A significant experimental and theoretical research program is ongoing worldwide on second-order nuclear transitions, recently triggered by the observation of the competitive double-$\gamma$ decay in $^{137}$Ba. Here we unambiguously confirm this discovery with an improved value on the double-photon versus single-photon branching ratio as $2.62 \times 10^{-6}$ (30). Our results, now covering a larger angular range, contradict conclusions from the discovery experiment, where the decay was interpreted to be dominated by a quadrupole-quadrupole component. Here, we find a substantial enhancement in the energy distribution consistent with a dominating octupole-dipole character and a rather small quadrupole-quadrupole element in the decay. We investigate these results in terms of both the quasiparticle phonon model built on energy-density functionals and the Monte Carlo shell model. We find that both these approaches give a consistent and satisfactory reproduction of the octupole-dipole coupling, while to explain the small quadrupole-quadrupole contribution an evolution of the internal nuclear structure with increasing valence protons relative to the magic tin chain is needed. This type of decay is sensitive both to details regarding spectral structures originating from high-energy electromagnetic strength as well as changes in internal nuclear structure at low excitation energy. Thus, this type of process is simultaneously sensitive to traditionally separated phenomena of nuclear structure physics and have an important impact on nuclear structure research by high precision spectroscopy.

Polarizability is a fundamental concept in physics and chemistry defined from the principles of electromagnetic interaction. It describes how applied electric or magnetic fields induce an electric or magnetic dipole, or higher-order multipole, moment in the matter under investigation. In nuclear physics, the simple concept of polarizability influences observables over a broad range of topics. For example, the static dipole polarisation of the shape of the ground and excited states in atomic nuclei is influenced by the coupling to high-energy collective modes like the giant dipole resonance (GDR) via virtual excitations. In this case the nuclear static dipole polarizability, $\alpha_{d}$, is obtained from the photonuclear population of excited states,

$$\alpha_{d;E1} = 2e \sum_n \frac{|\langle I_0 | E1 | I_n \rangle|^2}{E_n - E_0},$$

where the transition matrix elements of the wave functions correspond to the electric dipole transition, $E1$, between the ground state, $I_0$, and an excited state, $I_n$, with $e$ the elementary unit charge and $E_n$ the energy of the state.

By expanding the concept of polarizability beyond the scalar case, one can divide the polarizability tensor into separate components. Typically, these are either spatial components like the birefringence properties of crystals or electric and magnetic multipole components. Within the nuclear structure framework, this type of off-diagonal polarizabilities can appear in very weak second order processes. In the electromagnetic case, the off-diagonal nuclear polarizability can be defined analogous to equation (1) in terms of either electric and magnetic components, or
states, Due to the parity conserving properties of the strong force, these decays can only be observed between two different components of different multipolarities as

$$\alpha_{M2E2} = \sum_n \frac{\langle I_i||E2||I_n \rangle \langle I_n||M2||I_i \rangle}{E_n - \omega}$$  \hspace{1cm} (2)$$

or,

$$\alpha_{E3M1} = \sum_n \frac{\langle I_i||M1||I_n \rangle \langle I_n||E3||I_i \rangle}{E_n - \omega}.$$  \hspace{1cm} (3)

Due to the parity conserving properties of the strong force, these decays can only be observed between two different states, $I_i$ and $I_f$. In the definition above, the denominator depends on the interference frequency, $\omega$, of the emitted $\gamma$ rays and is assumed to be half of the initial state energy. This type of second-order electromagnetic processes of atoms was first discussed in the doctoral dissertation of Maria Göppert-Meyer\textsuperscript{3} where she estimated a probability for an atomic two-photon absorption process relative to the single-photon process to be approximately $10^{-7}$, first detected in CaF\textsubscript{2}:Eu\textsuperscript{2+} crystals.\textsuperscript{9} The work of Göppert-Meyer was later expanded also to cover second-order decay in weak interaction processes, known as double $\beta$ decay.\textsuperscript{5} Here a significant theoretical and experimental effort has been carried out in the last decade to measure the process and answer two of the major unanswered questions in physics: is the neutrino its own anti-particle and what is its mass? Thus, the nature of the double-$\gamma$ decay ties intimately to the double-$\beta$ decay both historically and scientifically. In both cases, an understanding of the nuclear matrix elements governing the second-order decay is a critical parameter for any theory that estimates the nuclear matrix elements of second-order decay processes.

Until recently, double-$\gamma$ decay has only been observed in exceptional cases where both the ground state and the initial state have a spin-parity $J^\pi = 0^+$ character for the doubly magic nuclei $^{16}$O, $^{40}$Ca\textsuperscript{5}, and $^{90}$Zr\textsuperscript{8}. Here single $\gamma$-emission is blocked, and only conversion-electron decay and double-$\gamma$ decay are allowed. In these experiments, the correlations between energies and angles of these $\gamma$-rays were used to determine the decay probabilities of electric and magnetic dipoles. Large state-of-the-art high-purity germanium (HPGe) detector systems\textsuperscript{9,10} have been used to search for the competitive $\gamma\gamma/\gamma$ decay where also the single $\gamma$ decay. Event though unsuccessful in that respect, these experiments successfully measured an E5 transition with the branching of $1.12(9) \times 10^{-7}$. It is only with recent instrumentation developments of detector materials that can provide both the energy and time resolution required\textsuperscript{11} that a discovery of this decay mode recently was announced\textsuperscript{12}. The discovery experiment consisted of five LaBr\textsubscript{3}:Ce detectors arranged in a planar configuration with relative angles of $72^\circ$ between the detectors, providing angular distribution data points at $72^\circ$ and $144^\circ$. That collaboration could announce the discovery of the $\gamma\gamma/\gamma$ decay with a statistical significance of $5.5 \sigma$ (standard deviations), near but above the typical discovery limit, $5 \sigma$. From the two angular data points as well as the energy spectrum of the individual $\gamma$ rays at $72^\circ$ angle, the off-diagonal polarizabilities $\alpha_{M2E2} = 33.9(2.8) \ e^2 fm^4/MeV$ and the $\alpha_{E3M1} = 10.1(4.2) \ e^2 fm^4/MeV$ polarizabilities were extracted. While the observation of the peak associated with $\gamma\gamma/\gamma$ decay was statistically clear, the nature of this decay was more uncertain, having the two dominating multipolarity combinations separated only by a small statistical difference, favouring the $\alpha_{M2E2}$ component\textsuperscript{13}.

Given the nature of this experiment to observe a longstanding prediction of a the fundamental concepts in quantum mechanics and quantum electro-dynamics, it is highly desirable to independently confirm this discovery. Some possibilities that have been under discussion to perform this independent confirmation is to either return to the HPGe approach with new and complex detector system and event processing like with the Advanced GAmma Tracking Array (AGATA) setup\textsuperscript{14,15} or highly charged radioactive ions\textsuperscript{16}. Here we report on an experiment using the ELI Gamma Above Neutron Threshold (ELIGANT) detector system\textsuperscript{17,18} in a new configuration\textsuperscript{19} similar to reference\textsuperscript{12}. Considering the very weak signal associated with such events, and in view of the large experimental difficulties associated with the measurements, we found necessary to re-optimize and upgrade the experimental setup, enabling the extraction of observables undetected in the discovery experiment.
1 Results

Experimental setup. The experiment was performed using eleven $3'' \times 3''$ CeBr$_3$ detectors from ELIGANT, shown in Figure 1a. While ELIGANT consists of both LaBr$_3$:Ce and CeBr$_3$ detectors, the CeBr$_3$ detectors were chosen to remove any possible source of background contribution from the natural radioactivity in lanthanum. The detector configuration was a circle with an inner radius to the front face of the scintillators of 40 cm. This distance was enough to separate true coincidences from multiple Compton scattering of single $\gamma$ rays using the photon time-of-flight (TOF), see Figure 1b. The relative angles between the eleven detectors were 32.7°, with an opening angle, given by the lead shielding, of ±3.4°. This gave five independent $\gamma\gamma$-correlation angles centered at: 32.7°, 65.5°, 98.2°, 130.9°, and 163.6°. The detectors were separated with a minimum of approximately 15 cm of effective lead shielding between two neighbouring detectors to remove any contribution from single Compton scattering between detector pairs at low angles. The setup was characterized both with an in-house GeANT toolkit[20] and a $^{152}$Eu source with an activity of 60 kBq. For a comprehensive overview, see reference [19]. The $\gamma\gamma/\gamma$-decay data on $^{137}$Ba were collected using a $^{137}$Cs source with an activity of 336 kBq for 49.5 days active data taking.

Energy spectra. From the data set obtained with the $^{137}$Cs source a $(\gamma_1, \gamma_2)$ coincidence matrix was constructed where the $\gamma$ rays were considered coincident if the time difference between them was less than one standard deviation from the prompt time distribution, $\Delta t_{1,2} \leq 655$ ps. This condition was obtained from the coincident 444 kilo electron-volt (keV) and 245 keV $\gamma$ rays from the $2^+ \rightarrow 4^+ \rightarrow 2^+$ decay chain in $^{152}$Sm following the electron capture decay of $^{152}$Eu. Corrections for detector efficiencies were done on an event-by-event basis[19]. A time difference of $20 \leq \Delta t_{1,2} \leq 820$ ns was used to estimate the uncorrelated background events with two detected $\gamma$ rays and subtracted after applying an appropriate scaling factor. To remove the background contribution from electron-positron pairs produced by cosmic rays a multiplicity-two condition was assigned together with an additional energy condition that $|E_1 - E_2| < 960 - (E_1 + E_2)$ keV. The full data set, as well as the different angular groups, were used to construct the summed energy spectra. The peaks were fitted assuming a quadratic background both with a Gaussian distribution as well as GeANT4 simulated data. Both fitting methods gave consistent results. The full summed spectrum of $E_1 + E_2$ with these conditions imposed is shown in Figure 2.

Branching. As an experimental observable to evaluate the relative decay probability we use the definition of the integrated differential branching ratio

$$\delta(E_1, E_2, \theta_{1,2}) = \frac{(4\pi)^2}{\Gamma_\gamma} \int^{E_2}_{E_1} d\omega \frac{d\Gamma_\gamma}{d\omega d\Omega d\Omega'} \bigg|_{\theta_{1,2}}.$$  \hspace{1cm} (4)$$

In this definition, $\Gamma_\gamma$ is the total single-$\gamma$ decay width, proportional to the size of the single-$\gamma$ peak. Given an angle, $\theta_{1,2}$, the differential decay is integrated over the frequency of the $\gamma$ ray, $\omega$. The frequency is proportional to the energy, and the integration limits are taken as the edges of the energy bin of interest. In the experimental spectrum a natural low-energy limit comes from the low-energy threshold of the detectors around 120 keV. However, to reduce the contamination from the 511 keV $\gamma$-rays originating from electron-positron annihilation, the integration limits $E_1 = 180$ keV and $E_2 = 331$ keV were chosen. The upper limit was chosen as the half of the total energy as we are not able to distinguish any relative ordering of the $\gamma$ rays. This procedure was performed for all combinations of $\theta_{1,2}$ and $\delta$ was evaluated as a function of angle. The results from this evaluation is shown in Figure 5.

This data can be directly fitted to the generalized polarizability functions of equation [5] discussed in the methods section, using only $\alpha_{M2E2}$ and $\alpha_{E3M1}$ as free parameters. Other components like $\alpha_{E2M2}$ or $\alpha_{M3E1}$ could in principle also contribute. However, the general polarizability functions are linearly dependent in the exchange of terms, weighted by the coefficients given by the Wigner $6j$ symbols, and this experiment is not sensitive to this ordering. These additional components are, furthermore, expected to be small. Thus, we restrict the discussion to the $\alpha_{M2E2}$ and $\alpha_{E3M1}$ polarizabilities from here on.
Figure 1: Experimental setup. (Top) Coincident $\gamma$ rays could originate either from true double-$\gamma$ decay events illustrated with red cones, or from multiple Compton scattering between detectors illustrated with blue cones. (Bottom) Multiple Compton scattering events were rejected by the time-of-flight of the $\gamma$ ray, shown in the blue histogram. The time condition for prompt $\gamma$-rays are shown as red dashed lines and verified with a $^{152}$Eu source.

Energy sharing distributions. The angular distributions themselves are not enough to completely distinguish between the contribution from the different polarizabilities. When calculating the goodness-of-fit ($\chi^2$), two local minima corresponding to either a large $\alpha_{M2E2}$ component or a large $\alpha_{E3M1}$ component appear. Instead, it is necessary to study the energy-sharing distributions between the two individual $\gamma$-rays. From equations (5) and (6) in the methods section it is clear that the energy dependence of the decay for the two different cases follows $\frac{d\Gamma_{\gamma\gamma}}{d\omega} \propto \omega^5\omega'^7$ for M2E2 and as $\frac{d\Gamma_{\gamma\gamma}}{d\omega} \propto \omega^3\omega'^5$ for E3M1 with $\omega' = 662 - \omega$. It is clear from these relations that the energy sharing distributions are expected to have a maximum at $E_\gamma = E'_\gamma = 331$ keV for the M2E2 type transitions, while an asymmetric maximum is expected at $E_\gamma = 200$ keV and $E'_\gamma = 442$ keV for the E3M1 type transitions.

For this purpose, $\delta$ from equation (4) was evaluated in separate slices of 30 keV energy difference between the low- and high-energy limit of $E_\gamma$. Figure 4 shows the results of these evaluations. A $\chi^2$ value was then calculated...
Figure 2: **Summed double-\(\gamma\) energy spectrum.** Black data points show the summed energy of two coincident photons detected in the CeBr\(_3\) detectors for events with a multiplicity of two. Gray data points show the sum energy spectrum when the multiplicity is larger than two, which mainly correspond to the background induced by cosmic ray showers. We also show the fit to the data of a quadratic background as a dashed red line and the fit of the background plus a Gaussian peak as a solid red line.
Figure 3: Angular distribution of the two photons of the double-γ decay. (Top) Illustration of the single-γ and the two types of double-γ decay discussed here. Here, M4 corresponds to the single-photon decay. The blue and pink decays show the lowest octupole-dipole and quadrupole-quadrupole components, respectively. (Bottom) The angular correlation of the two photons emitted in the double-γ decay from this work and reference [12] compared to the expected angular distributions of pure M2E2 and E3M1 decay.
Based on $\alpha_{\text{M2E2}}$ and $\alpha_{\text{E3M1}}$ simultaneously using the energy-integrated angular data points and the angle-summed energy data points. The resulting $\chi^2$ surface is shown in figure 4b. As seen here, the $\chi^2$ analysis from this data favours a large $\alpha_{\text{E3M1}}$ component, in contradiction with both the experimental interpretation and theoretical conclusions reported in reference [12].

### Theoretical calculations

The first step to understanding these results is to evaluate theoretical calculations of the polarization functions from equation (2), using the quasiparticle-phonon model (QPM) approach. The application of the QPM in the case of odd-mass spherical nuclei is discussed in detail in reference [22]. In particular, the nuclear structure of $^{137}$Ba was studied within the framework of this model in references [24, 25] and recently in reference [12]. In the work presented here, the calculations were built on the Energy-Density-Functional (EDF) theory coupled with the QPM to obtain magnetic and electric dipole spectral distributions for the $N = 81$ isotope $^{137}$Ba with one neutron hole in the closed $N = 82$ shell. The model parameters of the EDF+QPM approach are firmly determined from nuclear structure data or derived fully microscopically [27–29]. The theoretical results are shown in table 1 and agree with the data in terms of absolute branching strength, $\Gamma_{\gamma\gamma}/\Gamma_\gamma$. In addition, the EDF+QPM used here predicts a significantly larger $\alpha_{\text{E3M1}}$ than the value reported in reference [12] from the QPM, close to the experimental observations presented here. However, the relative magnitude of the $\alpha_{\text{M2E2}}$ and $\alpha_{\text{E3M1}}$ coefficients obtained from the EDF+QPM theory, as well as from reference [12] are different than the experimental results obtained in this work. In particular, the present measurement indicates that the $\alpha_{\text{M2E2}}$ coefficient is significantly smaller than previously reported, and at this level of complexity EDF+QPM it is not able to account for the apparent discrepancy with the experimental data.

To understand the origin of this discrepancy, the properties of the dominant, low-lying, states were investigated from another perspective using the state-of-the-art nuclear Monte Carlo shell model (MCSM) [30, 31]. These calculations were used to extract information from the three lowest-energy $7/2^+$ states, the five lowest-energy $5/2^+$ states, as well as the ground $3/2^+$ and isomeric $11/2^-$ states. Here, the wave function of the isomeric state with a spin-parity configuration of $J^\pi = 11/2^-$ is dominated by two $\pi g_{7/2}$ proton holes coupling to a $J^\pi = 0^+$ state and a single neutron hole in $\nu h_{11/2}$. The $J^\pi = 7/2^+$ state is different, however, as 90% of the neutron hole occupation is in $\nu d_{3/2}$, coupled
to a $2^+$ state of six valence protons in the $\pi g_{7/2}$ orbital. The $\nu g_{7/2}$ orbital itself is almost full with a hole occupation of only 4%. This is in contrast with the EDF+QPM results where the $2^+ \otimes d_{3/2}$ contribution is 38.7% and the $\nu g_{7/2}$ single-particle contribution is 51.3%. Thus, the odd-neutron contribution to the M2 transition rate in the MCSM would require a highly hindered transition between $\nu h_{11/2}$ and $\nu d_{5/2}$, or by utilising a minor $\nu g_{7/2}$ vacancy. This, gives rise to a rather small M2 transition within the MCSM, with a reduced transition probability, $B(M2) = 13.5 \times 10^{-3} \mu^2$fm$^2$, three orders of magnitude less than predicted by the EDF+QPM model where $B(M2) = 14.9 \mu^2$fm$^2$. This can explain the observed suppression of $\alpha_{E2M2}$. It is interesting to note that with increasing excitation energy, the MCSM predict a smooth change in orbital occupation from $\nu d_{5/2}$ to $\nu d_{5/2}$, constructively adding to the M2 transition strength for all the calculated $7/2^+$ transitions in contrast to the EDF+QPM where all higher-lying states act destructively. Table 1 lists the contributing low-lying matrix elements discussed here.

Table 1: Experimental and calculated $\alpha$ coefficients and $\gamma\gamma/\gamma$ decay branching ratios.

| Matrix element | EDF+QPM | MCSM | EDF+QPM | MCSM |
|----------------|---------|------|---------|------|
| $\Gamma_{\gamma\gamma}/\Gamma_\gamma$ | $2.6(30)$ | $\pm 8.8(50)$ | $\pm 36.4(20)$ | 
| $\Gamma_{E2}$ | $3.73(\ast)$ | $59.4$ | $20.7$ | 
| $\Gamma_{E3}$ | $0.196(\ast\ast)$ | $-3.34$ | $-34.3$ | 
| Literature | $2.05(37)$ | $33.9(28)$ | $10.1(42)$ | 
| QPM | $2.69$ | $42.6$ | $9.5$ | 

(*) The $\Gamma_{\gamma\gamma}/\Gamma_\gamma$ decay branching ratio is obtained from the EDF+QPM with gyromagnetic spin factor $g_\nu^{\text{eff}} = 0.6g_\nu^{\text{bare}}$, based on the individual reduced transition probabilities. The best fit for the decay ratio is obtained when choosing $g_\nu^{\text{eff}} = 0.7g_\nu^{\text{bare}}$ as $\Gamma_{\gamma\gamma}/\Gamma_\gamma = 2.8$. (**) For the MCSM the low value of the integrated decay ratio originates from the large predicted $\Gamma_\gamma$ width. When using the experimental $\Gamma_\gamma^{\exp}$, the branching becomes $\Gamma_{\gamma\gamma}/\Gamma_\gamma^{\exp} = 2.20$.

Table 2: Calculated matrix elements.

| Matrix element | EDF+QPM | MCSM | Matrix element | EDF+QPM | MCSM |
|----------------|---------|------|----------------|---------|------|
| $\langle 3/2^+M1||5/2^+ \rangle$ | $-0.11$ | $-0.139$ | $\langle 5/2^+||E3||11/2_1^+ \rangle$ | $-168$ | $57.2$ |
| $\langle 3/2^+M1||5/2^+ \rangle$ | $0.172$ | $5/2^+||E3||11/2_1^+ \rangle$ | $-81.9$ | 
| $\langle 3/2^+M1||5/2^+ \rangle$ | $0.183$ | $5/2^+||E3||11/2_1^+ \rangle$ | $-128$ | 
| $\langle 3/2^+M1||5/2^+ \rangle$ | $63.9$ | $39.0$ | $\langle 7/2_2^+||M2||11/2_1^+ \rangle$ | $1.14$ | $-0.0518$ |
| $\langle 3/2^+M1||5/2^+ \rangle$ | $-46.4$ | $7/2_2^+||M2||11/2_1^+ \rangle$ | $0.76$ | 

Transition matrix elements of the dominating transitions, in each model, calculated using the EDF+QPM and MCSM models for the lowest-energy states that contribute to the double-$\gamma$ decay in $^{137}$Ba. The EDF+QPM values for the magnetic transitions correspond to $g_\nu^{\text{eff}} = 0.6g_\nu^{\text{bare}}$, while the MCSM values correspond to $g_\nu^{\text{eff}} = g_\nu^{\text{bare}}$. This can explain the observed suppression of $\alpha_{E2M2}$. It is interesting to note that with increasing excitation energy, the MCSM predict a smooth change in orbital occupation from $\nu d_{5/2}$ to $\nu d_{5/2}$, constructively adding to the M2 transition strength for all the calculated $7/2^+$ transitions in contrast to the EDF+QPM where all higher-lying states act destructively. Table 2 lists the contributing low-lying matrix elements discussed here.

Regarding the $\alpha_{E3M1}$ component of the decay, the main components obtained from the EDF+QPM calculations are from the coupling of the single-particle mode with the surface vibrations of the even-even core. For the $5/2^+_1$ case the QPM state vector is dominated by $53.8\%[2d_{5/2}]$ neutron component with two main contributing ‘quasiparticle@phonon’ configurations as 15.1%$[3s_{1/2} \otimes 2^+_1]$ and 13.8%$[1h_{11/2} \otimes 3^+_1]$. As a consequence, due to the exchange of the collective $3^+_1$ octupole phonon, we obtain a rather strong E3 transition, consistent with our experimental observations. The structures of the $J^\pi = 5/2^+$ states within the MCSM are fragmented between a $d_{5/2}$ hole and a $d_{5/2}$ hole. As the E3 transition is not hindered between $h_{11/2}$ and $d_{5/2}$, the size of the matrix elements follow the population trend of the $d_{5/2}$ hole with increasing excitation energy. Thus, for these states the EDF+QPM and the MCSM give a consistent picture with a constructive addition to the strength for each successive state among the first three excited states with the main difference that in the EDF+QPM, the main contribution comes from the $5/2_1^+$ state.
while the MCSM predicts that the $5/2^+_{2,3}$ states are dominating.

2 Conclusions

In conclusion, we have unambiguously confirmed the recent discovery of the existence of the competitive double-photon decay process in atomic nuclei. The experimental setup was optimised for obtaining a clean signal over a wide angular range based on the expected intensities from the discovery of the decay mode. We find a significant M1E3 matrix element product contribution to the double-γ decay mode of $^{137}\text{Ba}$, contradicting the conclusions of the discovery experiment. From our calculations using the EDF+QPM and the MCSM nuclear models, we find that both models reproduce the octupole-dipole component consistently, but the nature and the strength of the quadrupole-quadrupole component, differ significantly. These results highlight the power of the double-γ decay as an experimental tool to extract detailed structural properties from atomic nuclei, and also the need for further experimental and theoretical investigations of this type of decay.

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Methods

Polarization functions. To obtain the nuclear polarizabilities, $\alpha_{p,S,L'/S,L}$, from the differential decay probability we follow the theoretical treatment in references [34, 35]. Here the differential decay probability can be expressed in terms of generalized polarization functions, $P_j^G(S, L, S', L')$, and Legendre polynomials, $P_l(\cos \theta)$, as

$$
\frac{d^3I_{\gamma \gamma}}{d\omega d\Omega'} = \frac{\omega'}{96\pi} \sum P_j^G(S'1L'1S1L1)P_j^G(S'2L'2S2L2) \sum a_l^G P_l(\cos \theta),
$$

where the generalized polarization functions are defined as

$$
P_j^G(S' L' SL) = (-1)^{S + S' 2\pi(-1)^{I_1 + I_f} \omega L_1 \omega' L'},
$$

$$
\left\{ \begin{array}{ll}
L & L' \\
I_f & I_i
\end{array} \right\} \alpha_{\Lambda S' L' / S L} + (-1)^{S + S'} \left\{ \begin{array}{ll}
L' & L \\
I_f & I_i
\end{array} \right\} \alpha_{S' \Lambda S L' / L' L}.
$$

The sums in equation (9) run over all the permutations of electric, $S = 0$, and magnetic, $S = 1$, combinations with multipolarity, $L$, allowed in the decay, and over all Legendre polynomials with non-zero coefficients. The general polarizability functions in equation (6) consist of a linear combination of the off-diagonal polarizabilities of the nucleus weighted by coefficients determined by the corresponding angular momentum algebra of the decay.

The quasiparticle-phonon model. The QPM Hamiltonian includes mean field, pairing interaction and separable multipole and spin-multipole interactions [22]. The mean field for protons and neutrons is defined as a Woods-Saxon potential with parameter sets derived self-consistently from a fully microscopic Hartree-Fock-Bogoliubov (HFB) calculations described in [26, 33]. The method assures a good description of nuclear ground-state properties by enforcing that measured separation energies and nuclear radii are reproduced as close as possible from a fully microscopic Hartree-Fock-Bogoliubov (HFB) calculations described in [26, 33]. Of particular importance in these studies is the determination of the isovector spin-dipole coupling constant which is extracted from comparison to data from [23] and fully self-consistent quasiparticle random phase approximation (QRPA) calculations using the microscopic EDF of [22]. Single-particle (s.p.) energies of the lowest-lying excited states in $^{137}$Ba are fine-tuned to experimental values to achieve the highest accuracy in the description of the experimental data. We point out that the s.p. energies problem is not a matter of the interaction parameters but originate in the quasiparticle spectrum, which indicates the necessity to go beyond the static mean-field formalism [23, 25].

The notation $\alpha_{j^\pm}$ is the quasiparticle creation operator with shell quantum numbers $j \equiv [(n, l, j)]$ and projection m; $Q_{\Lambda \mu i}^\pm$ denotes the phonon creation operator with the angular momentum $\lambda$, projection $\mu$ and QRPA root number $i$; $\Psi_0$ is the ground state of the neighboring even-odd nucleus and $\nu$ stands for the number within a sequence of states of given angular momentum $J^\pi$ and projection $M$. The coefficients $C_j^\nu$ and $D_j^\nu(\nu)$ are the quasiparticle and ‘quasiparticle ⊗ phonon’ amplitudes for the $\nu$ state. The coefficients of the wave function (7) and the energy of the excited states are found by diagonalisation of the model Hamiltonian within the approximation of the commutator linearization [22, 23]. The components $[\alpha_{j^\pm} Q_{\Lambda \mu i}^\pm]_M$ of the wave function (7) may violate the Pauli principle. The exact commutation relations between quasiparticle and phonon operators are used to solve this problem. The properties of the phonons are determined by solving QRPA equations from Ref. [22, 24]. The model basis includes one-phonon states with spin and parity $J^\pi = 1/2^\pm, 3/2^\pm, 5/2^\pm$ and excitation energies up to $E_\alpha = 20$ MeV. The calculations of the $\alpha$-coefficients of the double-γ decay probability of $^{137}$Ba include all low-energy excited states with spin and parity $J^\pi = 1/2^\pm, 3/2^\pm, 5/2^\pm, 7/2^\pm, 9/2^\pm$ and excitation energies up to $E_\alpha = 10$ MeV.

In the case of the E1 transitions, we have used effective charges $e_{p,n}^{\text{eff}} = (N/A)e$ (for protons) and $e_{n}^{\text{eff}} = -(Z/A)e$ (for neutrons) to separate the center of mass motion and ‘bare’ values for E2 and E3 transitions $e_p = e$ (for protons) and $e_n = 0$ (for neutrons), where $e$ is the electron charge. Following previous QPM calculations [23], the magnetic transitions are calculated with a quenched effective spin-magnetic moment $g_{\text{eff}}$. The influence of the $g_{\text{eff}}$ parameter on the experimental observables related to electromagnetic transitions of lowest-lying states and double-γ decay probability coefficients was investigated by carrying out EDF+QPM calculations for several choices of this parameter between 0.6 and 1 of the value of the ‘bare’ spin-magnetic moment, $g_{\text{bare}}$. The theoretical observations indicate that the values $g_{\text{eff}} = 0.6 - 0.7 g_{\text{bare}}$ are in agreement with our previous findings [22, 23, 25] reproduce quite well the experimental data on M1 and M2 transition strengths and the angular distribution of the two photons of the double-γ decay.
Monte Carlo shell model. In the MCSM, the approximated wave functions, $|\Psi_{N_b}\rangle$, are obtained as a superposition of spin ($I$) and parity ($\pi$) projected Slater determinant basis states, $|\phi_n\rangle$,

$$|\Psi_{N_b}\rangle = \sum_{n=1}^{N_b} \sum_{K=-I}^{I} f_{n,K}^{N_b} P_{M,K}^{I\pi} |\phi_n\rangle,$$

(8)

where $N_b$ is the number of basis states, $P_{M,K}^{I\pi}$ is the spin-parity projection operator, and the $f_{n,K}^{N_b}$ coefficients are obtained from diagonalizing the Hamiltonian. The set of basis states are selected by Monte Carlo methods and iteratively refined to minimize the ground state energy. The model space for these calculations included the $1g_{9/2}$, $1g_{7/2}$, $2d_{5/2}$, $2d_{3/2}$, and $3s_{1/2}$ even-parity orbitals, as well as the $1h_{11/2}$, $2f_{7/2}$, and $3p_{3/2}$ odd-parity orbitals. The two-body matrix elements were obtained from the JUN45 and SNBG3 data sets and the $V_{MU}$ interaction. To obtain the transition matrix elements effective proton and neutron charges $e_p = 1.25$ and $e_n = 0.75$, and gyromagnetic factors $g_{\ell,p} = 1$, $g_{\ell,n} = 0$, $g_{s,p} = 5.586$, and $g_{s,n} = -3.826$ was used. The calculations followed the procedure for the tin isotope chain closely. Said reference and references within contains a detailed description of the procedure.

Author contributions P.-A.S., L.C., E.A., D.L.B., C.M., and A.P. designed the experimental setup. P.-A.S., L.C., E.A., G.L.G., D.L., D.N., and T.P. collected the data. L.C., and D.N. wrote the software for data conversion. P.-A.S. wrote the software for data analysis and analysed the data. N.T., T.O., Y.T., and H.L. performed the theoretical calculations. P.-A.S. and D.L. performed the GEANT4 simulations. P.-A.S., L.C., N.T., T.O., D.L.B., and N.P. discussed the interpretation of the experimental and theoretical results. P.-A.S. and N.T. prepared the manuscript draft, and all authors read and contributed to the discussion of the final manuscript.

Data availability Raw data were obtained at the Extreme Light Infrastructure – Nuclear Physics facility, Romania. All the data used to support the findings of this study are available from the authors upon reasonable request.

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Competing Interests The authors declare that they have no competing financial interests.

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