New experimental access to the two-phonon octupole vibration in ^{208}Pb [*]

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Abstract

The feasibility of a new experimental method to identify two-phonon octupole vibrations is investigated for the example of ^{208}Pb. The 2^+ member of the $3_1^- \otimes 3_1^-$ multiplet can be excited with the (γ, γ') reaction and a sizable E1 decay to the 3_1^- level serves as a signature of the two-phonon character. Numerical estimates within the quasiparticle–phonon model indicate that such an experiment should be possible with state-of-the-art large volume Ge detectors like the EUROBALL cluster module.

1. Introduction

The search for the two-phonon octupole vibration in the doubly magic nucleus ^{208}Pb is one of the most outstanding nuclear structure problems with a long history. In a spherical nucleus like ^{208}Pb collective surface vibrations form excitation modes lowest in energy which are well described by a phonon approach. The relevance of their detailed understanding lies in two aspects. On one hand, coupling to low-lying surface

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oscillations is the decisive mechanism for the damping of nuclear excitations [1]. On the other hand, one expects characteristic multiplets of excited states in the low-energy spectra due to the coupling of two (or more) of these phonons. Investigating their properties provides a measure how far the phonon concept implying purely harmonic vibrations holds in real nuclei. The energy splitting of the multiplets furthermore permits valuable insight into the effects of the Pauli principle and the general properties of the phonon–phonon interaction in the coupling of elementary excitation modes.

States corresponding to $|2_1^+ \otimes 2_1^+; 0^+, 2^+, 4^+\rangle$ configurations are well established in many spherical nuclei. There is also evidence for the $1^- - 5^-$ quintuplet of $2_1^+ \otimes 3_1^-$ states near closed shells. In particular, the 1^- members are selectively excited in (γ, γ') experiments (see Refs. [2–4] and references therein) and rather complete experimental information on the multiplet exists [5] e.g. for the case of ^{144}Nd. On the other hand, experimental evidence for $|3_1^- \otimes 3_1^-; 0^+, 2^+, 4^+, 6^+\rangle$ states is rather sparse [6–10]. Two nuclei, ^{146}Gd and ^{208}Pb, are the most interesting candidates, since their first excited state is due to an octupole vibration and the members of this multiplet should be the lowest states of two-phonon nature in the respective nuclei.

The properties of the $3_1^- \otimes 3_1^-$ multiplet in ^{208}Pb have been studied in a variety of theoretical approaches [11–18]. Experimental searches have been performed using the (n,γ) reaction [19], internal conversion decay of 0^+ levels [20], inelastic hadron scattering [21], heavy ion (HI) Coulomb excitation [22,23] and γ-spectroscopy after deep inelastic reactions [24], but the contradictions of the results are confusing. The interpretation of the experiments described in Refs. [19,20] is rather model dependent. However, it was demonstrated recently by the observation of a characteristic E3–E3 decay sequence following excitation in inelastic neutron scattering that the $E_x = 5.241$ MeV, $J^\pi = 0^+$ level must contain a large $3_1^- \otimes 3_1^-$ component which makes it the best characterized two-phonon candidate in ^{208}Pb so far [25]. Evidence for the population of the $|3_1^- \otimes 3_1^-; 6^+\rangle$ state by two-step Coulomb excitation in the reaction ^{208}Pb $+$ ^{208}Pb has been claimed in Ref. [22]. However, because of the low cross sections in this experiment the projectile energy had to be chosen slightly above the Coulomb barrier where a substantial fraction of the total cross section is due to single nucleon transfer. It was therefore questioned in Refs. [21,24] that the γ-transition taken as signature of the two-phonon state in Ref. [22] belongs to ^{208}Pb, and a recent HI Coulomb excitation experiment at Argonne shows that the state seen must rather be assigned to ^{207}Pb [23].

Clearly, this situation is rather unsatisfactory asking for experimental clarification. The purpose of this note is twofold: to provide a microscopic study of the $3_1^- \otimes 3_1^-$ multiplet in ^{208}Pb and its decay properties to the g.s. and the one-phonon 3_1^- level, and based on these results to investigate the feasibility of a new experimental method in order eventually to identify the long-sought two-phonon octupole vibration. We propose to look for the excitation of the $|3_1^- \otimes 3_1^-; 2^+\rangle$ state in a nuclear resonance fluorescence (NRF) experiment. Although the NRF method is by far most sensitive to dipole excitations the recently demonstrated dramatic improvements of the detection limits [26] with a new generation of large volume HPGe detectors like the EUROBALL Cluster module effectively enable the observation even of weak electric quadrupole transitions. A clear
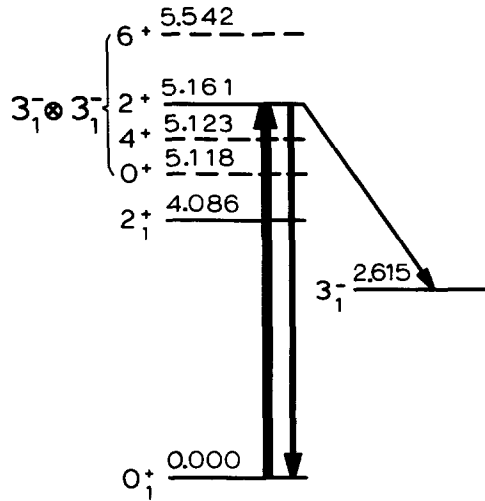
Fig. 1. Two-phonon octupole vibration multiplet in ^{208}Pb and levels relevant for the calculations described in the text. The experimental one-phonon energies are taken from the literature [27]. The multiplet energies are from a QPM calculation including of mixing between one- and two-phonon components in the excited state wave functions.

signature for the two-phonon nature of the 2^+ state would be a E1 decay to the 3_1^- one-phonon state, since none of the other 2^+ levels in the excitation energy region around the two-phonon octupole multiplet is expected to show a sizable partial decay width to the 3_1^- state.

The relevant levels and transitions are schematically summarized in Figure 1. We are interested in the g.s. excitation probability to the $|3_1^- \otimes 3_1^-; 2^+\rangle$ level (indicated by the upward arrow) and the partial decay widths to the g.s. and the 3_1^- state (denoted by downward arrows). The experimental energies of the 2_1^+ and 3_1^- vibrations in Fig. 1 are taken from the literature [27]. Next, we discuss the model calculations which also provide reliable estimates for the energies of the two-octupole phonon states indicated in Fig. 1.

2. Quasiparticle–phonon model calculations

The properties of the $3_1^- \otimes 3_1^-$ multiplet are calculated within the quasiparticle-phonon model (QPM) which successfully describes many different properties of single and multi-phonon low-lying states in spherical nuclei (see e.g. Refs. [28–32]). The model Hamiltonian includes terms corresponding to the mean field for neutrons and protons, monopole pairing and the residual interaction in a separable form. Excited states of even nuclei are described by wave functions which may include one-, two- and higher-phonon components. The properties of phonons, their excitation energies and their internal fermion structure are obtained by diagonalizing the model Hamiltonian in the random phase approximation (RPA); see Ref. [33] for details.

We introduce the phonon operator for the first 3^- state as

$$Q_{3\mu}^+ = \frac{1}{2} \sum_{j_1 j_2}^{N,Z} \left\{ \psi_{j_1 j_2} [\alpha_{j_1 m_1}^+ \alpha_{j_2 m_2}^+]_{3\mu} + (-1)^\mu \phi_{j_1 j_2} [\alpha_{j_1 m_1} \alpha_{j_2 m_2}]_{3-\mu} \right\}, \tag{1}$$

where j denotes a single particle level with quantum numbers n, l, j and m is its projection; α_{jm}^+ is a quasiparticle creation operator. The quantities $\psi_{j_1 j_2}$, $\phi_{j_1 j_2}$ denote the forward and backward amplitudes, respectively, of the RPA equations. The energy of this state corresponds to the first solution for multipolarity $J^\pi = 3^-$. In the calculation we used the single particle spectrum of Ref. [34]. It was adjusted to properties of low-lying states in odd nuclei of the lead region and later successfully used for different calculations in ^{208}Pb. The parameters of the residual interaction of the Hamiltonian were adjusted to reproduce the collectivity of the lowest 3^- state, i.e. the experimental B(E3, g.s. $\rightarrow 3_1^-$) value [35] for this level.

In a first step we assume that mixing between one- and two-phonon configurations in the low-energy spectra is negligible. This allows us to define the wave function of the two-phonon $|3_1^- \otimes 3_1^-; 2^+\rangle$ state as

$$|\Psi_{|3_1^- \otimes 3_1^-;2^+\rangle\mu}\rangle = \frac{1}{\sqrt{2}} \sum_{M_1 M_2} \langle 3M_1 3M_2|2\mu\rangle Q_{3-M_1}^+ Q_{3-M_2}^+ |\Psi_{\text{g.s.}}\rangle. \tag{2}$$

An E1 transition between the states described in Eqs. (1) and (2) as well as a direct decay of the 2^+ state to the ground state are forbidden in the pure boson picture. Only through exact commutation relations between phonon and quasiparticle operators, i.e. taking into account the fermion structure of phonons, one gets non-vanishing values. The reduced transition probability for the E1 decay ($|3_1^- \otimes 3_1^-; 2^+\rangle \rightarrow 3_1^-$) has the form

$$B(\text{E1}) = 171.5 \left| \sum_{j_1 j_2 j' j''} \langle j_2 \| M(\text{E1}) \| j_1 \rangle u_{j_1 j_2}^{(+)} \sum_J (-1)^J \begin{Bmatrix} j_2 & j_1 & 1 \\ j'' & j' & 3 \\ 3 & 3 & J \end{Bmatrix} \right.$$

$$\times e_{p(n)} \left[(\psi_{j_1 j'} \psi_{j_2 j''} \psi_{j'' j'} + \phi_{j_1 j'} \phi_{j_2 j''} \phi_{j'' j'}) \delta_{J,2} \right.$$

$$\left. + (\psi_{j_1 j'} \phi_{j_2 j''} \phi_{j'' j'} + \phi_{j_1 j'} \psi_{j_2 j''} \psi_{j'' j'}) (2J+1) \begin{Bmatrix} 1 & 3 & 2 \\ 3 & 3 & J \end{Bmatrix} \right] \Bigg|^2 \tag{3}$$

and for the E2 decay ($|3_1^- \otimes 3_1^-; 2^+\rangle \rightarrow$ g.s.)

$$B(\text{E2}) = 19.6 \left| \sum_{j_1 j_2 j'} e_{p(n)} \langle j_2 \| M(\text{E2}) \| j_1 \rangle v_{j_1 j_2}^{(-)} \psi_{j_1 j'} \phi_{j' j_2} \begin{Bmatrix} 2 & 3 & 3 \\ j' & j_2 & j_1 \end{Bmatrix} \right|^2, \tag{4}$$

where $\langle j_2 \| M(\text{E}\lambda) \| j_1 \rangle$ is the reduced matrix element of the electromagnetic transition; $v_{j_1 j_2}^{(-)} = u_{j_1} u_{j_2} - v_{j_1} v_{j_2}$ and $u_{j_1 j_2}^{(+)} = u_{j_1} v_{j_2} + v_{j_1} u_{j_2}$ are combinations of coefficients of the Bogoliubov transformation from particle (a_{jm}^+) to quasiparticle (α_{jm}^+) operators: $\alpha_{jm}^+ =$

Table 1
Excitation and decay properties of 2^+ member of two-phonon $3_1^- \otimes 3_1^-$ multiplet in ^{208}Pb. The results given in column I neglect the mixing of one- and two-phonon configurations in the wave functions of excited states, while it is included in the full calculation presented in column II. In brackets we present the results of test calculations varying the strength of the residual interaction. See text for details

	I (Ia, Ib)	II (IIa, IIb)
B(E2, g.s. $\rightarrow [3_1^- \otimes 3_1^-]_{2^+}$), e^2 fm^4	10.4 (5.1, 17.7)	40.2 (19.0, 54.6)
Γ_0(E2, $[3_1^- \otimes 3_1^-]_{2^+} \rightarrow$ g.s.), meV	6.58 (3.21, 11.2)	25.4 (12.0, 34.5)
$\Gamma_{3_1^-}$ (E1, $[3_1^- \otimes 3_1^-]_{2^+} \rightarrow 3_1^-$), meV	14.0 (12.6, 17.0)	13.7 (12.9, 14.3)
$\Gamma_{3_1^-}/\Gamma_0$	2.13 (3.93, 1.52)	0.54 (1.08, 0.41)

$u_j a_{jm}^+ + (-1)^{j-m} v_j a_{j-m}$; for ^{208}Pb these coefficients are either 0 or 1. The numerical factors appearing in Eqs. (3) and (4) are products of $(2\lambda + 1)$ factors for the phonon operators taken into account. We use effective charges $e_{p(n)} = 1(0)$ and $N/A(-Z/A)$ for E2 and E1 transitions, respectively.

3. Results and discussion

The partial decay widths of these transitions can be calculated from

$$\Gamma(\text{E1}) = 1.048(E_i - E_f)^3 B(\text{E1}, J_i \rightarrow J_f) \text{ eV}, \tag{5}$$

$$\Gamma(\text{E2}) = 8.076 \times 10^{-7} (E_i - E_f)^5 B(\text{E2}, J_i \rightarrow J_f) \text{ eV}, \tag{6}$$

where the $B(\text{E}\lambda)$ values are in units of e^2fm$^{2\lambda}$ and energies in MeV.

The results of the calculation are displayed in column I of Table 1. The resulting B(E2) excitation probability of the $|3_1^- \otimes 3_1^-; 2^+\rangle$ state from the ground state is about 300 times weaker than the known B(E2) value [36] for the transition to the first 2^+ state. The calculation also indicates a large branching ratio for the decay of this state into the first 3^- level. For the latter one has also a competing collective E3 transition with a large matrix element $\langle 3_1^- ||E3||3_1^- \otimes 3_1^-; 2^+\rangle$ corresponding to the one-phonon annihilation. However, because of the large multipolarity its contribution to the decay width is negligible.

Dealing with such a weak transition one has to consider possible small admixtures of other configurations which can be excited by strong transitions allowed in the boson space. The most important one in the region of the octupole two-phonon multiplet should be the 2_1^+ level, since the energy difference between these two states is estimated to be only a little more than 1 MeV, see Fig. 1. To take into account mixing in the $|3_1^- \otimes 3_1^-; 2^+\rangle$ state the model Hamiltonian was diagonalized for a set of 2^+ wave functions

$$|\Psi_{2^+}\rangle = \left\{ \sum_i S_i Q_{2^+ i}^+ + \sum_{\lambda_1 i_1 \lambda_2 i_2} \frac{D_{\lambda_1 i_1}^{\lambda_2 i_2} [Q_{\lambda_1 i_1}^+ Q_{\lambda_2 i_2}^+]_{2^+}}{\sqrt{1 + \delta_{\lambda_1,\lambda_2} \delta_{i_1,i_2}}} \right\} |\Psi_{\text{g.s.}}\rangle, \tag{7}$$

where the index i labels RPA root numbers for a given multipolarity λ^π. In this calculation all one- and two-phonon configurations built of $\lambda^\pi = 2^+, 3^-, 4^+$ phonons up to 13 MeV (i.e. including the complete isoscalar giant quadrupole resonance) were taken into account. The diagonalization with the wave functions of Eq. (7) yields eigen values and coefficients S_i, $D^{\lambda_2 i_2}_{\lambda_1 i_1}$ expressing the contributions of each one- and two-phonon configuration in the wave function, respectively.

Similar calculations were performed for all members of the two-octupole multiplet and the resulting energies are displayed in Fig. 1. The relevant transition strengths are summarized in column II of Table 1. One finds a negligibly small contribution of the isoscalar quadrupole resonance in the low-energy region, but a few percent admixture of the 2^+_1 configuration to the $|3^-_1 \otimes 3^-_1; 2^+\rangle$ state increases the excitation probability roughly by a factor of four. The decay width to the 3^-_1 state is practically unchanged since the magnitude of the E1 transition matrix element $\langle 3^-_1 \| M(\mathrm{E1}) \| 2^+_1 \rangle$ is comparable to the matrix element $\langle 3^-_1 \| M(\mathrm{E1}) \| 3^-_1 \otimes 3^-_1; 2^+ \rangle$.

To illustrate the effect of an admixture of the 2^+_1 component in the wave function of the $|3^-_1 \otimes 3^-_1; 2^+\rangle$ consider the following simplified two-state model. Let us write the wave function of the 3^-_1 and $|3^-_1 \otimes 3^-_1; 2^+\rangle$ states as

$$|\Psi_{|3^-_1\rangle}\rangle = 3^-_1 \,|\Psi_{\mathrm{g.s.}}\rangle , \tag{8}$$

$$|\Psi_{|3^-_1 \otimes 3^-_1; 2^+\rangle}\rangle = \left\{ \sqrt{\delta}\, 2^+_1 + \sqrt{1-\delta}\, |3^-_1 \otimes 3^-_1; 2^+\rangle \right\} |\Psi_{\mathrm{g.s.}}\rangle, \tag{9}$$

where $\delta \ll 1$ (in our calculation it is ≈ 0.01). We denote by the capital letter M the matrix element of a collective transition corresponding to one-phonon exchange and use the small letter m for weak transitions which are possible only due to rearranging of the fermions in the phonons. Then the γ-decay of the $|3^-_1 \otimes 3^-_1; 2^+\rangle$ state into the 3^-_1 state is determined by

$$B(\mathrm{E1}; |3^-_1 \otimes 3^-_1; 2^+\rangle \to 3^-_1) \sim |\sqrt{\delta}\, m^{\mathrm{E1}}_{2^+_1 \to 3^-_1} + \sqrt{1-\delta}\, m^{\mathrm{E1}}_{|3^-_1 \otimes 3^-_1; 2^+\rangle \to 3^-_1}|^2$$

$$\approx |m^{\mathrm{E1}}_{|3^-_1 \otimes 3^-_1|_{2^+} \to 3^-_1}|^2 \tag{10}$$

since $|m^{\mathrm{E1}}_{2^+_1 \to 3^-_1}| \approx |m^{\mathrm{E1}}_{|3^-_1 \otimes 3^-_1; 2^+\rangle \to 3^-_1}|$. On the other hand, for the γ-decay into the ground state

$$B(\mathrm{E2}; |3^-_1 \otimes 3^-_1; 2^+\rangle \to \mathrm{g.s.}) \sim |\sqrt{\delta}\, M^{\mathrm{E2}}_{2^+_1 \to \mathrm{g.s.}} + \sqrt{1-\delta}\, m^{\mathrm{E2}}_{|3^-_1 \otimes 3^-_1; 2^+\rangle \to \mathrm{g.s.}}|^2 \tag{11}$$

one has $|M^{\mathrm{E2}}_{2^+_1 \to \mathrm{g.s.}}|^2 \gg |m^{\mathrm{E2}}_{|3^-_1 \otimes 3^-_1; 2^+\rangle \to \mathrm{g.s.}}|^2$ and both terms give a contribution of the same order of magnitude. Thus, a small admixture of the 2^+_1 component affects the γ-decay properties of the two-phonon state into the ground state (see Table 1), but not into the one-phonon level.

The deduced transition probability for E1 decay $|3^-_1 \otimes 3^-_1; 2^+\rangle \to 3^-_1$ is equal to 0.73×10^{-3} $e^2\mathrm{fm}^2$. This is in good correspondence [2] to experimental values $B(\mathrm{E1}, |3^-_1 \otimes$

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[2] Theoretically, this transition was studied for ^{144}Sm within nuclear field theory [37] in reasonable agreement with the data, and within the QPM [38] which disagrees by several orders of magnitude. Therefore, we

$3^-_1; 2^+ \rangle \rightarrow 3^-_1) = 0.95 \times 10^{-3}$ e^{2}fm^2 and 1.20×10^{-3} e^{2}fm^2 for possible candidates reported in ^{96}Zr [8] and ^{144}Sm [10], respectively.

The QPM predictions presented here are mainly sensitive to the collectivity of the 3^-_1 state which determines the strength parameter of the residual interaction. To estimate how robust these predictions are we performed test calculations in which it was varied artificially by ±30 % with respect to the value determined by the experimental B(E3, g.s. $\rightarrow 3^-_1$). The results are included in columns Ia, IIa and Ib, IIb of Table 1, respectively. The γ-width of the $|3^-_1 \otimes 3^-_1; 2^+\rangle$ state for the decay into the ground state approximately changes by a factor of two for such a variation of the strength parameter while the decay width into the 3^-_1 state is stable within 20 %.

4. Proposed experimental test

The numbers presented in Table 1 can be converted to cross sections in a (γ, γ') experiment. The experimental goal is to identify the transitions and to determine spins of the intermediate and final state from the angular correlations ($0 \rightarrow 2 \rightarrow 0$ and $0 \rightarrow 2 \rightarrow 3$) by measuring at two angles (90° and 130°). Based on the recent experimental experience [26] the g.s. transition of the two-phonon 2^+ level should be easily measurable with a EUROBALL Cluster detector even for the least favourable case (column Ia). The observation of the $2^+ \rightarrow 3^-$ transition with roughly half the γ-energy is more challenging because of the steeply rising background in NRF experiments towards lower γ-energies.

A Monte Carlo simulation of the background expected in a ^{208}Pb(γ, γ') experiment at a bremsstrahlung end point energy of 7 MeV was performed with the code GEANT [39]. Two EUROBALL Cluster detectors were assumed under 90° and 130° in a geometry very similar to the one described in Ref. [26]. The contributions of resonant nuclear excitations to the background were neglected. The incoming photon flux was normalized to correspond to one week of beam time under the experimental conditions described in Ref. [26]. The background spectrum resulting after consideration of the Cluster efficiency properties with its statistical fluctuations is displayed in Fig. 2 for a photon energy interval $E_\gamma = 2.52$–2.58 MeV. It can be well described by a simple exponential function (dashed line). The effect of a $|3^-_1 \otimes 3^-_1; 2^+\rangle \rightarrow 3^-_1$ transition with a photon energy of $5.16 - 2.62 = 2.54$ MeV (see Fig. 1) is demonstrated by the superposition of a Gaussian (dotted line). Its width is taken from the experimentally determined resolution of the Cluster module and the height is normalized to the partial decay width predicted by the QPM (column II of Table 1). The transition can be clearly identified above the background. At 90°, despite the somewhat poorer peak-to-background ratio this still holds true on a 3σ level.

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repeated the calculation for ^{144}Sm using the present QPM approach. The result is B(E1, $|3^-_1 \otimes 3^-_1; 2^+\rangle \rightarrow$ 3^-_1) $= 2.9 \times 10^{-3}$ e^{2}fm^2 in excellent agreement with Ref. [37] taking into account their smaller effective charges. We have no explanation for the extremely small value reported in Ref. [38].
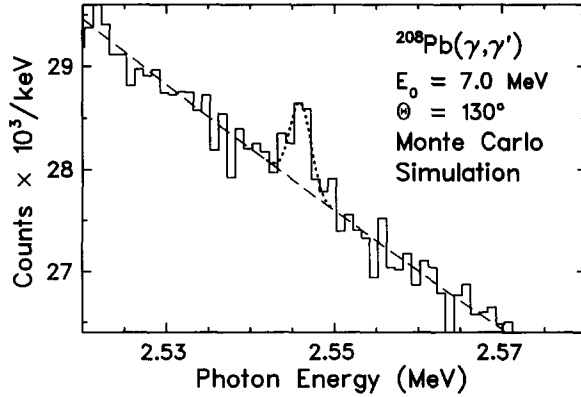
Fig. 2. Monte Carlo simulation of a spectrum of the ^{208}Pb(γ, γ') reaction for a bremsstrahlung end point energy of 7 MeV and a scattering angle of 130° using a EUROBALL Cluster detector. The statistics of the "data points" presented in form of a histogram correspond to one week of beam time under the experimental conditions of Ref. [26]. The background is well described by a simple exponential function (dashed line) for the shown energy interval. The signal of the predicted $B(E1, |3_1^- \otimes 3_1^-; 2^+\rangle \to 3_1^-)$ transition at 2.546 MeV (dotted line) is simulated by a Gaussian with a width determined by the experimental resolution of the Cluster module and the height normalized to the decay width predicted by the QPM.

5. Conclusions

To summarize, we have discussed the feasibility of a new experimental approach to identify the long-sought two-phonon octupole vibration in ^{208}Pb. By measuring simultaneously the E2 decay to the g.s. and the E1 decay to the 3_1^- level in the (γ, γ') reaction, the 2^+ member of the $3_1^- \otimes 3_1^-$ multiplet can be unambiguously identified. The description of the relevant transition probabilities has been derived in the QPM including mixing of one- and two-phonon configurations in the wave functions of excited states. The numerical results indicate that such an experiment should be possible with the new generation of large volume HPGe detectors like the EUROBALL Cluster module. At present, the only machine worldwide providing a sufficient bremsstrahlung flux in the energy range required is the Superconducting DArmstadt electron LINear ACcelerator S-DALINAC in Darmstadt. From the experimental information on the 0^+ and 2^+ states (eventually combined with new results on the 6^+ state from recent efforts to repeat the HI Coulomb excitation experiment of Ref. [22] with much improved sensitivity [40]) one may finally elucidate the degree of harmonicity of octupole vibrations in a doubly magic nucleus.

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