Harmonic Two-Phonon γ-Vibrational State in Neutron-Rich ^{106}Mo

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The neutron-rich ^{106}Mo nucleus has been studied by prompt γ-ray spectroscopy following the spontaneous fission of ^{248}Cm. The characteristics of the rotational band built on a state at 1435 keV identifies the band head as the best candidate for a harmonic double-phonon $K^\pi = 4^+$ γ-vibrational state observed so far.

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The advent of large and efficient γ-ray multidetector arrays expands possibilities for observing medium-spin states in neutron-rich nuclei formed as fission fragments in spontaneous or induced fission. A primary interest in extending the experimental data on neutron-rich nuclei is to search for changes in nuclear structure as isospin varies. But data on neutron-rich nuclides may also shed new light on nuclear structure questions that have been the focus of discussions over decades. One of these controversial subjects is the existence of two-phonon vibrational states in deformed nuclei [1–5]. Numerous low-lying rotational bands based on one-phonon quadrupole vibrations have been observed in these nuclides. When the vibration is ascribed to one phonon and has no component of angular momentum along the symmetry axis ($K = 0$), it is called a β vibration; when the vibrational phonon has a component of angular momentum along the axis given by the quantum number $K = 2$, it is called a γ vibration. The characteristics of one-phonon vibrations are well known experimentally and theoretically. The situation for the two-phonon bands, $\beta\beta$ with $K = 0$, $\beta\gamma$ with $K = 2$, and $\gamma\gamma$ with $K = 4$ or $K = 0$, is not as clear. Some models [1] predict that two-phonon states do not exist as collective levels. Others models, e.g., [2,3], claim that some collective states may exist, but that large anharmonicities will be present in the energy spectrum. Recently the first experimental evidence was found [6] for a $K^\pi = 4^+$ state in ^{168}Er with many of the characteristics expected for a two-phonon vibration. Although the observed state appears higher than the pairing gap, and so could in principle be of complicated multiparticle structure, its lifetime corresponds to the degree of collectivity predicted by the theoretical calculations. Only one state could be observed in the rotational band based on the proposed $\gamma\gamma$ level. A systematic search for two-phonon states was undertaken by Wu *et al.* [4,7], and several $K^\pi = 4^+$ candidates were proposed in the $A \sim 160$ region of nuclei with different degrees of anharmonicities and collectivity. Discussion of the properties and interpretation of these levels by Burke [5] added interest to the problem of the existence and details of two-quadrupole phonon states in this well-studied region of deformed nuclei. In heavier nuclei, only one $\gamma\gamma$ candidate has been observed [8]. This was in ^{232}Th where the 4^+ state was reached by Coulomb excitation. The observed band based on this state, although having few identified members, exhibits many characteristics of a band based on a two-phonon γ level. The excitation energy of the 4^+ state is slightly less than twice that of the single-phonon vibration, but lower than the pairing gap. Thus it is necessarily of collective character. The present Letter reports the observation of a well-developed rotational band in the neutron-rich deformed ^{106}Mo nucleus, which lies in a different region of the nuclear chart. This new band shows even more completely the characteristics expected for a band based on a *harmonic* two-phonon γ vibrational state with $K^\pi = 4^+$.

The ^{106}Mo nucleus was obtained in this work as a secondary fragment in the spontaneous fission of the ^{248}Cm isotope. The source was prepared by mixing about 5 mg of ^{248}Cm (yielding a fission rate of roughly 6.3×10^4 fissions/s) in the form of oxide with 65 mg of KCl and compressing the mixture into a 7 mm diameter pellet. In this way the fission fragments were stopped within a short time ($\sim$1 ps) and almost all prompt γ rays were emitted at rest. The radioactive source was placed in the center of the Eurogam2 array [9] located at the Centre de Recherches Nucléaires in Strasbourg. The Eurogam2 array, consisting in this experiment of 52 escape-suppressed spectrometers using 124 Ge detector elements, was augmented by the addition of 4 LEPS detectors. The acquisition system was triggered only when the number of Ge triggers (unsuppressed) was greater than 5. This considerably reduced events associated with β decay, whereas the events from prompt fission were much less affected since they have an average γ-ray multiplicity of roughly 10. A total of $\sim 2 \times 10^9$ threefold or higher-fold coincidence events was collected.

Before this work, several levels in ^{106}Mo had been identified in the β decay of ^{106}Nb [10] and the yrast band was observed in an experiment similar to the present

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one, but performed with fewer γ-ray detectors [11]. The extension of the level scheme, as shown in Fig. 1, was obtained by using one-dimensional spectra of γ rays in coincidence with two or three γ-ray energies. The gates were set on transitions either in ^{106}Mo or in the Xe isotopes, which are the fission partners of ^{106}Mo. Spin and parity assignments are based on previous work, on the observed decay paths of the levels, and on triple angular correlations of γ rays. The analysis of triple angular correlations of γ rays following spontaneous fission and detected in the Eurogam array is described in detail elsewhere [12].

Among the neutron-rich even-even Mo isotopes, ^{106}Mo has the lowest excitation energy for the first 2^+ state, $E(2_1^+)$, and the highest $E(4_1^+)/E(2_1^+)$ ratio, 3.04, which is close to the limit of 3.33 for a rigid rotor. This is an indication that the maximum deformation is attained for this isotope. A partial decay scheme obtained earlier [10] had suggested a rigid triaxial description of ^{106}Mo, with a rather large nonaxiality parameter ($\gamma = 19°$). However, such an interpretation is unable to describe the high-spin states obtained more recently [11] and in the present work. The predictions of the triaxial model [13] for these states lie at far too high excitation energies. A better description of ^{106}Mo is to consider it as an axially symmetric nucleus. The level sequence starting at 710.4 keV is then identified as the rotational band based upon the one-phonon γ vibrational level at 710.4 keV. This is supported by the fact that the ground-state band and the one-phonon γ band exhibit, as expected, similar moments of inertia. Indeed, fits to the two bands using the second order rotational energy formula

$$E(I,K) = E_K + A[I(I+1) - K^2] + B[I(I+1) - K^2]^2 \quad (1)$$

yield nearly the same value for the inertia parameters A: 25.5 and 25.3 keV for the yrast band and the γ band, respectively.

A striking feature of the level scheme is the presence of the band built on a level at 1434.6 keV excitation energy. This is well *below* the neutron pairing gap

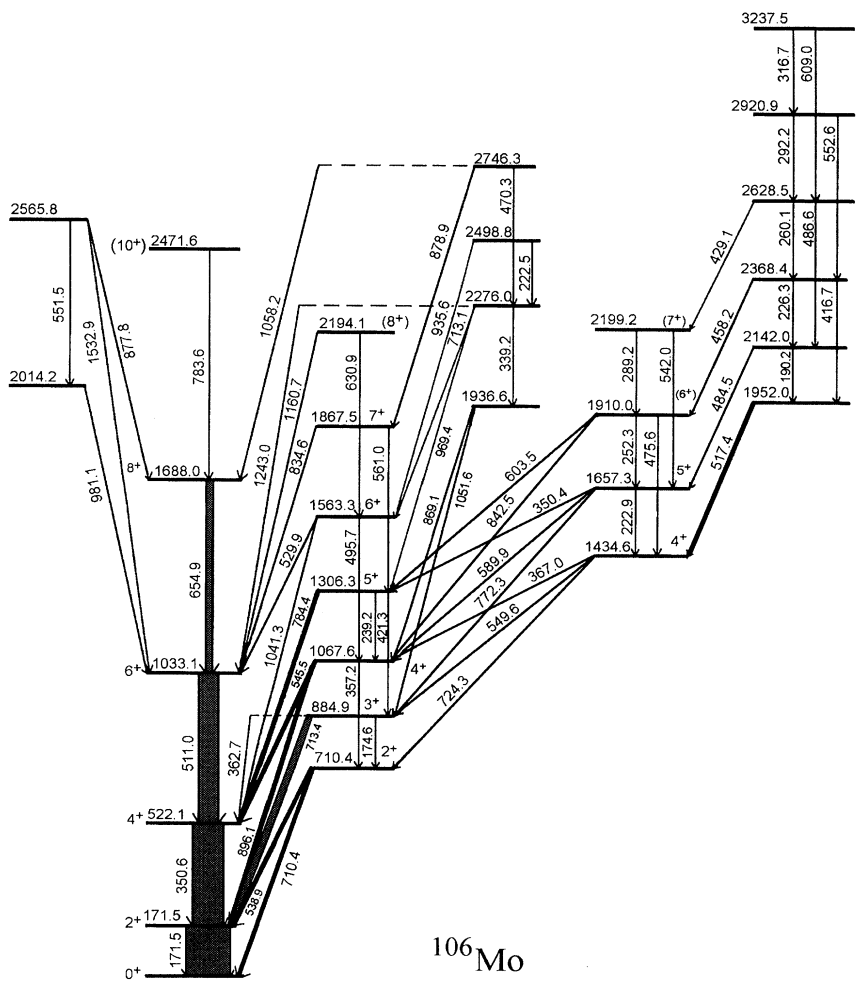

FIG. 1. Partial decay scheme for ^{106}Mo. The widths of the arrows are proportional to the γ-ray intensities.

energy $2\Delta_n \sim 2.1$ MeV and the proton pairing gap energy $2\Delta_p \sim 1.7$ MeV, as calculated from known nuclear masses. It thus is built on an intrinsic state of *collective* structure. The inertia parameter A of the ban is equal to 22.6 keV, and is close to the values for the ground-state and one-phonon γ bands. Such a feature is foreseen for quadrupole vibrational excitations since the moments of inertia are not expected to be altered greatly through small oscillations around the ground-state shape. The decay pattern of the members of this second sideband strongly suggests a spin assignment of $I = 4$ to the bandhead. The $I = 4$ assignment is corroborated by the similar ratios of directional correlations from oriented states (DCOR) obtained for the 724.3-538.9-171.5 and 357.2-538.9-171.5 keV γ cascades: DCOR = 0.86(6) and 0.92(4), respectively. In the same way, the $5_2^+ \rightarrow 3_1^+ \rightarrow 2_1^+ \rightarrow 0_1^+$ cascade yielding DCOR = 0.90(5) compares with the $5_1^+ \rightarrow 3_1^+ \rightarrow 2_1^+ \rightarrow 0_1^+$ cascade having DCOR = 0.99(5). Moreover, computed DCOR values, taking into account the geometries selected in the Eurogam array, the finite size of the detectors, and the γ-ray efficiencies, reproduce satisfactorily the experimental DCOR values when the $\Delta I = 0$ or $\Delta I = 1$ transitions in the different cascades are assumed to be predominantly $E2$ radiations. Further support for the spin assignment is provided by data from the β decay of ^{106}Nb [10]. This decay, with a Q_{β^-} value of 9.3 MeV, populates with roughly the same strength the three 4^+ states: the intensity $I_{\beta^-} = 21(7)$, 15(6), and 26(7) in arbitrary units for the 4_1^+, 4_2^+, and 4_3^+ states, respectively (the I_{β^-} value for the 4_1^+ state takes into account the new placing of the 784.4 keV transition in the level scheme). The strong $\Delta I = 2$ transitions observed in the present work between the second sideband and the $K = 2$ band exclude a negative parity assignment to the 1434.6 keV state. There are no transitions from the second sideband to the $K = 0$ ground-state band. We thus conclude that the bandhead at 1434.6 keV has $K = 4$ and positive parity.

There are two possibilities for the nature of this bandhead: either it is a candidate for a two-phonon γ state or it corresponds to a collective hexadecapole vibration. We will now make strong arguments in favor of the identification of the 1434.6 keV level as a two-phonon γ vibrational state.

(i) The $E_{K=4}/E_{K=2}$ ratio is equal to 2.00, the expected value for a $K^\pi = 4^+$ harmonic two-phonon vibrational state.

(ii) The dynamical moment of inertia of the proposed two-phonon band with bandhead at 1434.6 keV is almost equal to that of the one-phonon band with bandhead at 710.4 keV. This is shown by the similar slopes of the plots of the angular momentum along the rotational axis against the rotational frequency for the two bands (see Fig. 2). Such a feature has been suggested as a characteristic of multiphonon vibrational bands [7].

(iii) The branching ratios for decays out of the $K = 2$ and $K = 4$ bands agree very well with those predicted for members of bands based on one-phonon and two-phonon vibrational levels. For pure bands, i.e., no mixing between them, the following formula [14] gives the reduced electromagnetic transition probability between states with initial quantum numbers I_i, K_i, π_i, and final quantum numbers I_f, K_f, π_f:

$$B(\sigma\lambda; I_iK_i\pi_i \rightarrow I_fK_f\pi_f) = (1 + \delta_{K_f0})\langle I_iK_i\lambda(K_f - K_i)\,|I_fK_f\rangle^2\,|\langle K_f\pi_f|M(\sigma\lambda, K_f - K_i)\,|K_i\pi_i\rangle|^2. \quad (2)$$

Neglecting high multipolarities, i.e., $\lambda > 2$, this formula allows only electric quadrupole ($E2$) decays between bands with $\Delta K = 2$ and $\pi_i = \pi_f$. Furthermore,

$$\langle K_f(n_\gamma - 1)\,|M(E2, K_f - K_i = -2)|K_in_\gamma\rangle = \sqrt{n_\gamma}\langle K_f'(n_\gamma' = 0)\,|M(E2, K_f' - K_i' = -2)\,|K_i'(n_\gamma' = 1)\rangle, \quad (3)$$

where n_γ (n_γ') is the number of aligned γ vibrational phonons in the intrinsic state. Assuming that the intrinsic quadrupole moments of the $K = 4$ and $K = 2$ bands are equal, as reasonable assumption in view of the almost equal inertia parameters, one can apply these formulas to predict the ratios

$$R = [Y(I_1, K = 4 \rightarrow I_1 - 2, K = 4)/Y(I_1, K = 4 \rightarrow I_1 - 2, K = 2)] \times [Y(I_2, K = 2 \rightarrow I_2 - 2, K = 0)/Y(I_2, K = 2 \rightarrow I_2 - 2, K = 2)], \quad (4)$$

where the quantities Y are the γ decay probabilities from the appropriate levels I, K. Predicted values of the ratio R are compared with experimentally observed values (for transitions for which reasonably accurate decay probabilities have been measured) in Table I. It is seen that the agreement is excellent, supporting the assumptions made in the predictions. Moreover, this agreement rules out the possibility of a pure hexadecapole vibrational character of the $K^\pi = 4^+$ band. In such a case the decay to the one-phonon γ-vibrational state would imply the annihilation of a phonon and the creation of a different phonon, rendering thereby the use of Eq. (3) with $n_\gamma = 2$ inadequate.

Within the harmonic limit, collective models predict another rotational band built on a state with the two phonons coupled to $K = 0$ at an excitation energy close to that of the $K = 4$ bandhead. This band has not been identified in the present work, but the lower spin states of such a band are not expected to be appreciably populated

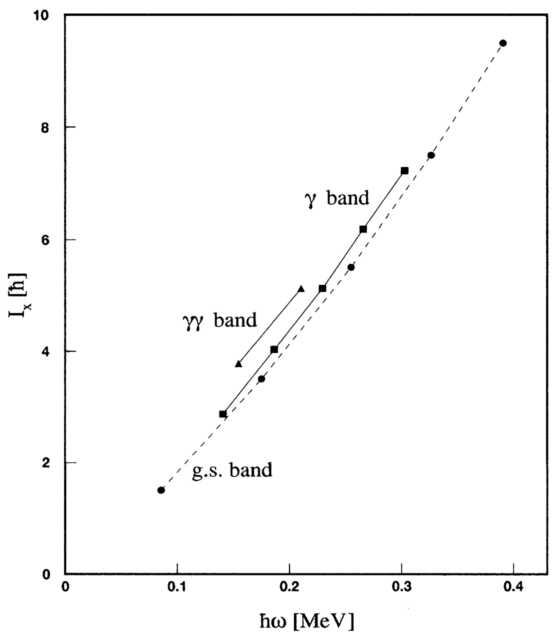

FIG. 2. Aligned angular momentum I_x as a function of the rotational frequency ω for the ground state and the γ and $\gamma\gamma$ bands. The slopes of the plots give the dynamical moments of inertia.

in spontaneous fission since they lie too far from the yrast line. In the case of anharmonic γ vibrations, the missing of a collective $K^\pi = 0^+$ two-phonon band has found a satisfactory explanation [1,3]. The search for these states in the harmonic situation would be of great interest.

Other band structures appear above 1.9 MeV, i.e., clearly above the proton pairing gap energy, in the partial level scheme presented in Fig. 1. For instance, the levels on the right-hand side of Fig. 1 most probably form a rotational band based on a two-quasiparticle state resulting from the breaking of a pair of identical nucleons. The ratios of energies of the intraband cascade transitions point to a $K = 4$ or 5 value, and the calculated moment of inertia of this band is, as expected, larger than those of the bands built on the ground state, and the one-phonon and two-phonon states.

In conclusion, a partial level scheme for the neutron-rich nucleus ^{106}Mo has been determined from the study of prompt, multifold-γ coincidences with the Eurogam2 array in the spontaneous fission of ^{248}Cm. It includes the yrast or ground-state band, a rotational band based on the one-phonon γ state, and also a state that presents the characteristics of a harmonic double-phonon $K^\pi = 4^+$ γ-vibrational state. The level scheme of ^{106}Mo thus yields the best example of a $K^\pi = 4^+$ harmonic two-phonon γ vibration yet observed in a deformed nucleus. Previous observations on transition strengths in the ^{232}Th nucleus [6], like the present data on ^{106}Mo, are also characteristic of a band with bandhead of two-phonon vibrational state nature. However, the proposed two-phonon state in ^{106}Mo is more *harmonic,* and it lies clearly *within* the pairing gap.

TABLE I. Predicted and experimental values for the ratio R defined by Eq. (4).

I_1	I_2	R	
		Pure harmonic picture	Experimental
6	4	1.22	1.29(26)
6	6	0.21	0.17(4)

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