

Structure of ^{240}Pu : Evidence for Octupole Phonon Condensation?

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The expanded level structure of ^{240}Pu available from the present study highlights the role of strong octupole correlations in this nucleus. In addition to a delayed alignment in the yrast band, the observations include the presence of both $I^+ \rightarrow (I-1)^-$ and $I^- \rightarrow (I-1)^+$ $E1$ transitions linking states of the yrast and negative-parity bands at high spin and the presence of an additional even-spin, positive-parity band deexciting exclusively to the negative-parity sequence. The observations appear to be consistent with expectations based on the recently proposed concept of octupole phonon condensation.

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Octupole correlations in nuclei result from the long-range, octupole-octupole interaction between nucleons occupying pairs of orbitals with $\Delta j = \Delta l = 3$. When both valence protons and neutrons occupy such states, the strength of the correlations can be such that rotational bands with alternating parity appear. These have commonly been interpreted in terms of the rotation of octupole-deformed nuclei [1,2]. The most striking examples of such behavior have been found in lanthanide ($A \sim 146$) and actinide ($A \sim 224$) nuclei. However, it has been realized for some time that this picture does not account for all of the experimental observations. Typically, the positive- and negative-parity states merge only at high spin. Moreover, a single rotational sequence rarely develops. Rather, the negative- and positive-parity sequences cross on close-lying trajectories in the energy-spin plane. An alternative interpretation for this behavior has recently been proposed in Ref. [3], where these states are understood as resulting from rotation-induced condensation of octupole phonons having their angular momentum aligned with the rotational axis. When the rotation of the condensate and the quadrupole shape of the nucleus synchronize, the collective motion becomes the familiar rotation of a static octupole shape. The small deviations mentioned above indicate that the synchronization is not fully reached. For less-well deformed nuclei, the resulting collective motion resembles that of a reflection-asymmetric tidal wave traveling on the surface of the nucleus [4].

In contrast to the situation depicted above, most negative-parity bands in the $A \sim 230$ – 250 actinide nuclei are described satisfactorily as rotational bands built on octupole vibrations. In this context, the 238 – ^{240}Pu isotopes

have remained somewhat of a puzzle. Specifically, Wiedenhöver *et al.* [5] reported that the sharp $i_{13/2}$ proton alignment observed in the yrast sequence of 241 – ^{244}Pu is absent within the same frequency range in the three lighter isotopes. This observation implies that, at the very least, there is a significant delay in the alignment process (octupole deformation has been shown to delay alignment processes—see Ref. [6]). Furthermore, at the highest spins, the yrast and the octupole states in $^{238,240}\text{Pu}$ appear to merge into a single band, although the interleaving $E1$ dipole transitions between the two sequences could not be observed. In ^{239}Pu , the positive- and negative-parity bands merge at high spin as well, and parity doublets seem to appear [7]. Additional indications for strong octupole correlations come from the fact that the decoupling parameters for the $K^\pi = 1/2^+$ and $K^\pi = 1/2^-$ bands in ^{239}Pu are of similar magnitude but opposite in sign [7], and, in the three isotopes, large dipole moments were inferred from the measured $B(E1)/B(E2)$ ratios in the medium-spin range where they are available [5,7]. Finally, it is worth noting that the large octupole strength in these specific Pu nuclei also manifests itself in the properties of their α decay [8].

This Letter focuses on ^{240}Pu , the even Pu isotope with the strongest octupole correlations. A new, high statistics measurement has expanded considerably the level structure of this nucleus and uncovered additional, unexpected spectral consequences of these correlations. All of the observations appear to find a satisfactory interpretation within the framework of octupole phonon condensation.

The experiment was carried out at the ATLAS accelerator facility at Argonne National Laboratory, with the Gammasphere array of 101 Compton-suppressed Ge spec-

trometers [9]. The approach is identical to that described in Ref. [5]. Thus, the so-called “unsafe” Coulomb excitation technique [10] was used at beam energies above the Coulomb barrier to enhance the feeding of the highest-spin states and the population of weak γ -ray cascades. A ^{208}Pb beam at an energy of 1300 MeV from ATLAS bombarded a ^{240}Pu target of ~ 0.35 mg/cm² thickness electroplated onto a 50-mg/cm² Au backing [11]. About 3×10^9 coincidence events were recorded when three or more suppressed Ge detectors fired in prompt coincidence. In the subsequent analysis, the data were sorted into three- and four-dimensional histograms using the RADWARE [12] and BLUE database [13] analysis packages. The latter made important contributions because of the ability to produce coincidence spectra at different detector angles which proved essential for the angular correlation measurements, as well as for the delineation of the highest-spin states where Doppler shifts and/or broadenings complicate the analysis. The resulting ^{240}Pu level scheme of Fig. 1 is

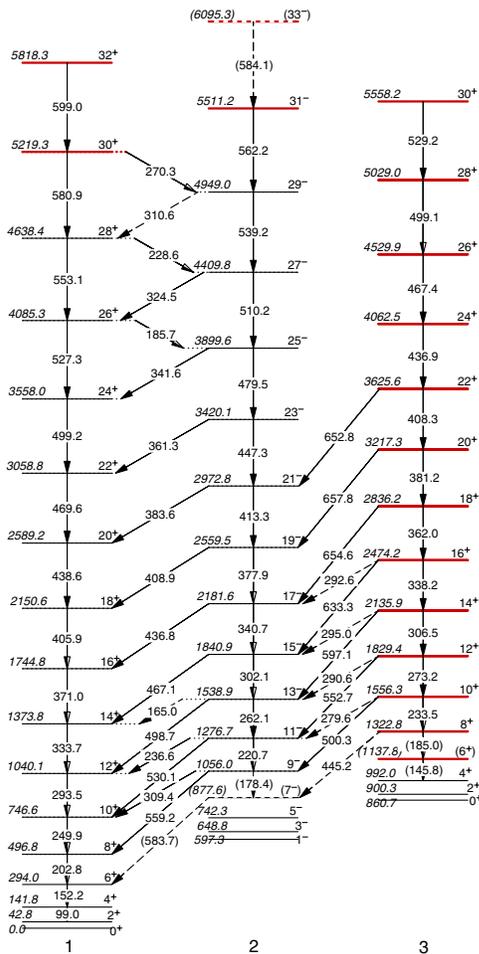


FIG. 1 (color online). Level scheme of ^{240}Pu with the new states observed in this work indicated by thick red lines; dashed lines and transitions energies in parentheses are used for γ rays either with a tentative placement or with no angular correlation information.

based on the observed coincidence relationships, as well as the measured angular correlations and γ -ray intensities. Further details of the data analysis and of the results will appear in a forthcoming publication which will also include data on $^{238,242}\text{Pu}$ [14]. They can also be found in Ref. [15].

With respect to Refs. [5,16–19], the highest-spin levels in the yrast and first negative-parity bands (bands 1 and 2 in Fig. 1) in ^{240}Pu have been delineated more accurately, hereby confirming the absence of any significant departure from a straightforward rotational behavior up to spins in excess of $30\hbar$ reported earlier [5]. Because of the high statistics accumulated, all of the levels of odd spin and negative parity up to 29^- have now been linked to the positive-parity yrast states by $E1$ transitions of the $I^- \rightarrow (I-1)^+$ type. More importantly, above spin $24\hbar$, where bands 1 and 2 appear to merge in a single sequence, three linking transitions of the type $I^+ \rightarrow (I-1)^-$, i.e., the 185.7-, 228.6-, and 270.3-keV dipole transitions have also been observed, establishing for the first time in a Pu isotope the interleaving pattern of $E1$ transitions commonly associated with octupole rotation. Further evidence for strong octupole correlations comes from the branching ratios between out-of-band and in-band transitions in band 2. For $I \leq 23\hbar$, these ratios were presented in Fig. 3 of Ref. [5]. They have now been extended throughout band 2, and their rise with angular momentum was found to persist to the highest spins (see Fig. 4.39 in Ref. [15]). With the usual assumption of a constant quadrupole moment $Q_0 = 11.6eb$ adopted from the measured $B(E2; 2^+ \rightarrow 0^+)$ transition rate [20], the induced intrinsic dipole moments D_0 are large ($D_0 \geq 0.2 e \text{ fm}$ for $I \geq 25\hbar$) and correspond to rates $B(E1) \geq 2 \times 10^{-3}$ W.u. Such values are larger than the $B(E1)$ strengths commonly observed for transitions linking negative-parity levels to positive-parity yrast states ($< 10^{-4}$ W.u.). They are, however, of the same order as those reported in the light Ra and Th isotopes [21], nuclei often viewed as some of the best examples of octupole rotors [2].

Built on three levels established previously in the decay of ^{240}Np [17], band 3 of Fig. 1 has been extended up to $I^\pi = 30^+$. Most surprisingly, from the 8^+ state upward, the deexcitation of this band proceeds solely towards the negative-parity levels of band 2 with $I^+ \rightarrow (I-1)^-$ dipole transitions of $E1$ character as well as with weaker $I^+ \rightarrow (I+1)^-$ γ rays [14,15]. The $E1$ multipolarity was established from the angular correlation analysis [14,15] showing that the linking transitions exhibit negative A_2 coefficients and small, positive A_4 coefficients (consistent with zero) of the same magnitude as those measured for the $E1$ transitions linking bands 2 and 1 [22]. No linking transitions between bands 1 and 3 were observed. For example, while the 14^+ band 3 member decays to the 13^- level of band 2 by a transition with an intensity of $\sim 0.7\%$ of the 249.9-keV yrast transition, the correspond-

ing upper limits for linking transitions to either the 12^+ or 14^+ band 1 states is $\leq 0.1\%$. This is a surprising and, to the best of our knowledge, unique observation in deformed nuclei throughout the periodic table. The $E1$ transitions linking bands 3 and 2 are similar in character to those linking bands 2 and 1: Within the same spin range, the $B(E1)/B(E2)$ branching ratios between out-of-band and in-band transitions for the former are of the same magnitude within errors as those observed for the latter (and the associated D_0 moments are large). Note that this result provides an explanation for the findings of Hoogduin *et al.* [23], who reported that, while in neighboring even nuclei strong $E0$ transitions link the first excited even-spin, positive-parity band to the yrast states, none was found for the band based on the 861-keV 0_2^+ level of ^{240}Pu . Rather, in this case, strong $E0$ deexcitation is associated with a rotational sequence built on the 1091-keV 0_3^+ state not observed in the present measurements.

In order to gain further insight into the nature of the excitations in ^{240}Pu , the evolution with rotational frequency $\hbar\omega$ of their alignments i_x and Routhians e' are displayed in Figs. 2 and 3, respectively. The striking absence of the strong $i_{13/2}$ proton alignment at $\hbar\omega \sim 0.25$ MeV seen in $^{242,244}\text{Pu}$ [5] is confirmed in the yrast sequence (band 1). The highest-frequency data may suggest that a first alignment is, in fact, delayed as expected in a nucleus with octupole deformation [6], but an extension of band 1 to higher spins is required before a firm conclusion can be reached. The alignment of band 2 grows gradually from a small value ($\sim 0.5\hbar$) at $\hbar\omega \sim 0.02$ MeV to a maximum of $\sim 3\hbar$ at $\hbar\omega \sim 0.20$ MeV, before remaining essentially constant up to $\hbar\omega \sim 0.28$ MeV. As a result, the value of Δi_x , the relative alignment of band 2 with respect to the yrast sequence (inset in Fig. 2) reaches $3\hbar$ in the medium-spin region, as expected for the alignment of

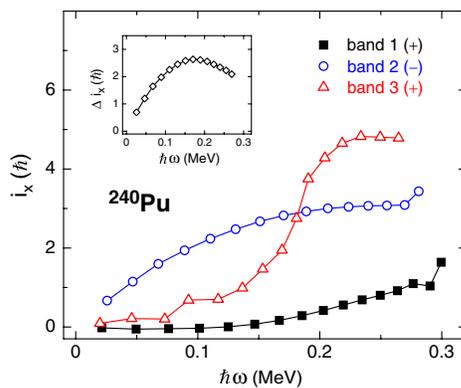


FIG. 2 (color online). Aligned spins i_x as a function of rotational frequency deduced for bands 1, 2, and 3 in ^{240}Pu . A collective reference with the Harris parameters $\mathcal{J}_0 = 71\hbar^2 \text{ MeV}^{-1}$ and $\mathcal{J}_1 = 320\hbar^4 \text{ MeV}^{-3}$ is adopted, which ensures that band 2 becomes flat for $\hbar\omega > 0.2$ MeV. The inset provides the relative alignment Δi_x of band 2 with respect to the yrast sequence.

an octupole phonon. Furthermore, a small decrease in Δi_x occurs for $\hbar\omega > 0.20$ MeV, a phenomenon also observed in nuclei such as ^{220}Ra and ^{222}Th [24,25] that are often considered to be among the best “octupole-deformed” rotors. Thus, the similarities between these rotors and bands 1 and 2 at high spin extend beyond the interleaving of the states in the two sequences accompanied by the characteristic pattern of $E1$ interband transitions to the more subtle behavior of relative alignments.

The aligned spin of band 3 (Fig. 2) starts from a low value ($< 1\hbar$), and a first, small irregularity in the evolution of i_x with $\hbar\omega$ occurs before 0.1 MeV, signaling the presence of a band crossing that can also be traced in the Routhian of Fig. 3. More importantly, at the point where a $3\hbar$ alignment is achieved in band 2, i.e., $\hbar\omega \sim 0.20$ MeV, a gain of $\sim 5\hbar$ has occurred in band 3, an i_x value that remains essentially constant at higher frequencies. This alignment gain translates into a slope change for the Routhian of band 3, and the ensuing trajectory would likely result in a crossing with the Routhians of bands 1 and 2 at frequencies beyond those reached in this work.

The recently proposed scenario of condensation of rotational-aligned octupole phonons [3] explains the observations in a natural way. Briefly, in this framework a prolate nucleus rotates with a frequency ω_2 , while multiphonon octupole tidal waves travel over its surface with a frequency ω_3 , which is $1/3$ of the vibrational frequency (see Fig. 4 in Ref. [3]). With increasing angular momentum, the one-phonon ($n = 1$) band crosses the zero-phonon ($n = 0$) band and becomes yrast. Then the two-phonon ($n = 2$) band becomes lowest in excitation energy following crossings with the $n = 0$ and $n = 1$ sequences. In the same way, the $n = 3$ phonon band comes down toward the yrast line as the angular momentum increases; i.e., boson condensation occurs; see Fig. 3(b) in Ref. [3]. In the ideal case of noninteracting harmonic phonons, these crossings occur at the same critical frequency $\omega_c = \omega_2 = \omega_3$, where the Routhians of multiphonon states coincide. At ω_c , the condensate corotating with the prolate nucleus resembles

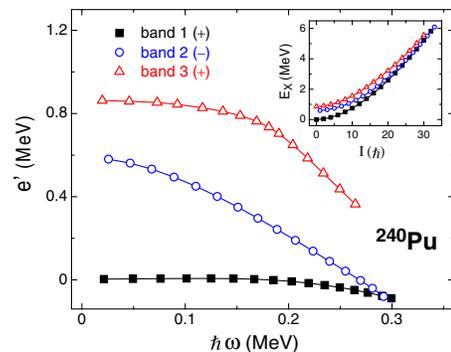


FIG. 3 (color online). Routhians (e') as a function of angular frequency ($\hbar\omega$) for states in bands 1, 2, and 3 of ^{240}Pu . The same reference as in Fig. 2 is used. Inset: The excitation energies (E_x) as a function of spin (I) for the same bands.

in a striking way an octupole rotor. In contrast to a static octupole rotor, ω_2 and ω_3 are not locked, and the lowest positive- and negative-parity bands oscillate around the ideal condensation line [Fig. 3(c) in Ref. [3]]. The anharmonicities of the octupole mode lead to an interaction between states with different phonon numbers but the same parity, resulting in a repulsion and mixing of the crossing bands.

Within this framework, bands 1, 2, and 3 in ^{240}Pu are associated at the highest rotational frequencies with $n = 0$, $n = 1$, and $n = 2$ phonons, and respective i_x values of approximately 0, 3, and $6\hbar$ result from the alignment of the octupole phonons with the rotation axis. The critical frequency is $\hbar\omega_c \approx 0.3$ MeV, where the $n = 0$ and $n = 1$ bands (bands 1 and 2) cross in Fig. 3. The Routhians of the $n = 0$ and $n = 2$ bands (bands 1 and 3) do not quite come together because they are associated with bands of the same parity. Because of the interaction, they repel each other. The corresponding state mixing gradually increases the i_x value of the $n = 0$ band by about $1\hbar$ and reduces i_x of the $n = 2$ band to about $5\hbar$. The similarity between the experimental Routhians e' and excitation energies E_x of the three bands (inset in Fig. 3) and the expectations presented in Figs. 3(b) and 3(c) of Ref. [3] is notable. Although the Routhian of the $n = 2$ band (band 3) could not be extended to the frequencies required to delineate the crossing and interaction with band 1, the data seem to indicate that such a crossing would likely occur above 0.3 MeV, pointing to the presence of anharmonicities. The interaction of the low-lying $n = 2$ band with the $n = 0$ band explains why the proton $i_{13/2}$ alignment is delayed (or absent). It is responsible for the down bend in the trajectory of the Routhian of band 1 at the highest frequencies and will delay a crossing with the s band (band with two aligned $i_{13/2}$ protons). More importantly, the $n = 2$ band will strongly interact with this s band and push it up in excitation energy, delaying its crossing with band 1.

The interpretation of band 3 as a two-phonon octupole structure above $\hbar\omega \sim 0.1$ MeV provides a natural explanation for the sole presence of $E1$ transitions between bands 3 and 2 as only transitions between states differing by a single phonon are allowed within such a vibrational picture. The mixing with the zero-phonon band at high spin accounts for the $E1$ transitions linking bands 1 and 2, with the presence of both $I^- \rightarrow (I-1)^+$ and $I^+ \rightarrow (I-1)^-$ γ rays of comparable strength [3,15]. As pointed out in Refs. [3,15], a substantial mixing of zero- and two-phonon bands does not differ much from a static octupole deformation.

In summary, the new data available for ^{240}Pu illustrate the impact of strong octupole correlations in ways that had not been seen before. A satisfactory interpretation of the observations appears to be achieved with the recently proposed concept of octupole condensation. To validate this interpretation further, additional experimental and

theoretical work is highly desirable. First, extending studies of ^{240}Pu to higher spins would provide missing information about the predicted crossing and interaction of bands 1 and 3. More detailed information on the electromagnetic transition rates will be important to substantiate this interpretation further. A search for the same phenomena in other nuclei, not only in the actinide region but also through other regions of the nuclear chart, is important in order to assess the generality of this exotic mode. Finally, a full microscopic description of this collective behavior is required as well.

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