2) and (3). 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. Quantitative data analysis

A quantitative analysis of the $\gamma\gamma$-angular distribution and of the distribution of energy amongst the simultaneously emitted γ quanta was restricted to the expected two dominant multipole channels: $M2 - E2$ and $E3 - M1$. Under this restriction equation (2) simplifies to

$$
\frac{d^5 \Gamma_{\gamma\gamma}}{d\omega d\Omega d\Omega'} = A_{qq}(\alpha_{M2E2}^2) + A_{od}(\alpha_{E3M1}^2) + A_x(\alpha_{M2E2} \cdot \alpha_{E3M1})
\tag{7}
$$

which only depends on the absolute values of the two unique-multipolar generalized polarizabilities α_{M2E2} and α_{E3M1} and on their relative sign. Apart from that, the quadrupole-quadrupole A_{qq}, the octupole-dipole A_{od} and their interference A_x terms are known [11] functions of the correlation angle θ_{12} according to the formulae from section 3.

From a fit of equation (7) to the 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) the following unique-multipolar generalized polarizabilities were obtained [11]

$$\alpha_{M2E2} = +33.9 \pm 2.8 \ \frac{e^2 \text{fm}^4}{\text{MeV}} \tag{8}$$

and

$$\alpha_{E3M1} = +10.1 \pm 4.2 \ \frac{e^2 \text{fm}^4}{\text{MeV}} \ . \tag{9}$$

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 [11]

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

The Quasiparticle Phonon Model [17] provides a rather satisfactory description of the structure of ^{137}Ba and was used in Ref. [11] for the first time for calculating unique-multipolar generalized polarizabilities. 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 [15]. 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 $\nu(2d_{3/2}^{-1})$ 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%.

5. Summary

The first observation for the competitive double-γ decay process has been presented. 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. Thanks to their good time resolution it was possible to demonstrate that the peaks in the coincident background-subtracted sum-energy spectra could not 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 studied double-γ decay branch in competition to the 662-keV $M4$ single-γ transition in ^{137}Ba. The data were well accounted for by a calculation using the Quasiparticle Phonon Model.

6. 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. We are looking into developing more efficient experimental approaches for the detection of $\gamma\gamma/\gamma$-decay reactions. 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 of state. It would also be desirable to clarify whether the nuclear models that are used to predict the $M^{(0\nu)}$ NMEs of $0\nu\beta\beta$-decay are capable of quantitatively describing 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.

7. Acknowledgements

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