Two-phonon octupole excitation in ^{146}Gd

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Based on experimental evidence from the ^{144}Sm$(\alpha,2n)$ reaction, the 3484.7-keV 6^+ state in ^{146}Gd is identified as the highest-spin member of the $3^- \otimes 3^-$ two-phonon octupole quartet. A previously unknown γ line of 1905.8 keV and $E3$ character feeding the 3^- octupole state has been observed. These results represent the first observation of a $6^+ \rightarrow 3^- \rightarrow 0^+$ cascade of two $E3$ transitions in an even-even nucleus and provide strong support for the interpretation of the 6^+ state as a two-phonon octupole excitation.

In the harmonic approximation for noninteracting phonons, a degenerate two-phonon multiplet should occur at twice the energy of the one-phonon state. For low-energy quadrupole ($\lambda = 2$) shape vibrations, multiphonon states have been characterized in many even-even nuclei; however, these structures frequently exhibit large anharmonicities, even in nearly spherical nuclei [1]. The octupole ($\lambda = 3$) mode in doubly magic nuclei likely comes much closer to the ideal harmonic oscillator as a result of the smaller vibrational amplitudes and the larger number of particles participating in the motion. Indeed, a two-phonon $3^- \otimes 3^-$ quartet ($J^\pi = 0^+, 2^+, 4^+$, and 6^+) should occur at twice the energy of the octupole phonon and is expected to decay with $B(E3)$ values which are twice that of the $3^- \rightarrow 0^+$ transition [1,2]. Unfortunately, two-phonon excitations of the octupole type have proven difficult to identify.

It is not surprising that ^{208}Pb, with its 3^- first-excited state at 2614 keV and $B(E3; 3^- \rightarrow 0^+)$ of 34 W.u. [3], has been the subject of many searches for the two-phonon octupole quartet at about 5.2 MeV, that is, at $2\hbar\omega_3$ [4]. Firm evidence, an observed cascade of two $E3$ transitions from a 0^+ state at 5241 keV, for the lowest-spin member of the two-phonon quartet in ^{208}Pb has been obtained [5]. In addition, candidates for the 2^+ and 4^+ members of the quartet at nearly harmonic energies have been proposed, but these identifications are based primarily on energy arguments and $E1$ transition rates; the signature cascade of $E3$ transitions was not observed [6]. Transfer reaction and inelastic light-ion scattering studies [7] support these assignments, but no clear candidate for the 6^+ member in ^{208}Pb has been identified, although a number of experiments have been performed specifically to observe this excitation.

When the nucleons of one kind form a closed-shell configuration and the Fermi surface for the other kind lies near the middle of a major shell, a situation characterized by significant anharmonic effects might be expected. These circumstances may occur, in particular, when one of the low-energy excitations, such as $p_{3/2} \rightarrow g_{9/2}$, $d_{5/2} \rightarrow h_{11/2}$, or $f_{7/2} \rightarrow i_{13/2}$, contributes prominently to the octupole mode. The nucleus ^{146}Gd represents indeed such a case, where the $\pi h_{11/2} d_{5/2}^{-1}$ component contributes about half of the 3^- excitation [8].

For many years, it has been recognized that ^{146}Gd, the only known even-even nucleus besides ^{208}Pb with a 3^- first excited state, displays many of the properties of a doubly magic nucleus [9], with $B(E3; 3^- \rightarrow 0^+) = 37$ W.u. [10]. States of two-phonon octupole character in ^{147}Gd and ^{148}Gd built on the $\nu f_{7/2}$ ground state and the $(\nu f_{7/2}^2)6^+$ state, respectively, have been characterized through the observation of cascades of two stretched $E3$ transitions [11–13]. Their energies and the measured $E3$ strengths are quite accurately predicted in elementary calculations. Similar two-phonon states have been identified in ^{146}Sm, but they have not been fully characterized through transition rate measurements [14].

The reason why two-phonon octupole states in ^{147}Gd and ^{148}Gd have been observed is that these excitations are fortuitously yrast and are, consequently, well populated in fusion-evaporation reactions. In ^{148}Gd the yrast 6^+ state is attributed to the coupling of the two valence $f_{7/2}$ neutrons outside the $N = 82$ closed shell and lies at 1811-keV excitation. The 6^+ two-phonon octupole state is expected at approximately 700 keV above the yrast 6^+ and thus receives much smaller population in fusion-evaporation reactions.

In view of the success in identifying two-phonon octupole states in nuclei such as ^{147}Gd and ^{148}Gd, it is not surprising that the more straightforward excitations involving only two phonons have also been sought in ^{146}Gd, the core nucleus. About 20 years ago, an extensive search conducted in the

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region of the expected two-phonon excitations with the ^{144}Sm$(\alpha,2n)$ reaction [15] resulted in a significant extension of the knowledge of the ^{146}Gd level structure. Among the observed states were 6^+ states at 3457 and 3485 keV, which were characterized by $E1$ decay to the 5^- yrast state. These states were interpreted as the highest-spin member of the two-phonon octupole quartet and the expected $(d_{5/2}^{-1}g_{7/2}^{-1})6^+$ two-proton hole state, but a distinction between the possible assignments for these states was not possible from the available data.

In the years since these studies, γ-ray detection methods have made significant strides, particularly in the application of arrays of large-volume Ge detectors. In view of these advances and the availability of α-particle beams, the facilities at the University of Cologne were appropriate for reinvestigating the excited states in ^{146}Gd with the ^{144}Sm$(\alpha,2n)$ reaction. A beam energy of 26.3 MeV had been demonstrated to produce the optimal population of nonyrast states, and higher α-particle energies yield only additional yrast population [15]. The ^{144}Sm target (3.0 mg/cm^2, enriched to 97.6%, on a thick gold backing) was surrounded by a compact array of nine individual Ge detectors at an average distance of 10 cm and angles of 90°, $\pm45°$, and $\pm 35°$ to the beam direction. Five of these detectors had anti-Compton shields. In addition, a EUROBALL cluster detector comprised of seven coaxial Ge detectors was placed at 90°. This detector can be used as a nonorthogonal Compton polarimeter by employing all possible combinations of adjacent crystals, one as a scatterer and the other as an analyzer. The characteristics and figures of merit of such a polarimeter have been examined in detail [16]. The polarization sensitivity as a function of the γ-ray energy was determined by using internal calibration points of transitions with well-known character and multipolarity. The cluster detector, operated in "add-back" mode, also served to increase the efficiency for high-energy γ rays. Additional information used for the characterization of the γ rays was the anisotropy, which was calculated as a ratio of the γ-ray intensity at 40° (the data taken at 35° and 45° were summed in order to increase the statistics) and at 90°. The total photopeak efficiency of the array was 1.1% at 1.33 MeV. This experimental arrangement provided excellent γ-γ coincidence and very useful γ-ray polarization data, but only limited information on the angular distributions of the γ rays. The angular distribution data were generally sufficient to obtain the sign of a_2, which, combined with the polarization information, permitted the determination of multipolarities of most transitions. Overall, the detection sensitivity of this measurement was about 10 times better than the previous measurements [15].

Because the angular momentum transferred in the ^{144}Sm$(\alpha,2n)$ fusion-evaporation reaction is low, states of low-to-intermediate spin lying above the yrast line in ^{146}Gd are populated in the region where the two-phonon octupole states are anticipated. We have generally confirmed the results of the previous study [15] and, in only a few cases, have they been modified. In addition, we have placed 44 new levels and γ rays deexciting 26 levels seen in in-beam fusion evaporation reactions for the first time [17]. Most of these new states lie within 2 MeV of the yrast line. In addition, we have been
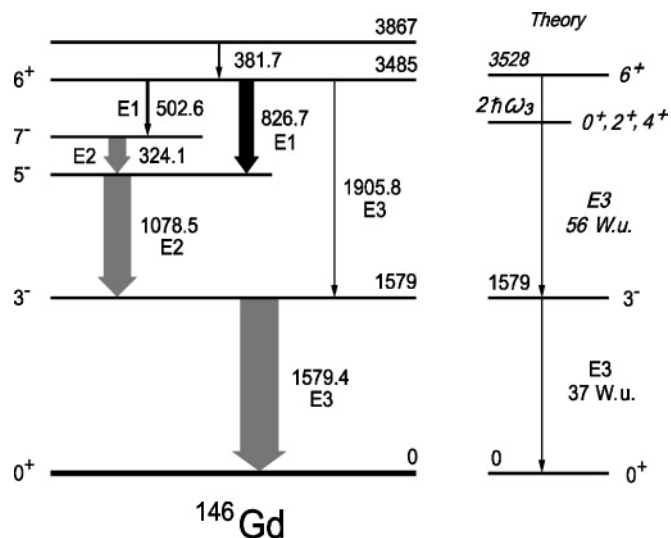

FIG. 1. Partial level scheme of ^{146}Gd showing transitions that populate and deexcite the 3484.7-keV 6^+ level. The thicknesses of the γ lines shown in black represent their intensities; those shown in gray have had the intensities divided by 20. Calculated results for the energy and $E3$ decay strength of the $(3^- \otimes 3^-)6^+$ are given to the right in italics. The $B(E3) = 37$ W.u. is derived from the 1579-keV level $t_{1/2}$ value of Ref. [11]. See text.

able to detect crucial new decay branches from previously known in-beam fusion evaporation reaction levels. As will be shown subsequently, these weak branches permitted us to increase the confidence of our spin and parity assignments. The aforementioned 6^+ levels at 3457 and 3485 keV characterized by Yates *et al.* [15] were also observed in the current work, and important new information for both states has emerged. The new data firmly place a Doppler-broadened 1877-keV γ ray as the transition from the 3457-keV state to the 3^- level and exclude the previous 6^+ assignment for that state. In Ref. [15], the 3485-keV state was assigned as $J^\pi = 6^+$ from an observed 826.9-keV $E1$ transition to the yrast 5^- state and the population yield of the state. The higher sensitivity of the new measurements permitted us also to observe a 502.6-keV transition from this level to the yrast 7^- state (see Fig. 1). From the angular distribution and polarization data (Table I), this γ ray was also firmly assigned as an $E1$ transition, so the level spin must indeed be 6^+. Furthermore, a 1905.8-keV γ ray from this level is observed to feed the 3^- octupole state. Conclusive supporting evidence is adduced from the spectrum of γ rays in coincidence with the newly identified 381.7-keV transition, which weakly populates the 3485-keV state. Figure 2 illustrates portions of this spectrum and confirms that all three of these transitions originate from the 3485-keV level. We have thus established a firmly assigned 6^+ state with a newly adopted energy of 3484.7 ± 0.3 keV that decays by a cascade of two $E3$ transitions to the ground state. Such a $6^+ \rightarrow 3^- \rightarrow 0^+$ cascade is, of course, the expected decay signature of the 6^+ member of the two-phonon octupole quartet.

The primary contributions to the anharmonicities of the two-phonon $3^- \otimes 3^-$ states arise from the microscopic composition of the octupole phonon, where the $\pi h_{11/2}d_{5/2}^{-1}$

TABLE I. Data from the ^{144}Sm$(\alpha,2n\gamma)$ reaction at $E_\alpha = 26.3$ MeV for γ rays associated with the feeding and deexcitation of the 3484.7-keV 6^+ two-phonon octupole state of ^{146}Gd.

Energy (keV)	Intensity	Anisotropy[a] (40°/90°)	Polarization Exp. Calc[b]	Adopted multipolarities
324.1(1)	458(22)	1.49(10)	0.51(14) 0.56	$E2$[c]
381.7(3)	1.5(6)	0.9(5)	0.2(9)	
502.6(1)	3.2(4)	0.67(12)	0.36(24) 0.21	$E1$
826.7(1)	20.5(14)	0.84(8)	0.46(19) 0.33	$E1$[c]
1078.5(1)	712(37)	1.47(11)	0.61(21) 0.62	$E2$[c]
1579.4(1)	1000	1.63(10)	0.55(20) 0.84	$E3$[c]
1905.8(6)	1.3(6)	1.2(9)	0.8(10) 0.70	$E3$

[a]Anisotopy values less than unity correspond to negative a_2 values, and those greater than unity correspond to positive a_2 values.
[b]Polarization values expected for the adopted pure multipolarities are indicated.
[c]Multipolarity is well established from angular distribution coefficients, from a_2 and a_4 values, and from internal conversion coefficients [15].

configuration is dominant [8]. The observed anharmonicities in the $\pi h_{11/2} \otimes 3^-$ multiplet in ^{147}Tb [8] are of the same origin and can be used to provide the required input for calculating the two-phonon anharmonicities. The coupling process is shown

FIG. 2. Portions of the γ-ray spectrum observed at 90° in coincidence with the 381.7-keV transition selected in all the detectors of the array. The γ rays of interest (see Fig. 1) are labeled with their energies (in keV).

in the interaction diagram of Fig. 3(a), which represents the exchange of the particle with the phonon. The energy shifts for the seven members of the multiplet in ^{147}Tb from the $\hbar\omega_3$ unperturbed energy are

$$\Delta E = -7 \times \begin{Bmatrix} 3 & 11/2 & 5/2 \\ 3 & 11/2 & J \end{Bmatrix} \times \frac{M^2}{2\Delta}.$$

From the splitting of the $J^\pi = 17/2^+$ and $15/2^+$ yrast states, the highest-spin members of the $\pi h_{11/2} \otimes 3^-$ septet in ^{147}Tb, and taking the energy denominator, $\Delta = \varepsilon(h_{11/2}) - \varepsilon(d_{5/2}) - \hbar\omega_3$, as 1.5 MeV [11], the particle-phonon interaction matrix element is calculated to be $M = 1.13$ MeV.

The two-phonon exchange process illustrated in the fourth-order diagram of Fig. 3(b) is analogous to the exchange coupling in Fig. 3(a). As the interaction vertices in Figs. 3(a) and 3(b) are the same, we can use the empirical value of M as extracted above from the ^{147}Tb data. Therefore, the anhamonicities for the two-phonon states are

$$\Delta E(3^- \otimes 3^-) = 98 \times \begin{Bmatrix} 11/2 & 5/2 & 3 \\ 5/2 & 11/2 & 3 \\ 3 & 3 & J \end{Bmatrix} \times \frac{M^4}{\Delta^3}.$$

For the 6^+ member, this calculation gives

$$\Delta E(3^- \otimes 3^-)6^+ = 0.846 \times (1.1 \text{ MeV})^4/(1.5 \text{ MeV})^3$$
$$= +0.37 \text{ MeV},$$

and the predicted level energy, $E(3^- \otimes 3^-)6^+ = 3.53$ MeV, is in good agreement with experiment. Additional details of the calculations are given in Ref. [11].
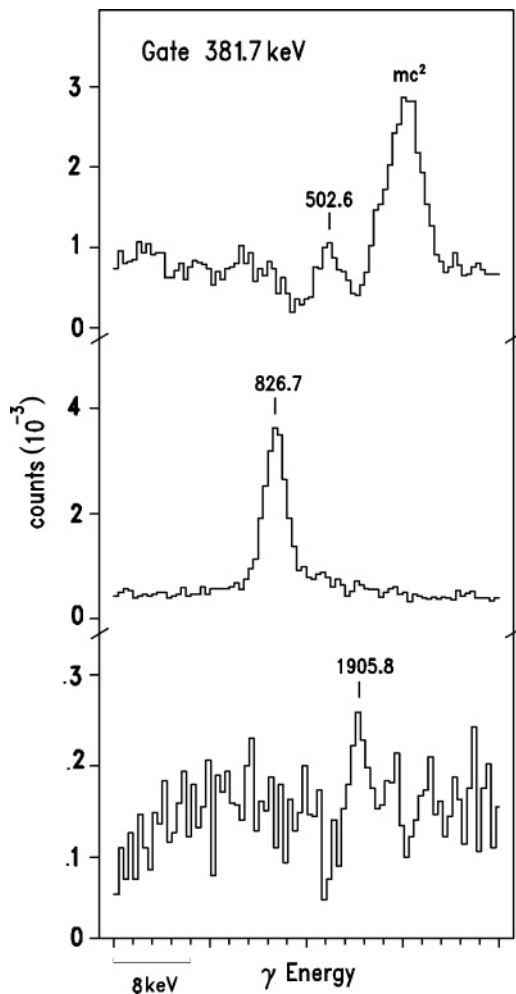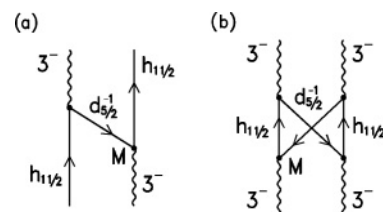

FIG. 3. (a) Coupling of the $h_{11/2}$ valence proton of ^{147}Tb to the core octupole phonon. (b) Phonon coupling in ^{146}Gd.

An analogous diagram specifies the anharmonicity of the $B(E3)$ transition strength. With the empirical $B(E3)$ value of the ^{146}Gd core, 37 W.u., and an estimated 5 W.u. for the $(\pi h_{11/2} \pi d_{5/2}^{-1})3^- \to 0^+$ single-particle strength, one obtains a reduction in the $B(E3)$ of 18 W.u., that is, $B(E3; 6^+ \to 3^-) = 56$ W.u., as explained in Ref. [11]. The validity of this approach has already been discussed and tested experimentally in calculating the $E3$ strengths of the decays of the two-phonon octupole states in ^{147}Gd and ^{148}Gd [11–13].

From the predicted $B(E3; 6^+ \to 3^-)$ of 56 W.u. and the data in Table I, the lifetime of the 3484.7-keV state is calculated to be 10 ± 5 ps, with the large uncertainty arising primarily from the γ-ray branching ratio. The $B(E1)$ values of the 502.6- and 826.7-keV transitions are then 3×10^{-5} and 4×10^{-5} W.u., respectively. In addition to these observed $E1$ transitions, it might be expected that an $E2$ transition to the 4^+ $(\pi d_{3/2} d_{5/2}^{-1})$ state at 2612 keV would occur. If this γ ray is present, it is below the detection sensitivity of the present measurements ($I_\gamma < 0.4$ intensity units) and would correspond to a $B(E2) < 0.04$ W.u., a value that is certainly small but not remarkable.

With the identification of the 6^+ two-phonon octupole state, it is only natural to ask where the remaining members of the quartet are located. It seems unlikely that the decays of these states would occur by $E3$ transitions, as is the case for the 6^+ member of the quartet. Nonetheless, candidates for each of these states exist.

Several 4^+ states have been observed in the present work, but only two of these occur below 3.4 MeV. The lowest of these, at 2612 keV, has been firmly assigned to the $\pi d_{3/2} d_{5/2}^{-1}$ multiplet [15]. The next level, at 2967 keV, decays only by an $E1$ transition to the 3^- octupole phonon and has been suggested as the $(3^- \otimes 3^-)4^+$ state, primarily from energy considerations [15]. Although this state is 190 keV below $2\hbar\omega_3$ and no clear-cut signature for the two-phonon character of this state is observed, no other configuration assignment seems more reasonable.

Contrary to the difficulties in assigning the 4^+ quartet member, the assignments of the 2^+ and 0^+ two-phonon states are more promising. Configuration-specific information comes from β^+/EC decay of the 8-s ground state of ^{146}Tb [18], with the assigned configuration $(\pi d_{5/2}^{-1} \nu d_{3/2}^{-1})1^+$ [19]. Here, the decay of the odd proton leads to the $(\pi d_{5/2}^{-2})0^+$ configuration largely contributing to the ^{146}Gd proton pair vibration state whereas decay of a paired $d_{5/2}$ proton populates also the $(\pi d_{5/2}^{-2})2^+$ state. Two $d_{5/2}$ proton holes play, of course, a significant role in the two-phonon octupole states in ^{146}Gd and might well admix with the $(3^- \otimes 3^-)0^+$ and 2^+ levels. The ^{146}Tb β decay excites two 2^+ states, at 3186 and 3233 keV in ^{146}Gd, that can be identified with these configurations, with a slight preference of $(3^- \otimes 3^-)2^+$ for the higher state. We also note that these states have been populated in (p,t) experiments with small cross sections and $L = 2$ [20].

Four excited 0^+ states are known in ^{146}Gd below 4 MeV. The lowest of these, at 2165 keV, was ascribed to the proton pair vibration based on its observed $E0$ decay and energy arguments [21]. The large cross section for population of the 3020-keV 0^+ state in the (p,t) reaction led to its unequivocal characterization as the neutron pair vibration [22]. This state is populated in the β decay of the ^{146}Tb 1^+ state. The next higher 0^+ state at 3485 keV, originally observed in internal conversion electron measurements following the $(\alpha,2n)$ reaction [23], is the highest state observed in ^{146}Tb 1^+ β decay to be fed with significant β intensity and is, therefore, the best candidate for the $(3^- \otimes 3^-)0^+$ quartet member. Its energy is in sharp contrast with our calculation, which predicts it very close to $2\hbar\omega_3$.

In conclusion, we have identified for the first time the fully aligned 6^+ two-phonon octupole excitation in an even-even nucleus, ^{146}Gd, by observation of the $6^+ \to 3^- \to 0^+$ cascade of $E3$ transitions. In addition, we have proposed candidates for the 4^+, 2^+, and 0^+ members of the $3^- \otimes 3^-$ quartet. Our results provide the first evidence on the nuclear octupole degree of freedom governing the interactions between octupole phonons in nuclei. Whereas the energy of the maximally aligned 6^+ member of the quartet is found to be in excellent agreement with expectations for the pure octupole-octupole interaction, our proposed candidates for the 4^+ and 0^+ members are apparently distorted by admixtures from nonoctupole contributions.

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