PHYSICAL REVIEW C **81**, 041306(R) (2010)

Possible double-octupole phonon band in ^{238}U

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(Received 1 April 2010; published 28 April 2010)

The level scheme of ^{238}U has been extended using the so-called unsafe Coulomb excitation technique. One positive-parity band was uncovered for the first time, and its most important features can be related to a double-octupole phonon excitation. This band decays to the known $K = 0$ octupole band via $E1$ transitions, with strengths much larger than those to the ground-state band. It also decays to the $K = 1$ and 2 octupole bands. Comparisons among the proposed zero-, one-, and two-phonon bands in ^{238}U and those in ^{240}Pu shed more light on the recently proposed concept of rotationally aligned octupole phonon condensation.

DOI: 10.1103/PhysRevC.81.041306 PACS number(s): 21.10.Re, 23.20.Lv, 27.90.+b

Octupole correlations play an important role in determining the low-level structure of nuclei throughout the periodic table. Microscopically, these result from the long-range, octupole-octupole interaction between nucleons occupying pairs of orbitals with $\Delta j = \Delta l = 3$. The coupling of such pairs in opposite-parity orbitals located near the Fermi level can lead to reflection-asymmetric deformation. Octupole correlations are often coupled to other degrees of freedom. This is particularly the case in the actinide region [1–6], where signatures of the coupling of octupole correlations with the quadrupole deformation have been identified in two distinct collective modes; that is, octupole rotation and octupole vibration. A signature of the rotation of an octupole-deformed, even-even nucleus is the presence of rotational bands with levels of alternating parity, connected by strong electric-dipole transitions [i.e., $(I+1)^{\pm} \rightarrow I^{\mp}$] [2,3]. The large $B(E1)$ values of the connecting transitions are interpreted as resulting from the presence of an intrinsic electric dipole moment. In even-even nuclei, octupole vibrations have been associated with pairs of sequences of positive- and negative-parity levels well separated in energy. In addition to the $K^{\pi} = 0^-, 1^-, 2^-$, and 3^- one-phonon bands, bands with K^{π} varying from 0^+ up to 6^+ built on the double-octupole phonon should also exist. Forharmonic vibrations, these bands should decay to the one-phonon states only by fast $E1$ transitions, and the bandhead energy of the two-phonon $K^{\pi} = 0^+$ band should be twice that of the $K^{\pi} = 0^-$ bandhead. Finding the predicted double-octupole phonon excitations in the actinide region has proved elusive experimentally. However, a recent study of ^{240}Pu proposed evidence for such an excitation [7]. This nucleus exhibits properties somewhat in between those of an octupole vibrator and an octupole rotor. On one hand, it appears to behave as an octupole vibrator at low spin with the 1^- state located at 597 keV in energy. On the other, at high spin ($I \geqslant 23\hbar$), the yrast and the octupole bands appear to merge into a single sequence of levels with alternating spin and parity. More importantly, when the two bands become interleaved, three linking $E1$ transitions are observed to decay from the positive-parity band to the negative-parity sequence, and ^{240}Pu truly looks like an octupole-deformed rotor [6,7]. This apparent onset of octupole deformation has been attributed to the stabilization of the asymmetric shape by rotation [6]. The parity doublets observed at high spin in ^{239}Pu were also interpreted as the result of octupole deformation. In contrast, the structure of the isotone ^{237}U [8] can be classified as that of a typical octupole vibrator.

An important signature making ^{240}Pu unique concerns the band built on the first excited 0^+ state: it was found to decay solely via $E1$ transitions to the negative-parity band, and, in the $\hbar\omega > 0.20$ MeV region, exhibits an alignment of $\sim 5\hbar$ with respect to the yrast band [7]. Based on these observations, it was suggested that this band is associated with a double-octupole phonon excitation. Furthermore, the signatures laid out in Ref. [7] for these three bands are consistent with a description in terms of multiple-octupole phonon condensation, a concept first proposed by Frauendorf [9]. From a classical point of view, the condensation of rotationally aligned octupole phonons happens when the angular velocity of an octupole wave running over the surface of the quadrupole-deformed nucleus is equal to the angular velocity of this rotating prolate body. At this point, the quadrupole and octupole distortions combine into a heart-shaped object, and the classical image of a rotating, octupole-deformed nucleus is recovered [9].

In this paper, a candidate double-octupole phonon band decaying mainly to known one-octupole phonon bands is reported in ^{238}U, and the observations are compared with those in ^{240}Pu. Over the entire accessible frequency range ($\hbar\omega \leqslant 0.3$ MeV), no quasiparticle alignment is observed in ^{240}Pu. Hence, the impact of such a process on octupole phonon condensation has not yet been established. In contrast, the presence of a backbending in ^{238}U [4] provides an opportunity to investigate the interplay between octupole phonon condensation and the effects of particle alignment. In addition, the excitation energy of the proposed two-phonon band in ^{238}U is higher than that in ^{240}Pu, and possibly high enough to place it in the region where several excited quasiparticle states are expected. This opens up the possibility of investigating the impact on the band's properties of mixing with these quasiparticle excitations.

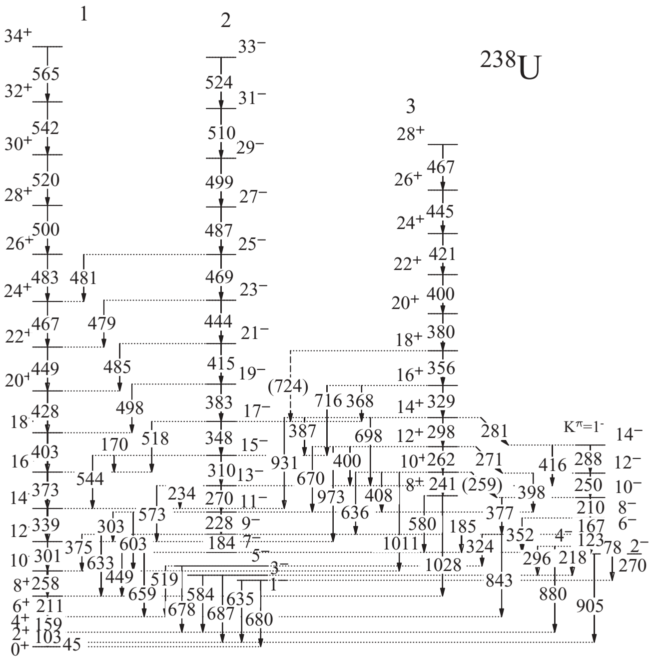

FIG. 1. Partial level scheme of ^{238}U. Band 3 is new to this work, as are the 542- and 565-keV transitions of band 1, the 524-keV γ ray in band 2, and the 185- and 416-keV deexcitations from the $K^\pi = 1^-$ band.

The low-lying structure of ^{238}U relevant to this work has been studied by Coulomb excitation with an 18-MeV α beam [10]. The low-spin members of the $K^\pi = 0^-$, 1^-, and 2^- octupole vibrational bands, together with two $K^\pi = 0^+$ states at 927 and 992 keV, were observed. Eight 2^+ states, located between 966 and 1782 keV, were also identified. Among these, only the 2^+ level at 1530 keV was suggested to possibly be of two-phonon octupole character, based on the large dipole moments inferred from the associated decay transitions [10]. It has been predicted that a 0^+ excited state decaying to the 0^- level with a large $B(E3)$ value should exist around 1.16 MeV in ^{236}U [11]. Even though this state has not yet been identified in this nucleus, it might still be possible to observe a similar level in ^{238}U.

It has been established by g-factor measurements on the high-spin yrast states [12] that the upbending of the ground-state band at $\hbar\omega \sim 0.25$ MeV in ^{238}U results from the rotational alignment of a pair of $i_{13/2}$ protons, with corroborating evidence from blocking arguments in the neighboring odd nuclei ^{237}Np [13,14] and ^{237}U [8]. The three octupole bands built on the respective principal quantum numbers $K^\pi = 0^-$, 1^-, and 2^- have been extended to high spin by the so-called unsafe Coulomb excitation technique with ^{209}Bi beams at energies of 1130 and 1330 MeV [4]. The alignment gain for these bands was attributed to the crossing of the octupole Routhians by negative-parity, two-quasiparticle states located near the Fermi surface [4]. Since a phonon excitation can be described by a linear combination of two-quasiparticle configurations in the framework of the random phase approximation (RPA), it was argued in Ref. [4] that the Coriolis field becomes strong enough at high spin to break the octupole phonon, at which point the individual aligned two quasiparticles are released from the octupole coupling.

In the present work, high-spin states in ^{238}U were again populated with the unsafe Coulomb excitation technique pioneered in Ref. [4]. The isotopically enriched ^{238}U target ($\geqslant$99%, 48 mg/cm^2 thick) was bombarded with a 1400-MeV ^{207}Pb beam, delivered by the ATLAS superconducting linear accelerator. Gamma rays were detected with the Gammasphere [15] array composed of 101 Compton-suppressed Ge detectors. The detection efficiency for γ-ray energies below 200 keV was improved by operating the spectrometer with the timing discriminators in leading edge rather than in constant fraction mode.

In excess of 10^9 three- or higher-fold coincidence events were accumulated and sorted into three-dimensional and four-dimensional histograms using the RADWARE analysis software [16]. These histograms contained only γ rays emitted within a ±15-ns prompt time window with respect to the beam. The double- and triple-gated coincidence spectra were extracted using the generalized background subtraction algorithm of Ref. [17] and provided the basis for the construction of the level scheme. Information on the multipolarity of the transitions with sufficient yields was obtained from an angular distribution analysis. For weaker γ rays, the anisotropy ratio R, defined as the intensity ratio of transitions observed in the 32°, 37°, 143°, 148°, and 163° detector rings to those detected in the 79°, 81°, 90°, 99°, and 101° rings, was used to deduce the multipolarity. In general, stretched-quadrupole transitions are associated with $R > 1$ values, while pure dipole transitions are characterized by $R < 1$. Because only prompt γ rays are considered, each transition was restricted to either $E1$, $E2$, or $M1$ character or to the appropriate mixing of these multipolarities.

A partial level scheme for ^{238}U is given in Fig. 1. The primary feature of interest is the newly discovered band 3. It is weakly populated, with an intensity of only 0.06(1)% relative to that of the ground-state band (GSB). In contrast, the intensity of band 2 is about 1.10(1)% of the GSB. Figure 2 provides a summation of double-gated coincidence spectra for this band

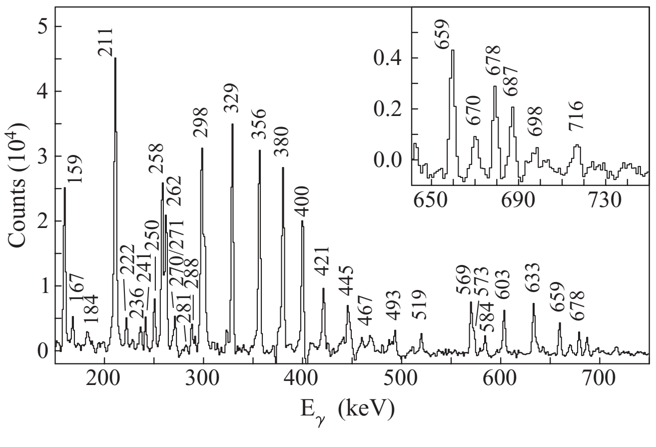

FIG. 2. Sum of coincidence spectra double gated on all the transitions in band 3 except the 241- and 467-keV γ rays. The inset is the expansionof the spectrum between 640 and 750 keV.

to illustrate the quality of the data. The inset is an expanded part of the spectrum indicating the transitions connecting bands 2 and 3. The spins and parities of bands 1 and 2 have been assigned in previous works and were also evaluated in Ref. [18]. These agree with the results from the aforementioned angular distribution analysis. The stretched-$E2$ character of all γ rays in band 3 was derived from the measured angular distributions and anisotropy ratios. The dipole character of the 670-keV transition linking bands 3 and 2 is consistent with the measured coefficients $A_2/A_0 = -0.13(2)$ and $A_4/A_0 = 0.04(4)$. In addition, the anisotropy ratios for the 636- and 698-keV linking transitions are 0.89(6) and 0.84(5), respectively, establishing their dipole character as well. These observations, when combined with the decay pattern to band 2, result in the spin values for band 3 proposed in Fig. 1. The parity of this band cannot be determined in a trivial manner based solely on the dipole character of these linking transitions. The decay out of the $I = 12$ level of band 3 to states in the $K^\pi = 1^-$ band occurs only through a 271-keV γ ray without a branch to the 10^- state (Fig. 1). With a negative-parity assignment for band 3, a 521-keV $E2$ transition would be expected as a link between the level of interest and the 10^- state, and would compete favorably with the 271-keV γ ray. Furthermore, it would be stronger than the 670-keV γ ray, which would be of $M1$-$E2$ character under the negative-parity assumption. In contrast, such a 521-keV transition would be absent for a positive-parity assignment as an $M2$ transition is unlikely to compete with an $E1$ link, and these arguments settle the positive parity of band 3.[1]

One of the expectations for a double-octupole phonon band is that its 0^+ bandhead be located at twice the energy of the one-phonon 1^- state. Despite an extensive search for in-band transitions at the bottom of band 3 in the hope of reaching the presumed 0^+ bandhead, no γ rays deexciting states with spin below 6^+ could be found. A reason for this lack of signal is that the in-band transitions compete with the deexcitations out of the band of higher energy that take away the dominant portion of the decay strength. This is demonstrated by the sudden drop in intensity of the 241-keV peak when compared to that of the 262-keV transition (Fig. 2). In addition, the lower-energy transitions tend to decay through the electron conversion process, making the identification of lower-spin band members even more challenging. However, the energies of these unobserved states can be extrapolated from the known levels. The 0^+ bandhead energy is estimated to be 1162 keV by adopting a simple relation between level energies and spins, $E(I) = E(0) + aI(I+1) + bI^2(I+1)^2$, which is applicable as long as alignment processes are still small in band 3. This value is about 200 keV higher than the energies of the two known excited 0^+ states [18], but about 200 keV lower than twice the excitation energy of the 1^-_1 state. With this extrapolated energy, the energy ratio $E(0^+)/E(1^-) = 1.71$ is surprisingly close to the value predicted for ^{236}U of 1.71 [11]. In addition, an extrapolated energy for the 2^+ state is about 1197 keV, which is not among the eight 2^+ states between 966 and 1782 keV observed in the Coulomb excitation study, and is much lower than the suggested two-octupole phonon 2^+ state at 1530 keV [10]. Although these estimated values require experimental confirmation, they suggest that anharmonicity plays an important role in explaining the properties of band 3, as was also the case in ^{240}Pu [7].

Band 3 predominantly feeds into the three known $K^\pi = 0^-$ (band 2), 1^-, and 2^- (not shown in Fig. 1; see Ref. [4]) octupole bands with a weaker branch to the GSB. The data allow an extraction of the decay-out branching ratios only in a limited number of cases because of the presence of numerous doublets and other contaminant transitions in the spectra. Nevertheless, the relative intensities of the deexciting γ rays from the $I^\pi = 12^+_3$ state toward the various bands are measured as $I(670{:}12^+_3 \to 11^-_1)/I(262{:}12^+_3 \to 10^+_3) = 15(1)\%$, $I(271{:}12^+_3 \to 12^-_1)/I(262{:}12^+_3 \to 10^+_3) = 13(1)\%$, and $I(973{:}12^+_3 \to 12^+_1)/I(262{:}12^+_3 \to 10^+_3) = 4.4(5)\%$. The γ rays associated with the decay to the $K^\pi = 2^-$ band [4] cannot be observed because Gammasphere is not sensitive to these low-energy transitions, but the coincidence evidence indicates that the decay strength to this band is sizable, of the order of 5%. Such a deexcitation pattern is consistent with the expected scenario for a double-octupole phonon band where the decay proceeds mainly toward the one-octupole phonon states. The measurable branch to the GSB, which was not observed in ^{240}Pu [7], is a first indication of a sizable mixture of the state wave functions in band 3, and might be associated with the anharmonicity as well.

Because the level structure of ^{238}U is considered to be octupole vibrational in character, its intrinsic dipole moment (D_0) at equilibrium is equal to zero, while its quadrupole moment (Q_0) is well defined. As a result, the ratio of $B(E1; I^\pm \to (I-1)^\mp)/B(E2; I^\pm \to (I-2)^\pm)$ is no longer a good measure of octupole deformation, but rather an indication of the strength of octupole correlations relative to the quadrupole collectivity. From the available branching ratios, the $B(E1; I^+_3 \to I^-_2 - 1)/B(E2; I^+_3 \to I^+_3 - 2)$ values are $1.8(2) \times 10^{-9}$ and $0.5(1) \times 10^{-9}$ fm^{-2} for the 10^+_3 and 12^+_3 states of band 3, respectively. Even after the branch to the $K^\pi = 1^-$ band is accounted for, these are still much smaller than the values extracted for the 11^-_2 [$1.4(1) \times 10^{-8}$ fm^{-2}] and 13^-_2 [$1.2(1) \times 10^{-8}$ fm^{-2}] states of band 2. This result is distinctly different from that in ^{240}Pu [7], where the $B(E1)/B(E2)$ ratios are of the same magnitude for bands 2 and 3 within the same spin range [7]. This observation can perhaps be viewed as an indication that band 3 of ^{238}U is the result of mixing between the double-octupole phonon and quasiparticle states. This mixing can be related to the high excitation energy of band 3. The single-particle Routhians (e') for all the bands are plotted in Fig. 3 as functions of the rotational frequency $\hbar\omega$ using the Harris parameters $J_0 = 65\,\text{MeV}^{-1}\hbar^2$ and $J_1 = 365\,\text{MeV}^{-3}\hbar^4$ [19]. When compared with RPA calculations (Fig. 12 in Ref. [4]), the Routhian of band 3 in ^{238}U is located in the region where two quasineutron

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[1] Note that, because the energy of another possible interband transition connecting the $I = 14^+$ and $I^\pi = 12^-$ levels is equal to the energy of the 569-keV transition in ^{207}Pb, the observation of this transition in the double-gated spectrum of 329- and 250-keV γ rays is the result of the cross correlation between ^{238}U and ^{207}Pb, and does not contradict the positive-parity assignment for band 3.

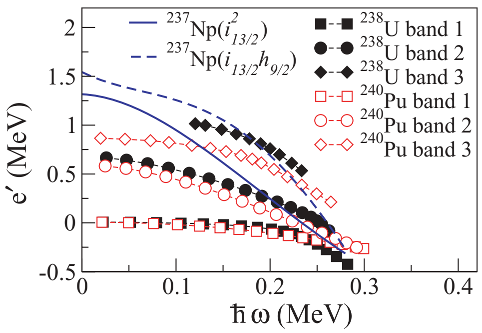

FIG. 3. (Color online) Routhians for bands 1, 2, and 3 of ^{238}U and ^{240}Pu. The constructed Routhians for $\pi i_{13/2}^2$and $\pi i_{13/2}h_{9/2}$ configurations based on the experimental data of ^{237}Np [14] are plotted as lines.

and two quasiproton states can be found. As a consequence, the probability of mixing between quasiparticle and phonon states is enhanced. Since, at any given frequency, the Routhian of band 3 is lower in ^{240}Pu than in ^{238}U, the double-octupole phonon band in the former nucleus likely avoids such mixed character.

To further understand the nature of this newly observed band in ^{238}U, the alignment i_x, as a function of the rotational frequency $\hbar\omega$, for bands 1–3 in both ^{238}U and ^{240}Pu is presented in Fig. 4(a), using the Harris parameters given above. It can be seen that the alignments of the bands in ^{238}U and ^{240}Pu follow the same pattern for $\hbar\omega \leqslant 0.20$ MeV, pointing to a similar underlying intrinsic structure in this frequency range. The alignments deviate from each other at larger $\hbar\omega$, where an upbend is observed in ^{238}U. As discussed earlier, the upbend in the GSB is a consequence of a crossing with a band associated with a rotationally aligned pair of $i_{13/2}$ quasiprotons. It has also been suggested that the backbending of the octupole phonon band results from the crossing with a negative-parity two-quasiparticle band [4]. In Fig. 3, Routhians associated with the $\pi i_{13/2}^2$ and $\pi i_{13/2}h_{9/2}$ configurations were constructed from the available data in the neighboring nucleus ^{237}Np [14]. Even though the constructed values for the $\pi i_{13/2}h_{9/2}$ states are distorted because of the unblocked $\pi i_{13/2}^2$ alignment in the $h_{9/2}$ band of ^{237}Np [14], the Routhian trajectories suggest that the one-phonon band (band 2) in ^{238}U is crossed by this negative-parity two-quasiparticle band. At the same time, it can be seen that the Routhian of the two-phonon band in ^{238}U is located so high in energy that the crossing with the $\pi i_{13/2}^2$ band is avoided. Ideally, the alignment of an aligned double-octupole phonon is $6\hbar$. In Ref. [7], it is suggested that in ^{240}Pu the maximum alignment of the two-phonon band is reduced somewhat to $\sim 5\hbar$ because of the admixture between zero- and two-phonon states at high spin. The situation of band 3 in ^{238}U appears to be more subtle than that in ^{240}Pu. The alignment reaches $6\hbar$ around $\hbar\omega \sim 0.21$ MeV, consistent with the value for an aligned double-phonon octupole band. Beyond $\hbar\omega \sim 0.23$ MeV, i_x continues to increase, presumably as a consequence of mixing with two-quasiparticle states other than the first pair of $i_{13/2}$ quasiprotons. As illustrated in Fig. 4(b), the alignment difference between the one-phonon and GS bands (Δi_x : band 2 − band 1) for ^{238}U is almost identical to that for ^{240}Pu before the impact of the aligned orbitals becomes visible. In addition, for $\hbar\omega \leqslant 0.21$ MeV, the difference between the two-phonon and GS bands (Δi_x : 3 − 1) in ^{238}U is about $1\hbar$ higher than that in ^{240}Pu. Presumably this additional $1\hbar$ alignment also comes from the quasiparticle states admixed into this band.

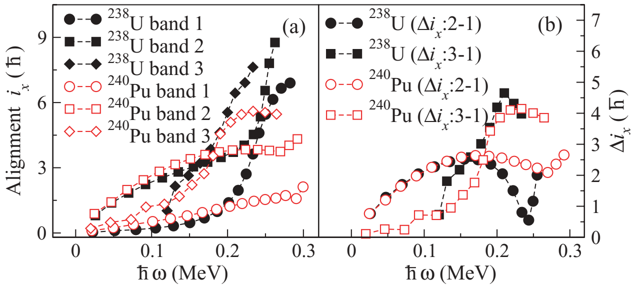

FIG. 4. (Color online) (a) Alignments of bands 1, 2, and 3 of ^{238}U and ^{240}Pu. (b) Alignment differences between bands 2 and 1, and bands 3 and 1 in both nuclei.

The recently proposed scenario of rotationally aligned octupole phonon condensation can be applied to associate bands 1, 2, and 3 in ^{240}Pu with $n = 0$, 1, and 2 octupole phonons, respectively. Even though it has been demonstrated previously that the situation in ^{238}U is more complex owing, at least in part, to band crossings at high spin, as well as to band mixing, the trajectories of the Routhians for the three bands follow a remarkably similar pattern to those in ^{240}Pu before the backbending (Fig. 3). When compared to the Routhians shown in Figs. 3(b) and 3(c) of Ref. [9], it appears that the convergence of the three bands in ^{238}U is interrupted by the crossing with the aligned two-quasiparticle states. Nevertheless, when the Routhian trajectories are extrapolated while the points beyond the band crossing are neglected, it is clear that condensation would occur at a frequency higher than 0.28 MeV, the value where the bands in ^{240}Pu would cross each other. The higher projected condensation frequency in ^{238}U is consistent with what has been described in Ref. [9], where the condensation frequency was determined as Ω_3/i for a harmonic octupole vibration with frequency Ω_3. Ideally, each phonon carries $i = 3\hbar$ of angular momentum aligned with the rotational axis. Assuming that the rotational energy does not change with n, the excitation energy of the n-phonon states in the rotating frame (relative Routhian) is $E_n' = \hbar\Omega_3(n + 1/2) - ni\omega$, and the condensation frequency, where all $E_n'(\omega_c)$ coincide, is $\omega_c = \Omega_3/3$. From the differences between the zero- and one-phonon Routhians in Fig. 3 at $\hbar\omega = 0.20$ MeV, one finds for ^{240}Pu $\Omega_3 = 0.84$ MeV corresponding to $\omega_c = 0.28$ MeV and $\Omega_3 = 0.90$ MeV corresponding to $\omega_c = 0.30$ MeV for ^{238}U, which indicates the delay of the octupole condensation in the latter. From the differences between the zero- and two-phonon Routhians in Fig. 3 at $\hbar\omega = 0.20$ MeV, one finds $\Omega_3 = 0.84$ MeV for ^{240}Pu and $\Omega_3 = 1.01$ MeV for ^{238}U. The fact that the two-phonon band lies at an energy higher than twice the one-phonon energy reflects the anharmonicity of the octupole mode. As discussed in Ref. [9], this type of anharmonicity stretches the condensation point into a narrow frequency interval.

According to the description of Ref. [9], condensation sets in only when the angular velocity for both the octupole and quadrupole components reaches the critical value. For ^{240}Pu, this condensation was realized at $\hbar\omega \sim 0.28$ MeV because of strong octupole correlations. These correlations are also thought to be responsible for the delay in the two-quasiparticle alignments and the critical angular velocity is reached without interruption as a result. The situation is different in ^{238}U. The crossing of the aligned two-quasiparticle states happens at a lower frequency (~0.24 MeV), substantially below the estimated condensation frequency of 0.30 MeV, and leads to a transition from a collective octupole state to an aligned quasiparticle one before condensation sets in. Figure 3 clearly indicates that the ^{238}U Routhians for bands 1 and 2 stop converging toward the same frequency after band crossings take place. In other words, the process of octupole phonon condensation is not completed due to mixing with the aligned two-quasiparticle states. The two isotones illustrate the delicate balance between octupole condensation and $i_{13/2}$ proton alignment: in ^{238}U the rotationally aligned band crosses the GSB before the two-phonon octupole band is close enough to strongly mix with it, which terminates condensation. In ^{240}Pu, the two-phonon band mixes with the GSB earlier. This stronger mixing lowers the energy of the yrast levels and generates strong octupole correlations, both preventing (or delaying) the crossing with the aligned-proton band, which is seen in ^{242,244}Pu at 0.25 MeV, slightly higher than in ^{238}U. It is interesting that the relatively small differences in the condensation and alignment frequencies have such a dramatic consequence.

As mentioned above, even though the Routhian of band 3 in ^{238}U is not low enough in energy to avoid mixing with other quasiparticle states, it still exhibits features characteristic of a two-phonon octupole structure. First, the strongest decay-out branch is to the one-phonon band via $E1$ transitions. Second, together with the other two bands, it behaves in a way similar to that of their counterparts in ^{240}Pu in terms of alignments and Routhians, at least in the low-frequency region free of mixing and alignments. The difference between its observed properties and the expectations as a pure two-phonon octupole band is attributed to configuration mixing. The observable branch to the ground-state band is one of the results of this mixing. Another is that the $B(E1)/B(E2)$ ratios are smaller for band 3 than for band 2 in the same spin range. Finally, the additional $1\hbar$ alignment, as compared to that of band 3 in ^{240}Pu, is very likely caused by the mixed quasiparticle states as well.

This work is supported in part by the US Department of Energy, Office of Nuclear Physics, under Contract No. DE-AC02-06CH11357.

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