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Strength of octupole correlations in the actinides: contrasting behavior in the isotones ^{237}U and ^{239}Pu

S. Zhu [a], R.V.F. Janssens [a], G.J. Lane [b,c], I. Wiedenhöver [d], M.P. Carpenter [a], I. Ahmad [a], A.P. Byrne [b], P. Chowdhury [a,e], D. Cline [f], A.N. Deacon [g], G.D. Dracoulis [b], S.J. Freeman [a,g], N.J. Hammond [a], G.D. Jones [h], T.L. Khoo [a], F.G. Kondev [a], T. Lauritsen [a], C.J. Lister [a], A.O. Macchiavelli [c], E.F. Moore [a], D. Seweryniak [a], J.F. Smith [g], C.Y. Wu [f]

[a] *Argonne National Laboratory, Argonne, IL 60439, USA*
[b] *Australian National University, Canberra ACT 0200, Australia*
[c] *Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA*
[d] *Florida State University, Tallahassee, FL 32306, USA*
[e] *University of Massachusetts Lowell, Lowell, MA 01854, USA*
[f] *University of Rochester, Rochester, NY 14627, USA*
[g] *University of Manchester, Manchester M13 9PL, United Kingdom*
[h] *University of Liverpool, Liverpool L69 7ZE, United Kingdom*

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Abstract

A study of high spin states in the odd-neutron isotones ^{239}Pu and ^{237}U is reported. Striking differences were found in the high-spin properties of rotational bands built on the $1/2^+[631]$ ground states in these two nuclei. These differences mirror those observed in the even–even Pu and U immediate neighbors and appear to be related to the strength of octupole correlations.

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Octupole correlations play an important role in determining the low level structure of nuclei throughout the periodic table. This is particularly the case in the actinide region [1–7], where two distinct collective modes have been identified. An octupole vibration has been associated with sequences of negative parity levels starting at excitation energies of 0.5 MeV or more with respect to the ground state. These are understood in terms of rotational bands built on octupole phonon

E-mail address: janssens@anl.gov (R.V.F. Janssens).

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excitations. Many of the band members with quantum numbers I^- decay through characteristic E1 transitions towards both the $(I+1)^+$ and $(I-1)^+$ yrast states. The case of ^{238}U is a good example: three octupole bands with respective principal quantum numbers $K = 0$, 1 and 2 have been identified by Ward et al. [5]. In nuclei where these correlations are much stronger, these negative parity states lie even lower in excitation energy, and stable octupole deformation may occur. In nuclei around ^{146}Ba and ^{224}Th, bands with levels of alternating spin and parity, connected by strong electric-dipole transitions (i.e., $(I+1)^+ \to I^-$ and $I^- \to (I-1)^+$), are recognized as the best experimental evidence for the rotation of octupole-deformed nuclei. It has been shown empirically that rotation can stabilize the octupole shape [3,4,6]. Furthermore, in odd-mass nuclei, the most striking evidence for octupole deformation is provided by the presence of so-called parity doublets, i.e., pairs of states nearly degenerate in excitation energy with the same spin, but opposite parity [8].

The energy displacement between the positive and negative parity states in such collective sequences can be used as an empirical measure of the strength of the octupole correlations. This displacement has been investigated by, among others, Jolos and von Brentano [9,10] who showed that it depends strongly on angular momentum. These authors also suggested that nuclei exhibiting dynamical octupole effects at low spins might develop static octupole deformation at sufficiently high angular momentum. Thus far, experimental evidence for such an evolution in octupole character has only been proposed in one instance. Wiedenhöver et al. [7] reported that the sharp $i_{13/2}$ proton alignment observed in ^{242,243,244}Pu is not present within the same frequency range in the lighter ^{238,239,240}Pu nuclei, implying at the minimum a significant delay in the alignment process (octupole deformation has been shown to delay alignment processes, see Ref. [11]). Furthermore, at the highest spins, the yrast and the octupole bands in ^{238,240}Pu appeared to merge into a single sequence of levels with alternating spin and parity, and large intrinsic dipole moments were inferred from the measured $B(\mathrm{E1})/B(\mathrm{E2})$ ratios. In addition, parity-doublets appeared to occur in ^{239}Pu [7]. Thus, the experimental evidence suggested that a transition from an octupole vibration to stable octupole deformation may have occurred. It is worth noting that the large octupole strength in these specific Pu nuclei also manifests itself in the properties of their ground state α decay [12].

It is the purpose of the present Letter to explore the strength of the octupole correlations in this region further by comparing the behavior of the odd ^{239}Pu nucleus with that of its isotone ^{237}U. Odd-A nuclei offer the potential for new insight at the cost of added complexity with respect to even–even nuclei since the coupling of the odd particle to the core is likely to be affected by the presence of octupole correlations. For example, microscopic investigations of odd-mass nuclei with the inclusion of octupole deformation suggest a dependence of the correlation strength on the principal quantum number Ω of the single-particle orbital occupied by the odd nucleon [8,13]. Partial, preliminary results on ^{239}Pu and ^{237}U have been presented earlier [7,14]. In these two nuclei it has now been possible to follow octupole excitations up to high spin and notable differences between the behavior in the two isotones have been found. The findings shed new light on the issues raised above, in particular, on the possible evolution with rotational frequency from an octupole vibration to octupole deformation.

High-spin states in ^{239}Pu were populated with the so-called “unsafe” Coulomb excitation technique pioneered in Ref. [5], while ^{237}U was produced via a one-neutron pickup reaction on a ^{238}U target. The 98% isotopically enriched ^{239}Pu target ($\sim 0.3\ \mathrm{mg/cm^2}$ thick), electroplated on a $50\ \mathrm{mg/cm^2}$ Au foil [15], was bombarded with a 1300-MeV ^{207}Pb beam. For the investigation of ^{237}U, the same ^{207}Pb beam was used, but at an energy of 1400 MeV. The ^{238}U target, isotopically enriched to $\geqslant 99\%$, was $48\ \mathrm{mg/cm^2}$ thick. The ^{207}Pb beams were delivered by the ATLAS superconducting linear accelerator at Argonne National Laboratory. Gamma rays were detected with the Gammasphere [16] array of 101 Compton-suppressed HPGe detectors. For the ^{237}U measurement, the detection efficiency for low-energy photons was greatly improved by taking advantage of the recently installed capability to operate the timing discriminators in leading edge rather than in constant fraction mode for γ-ray energies below 200 keV. This capability was not available at the time of the ^{239}Pu measurement and, in the latter case, the discriminators were operated in the customary constant fraction mode.

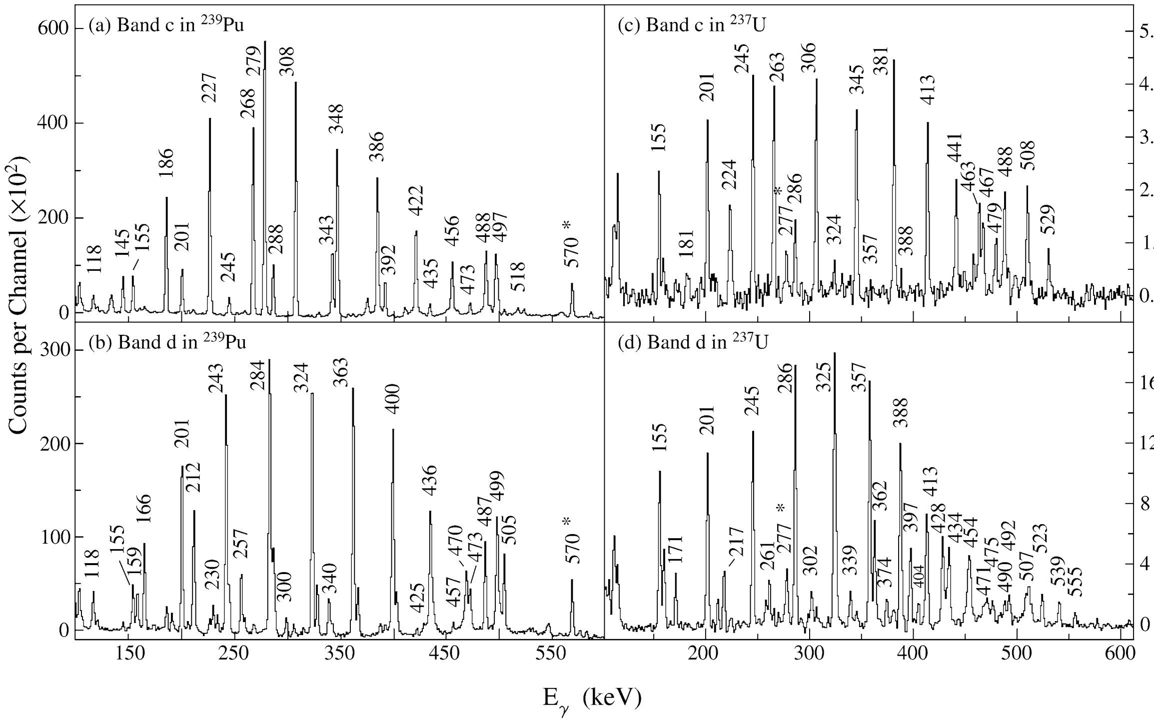

Fig. 1. Summed double-gated spectra for octupole partner bands *c* (figure (a)) and *d* (figure (b)) in ^{239}Pu and summed triple-gated spectra for octupole partner bands *c* (figure (c)) and *d* (figure (d)) in ^{237}U. Most of the connecting E1 transitions are clearly seen in the figures. Cross-coincident transitions from projectile-like binary reaction partners are marked by an asterisk.

In excess of 10^9 three or higher fold coincidence events were accumulated with each target and stored onto magnetic tapes for further analysis. In the case of the ^{239}Pu target, the contribution of the Au backing to the γ-ray yield is substantial. The data were sorted into three-dimensional (E_γ–E_γ–E_γ, cube) and four-dimensional (E_γ–E_γ–E_γ–E_γ, hypercube) histograms using the Radware analysis software [17]. These histograms contained only γ rays emitted within a ± 20 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. [18] and provided the basis for the construction of the level schemes presented below. Information on the multipolarity of the transitions was obtained from an angular correlation analysis. To this effect, a number of coincidence histograms corresponding to specific angle combinations between the Gammasphere detectors were obtained and analyzed, as described in Ref. [19].

The level structure of ^{239}Pu known prior to this work has recently been summarized in Ref. [20]. For the purpose of the present analysis, however, the results of studies using inelastic excitation with a ^{117}Sn beam [21] and fusion–evaporation with the ^{238}U$(\alpha, 3n)$ reaction [22] are most relevant. The quality of the present data is illustrated in panels (a) and (b) of Fig. 1, and the relevant level scheme is given in Fig. 2. The yrast structure, composed of the two signature partner bands *a* and *b* assigned to the $1/2^+[631]$ configuration of predominant $d_{5/2}$ parentage, has now been extended to levels with assigned quantum numbers $(55/2^+)$ and $(53/2^+)$. The angular correlation analysis supports the stretched-E2 character of all in-band transitions, with the exception of the highest level in each sequence where statistics on the correlation coefficients proved insufficient. In addition, eight new inter-band transitions linking bands *a* and *b* have also been identified. More importantly in the present context, the octupole excitations built on the *a* and *b* signature partners, tentatively proposed in Ref. [21],

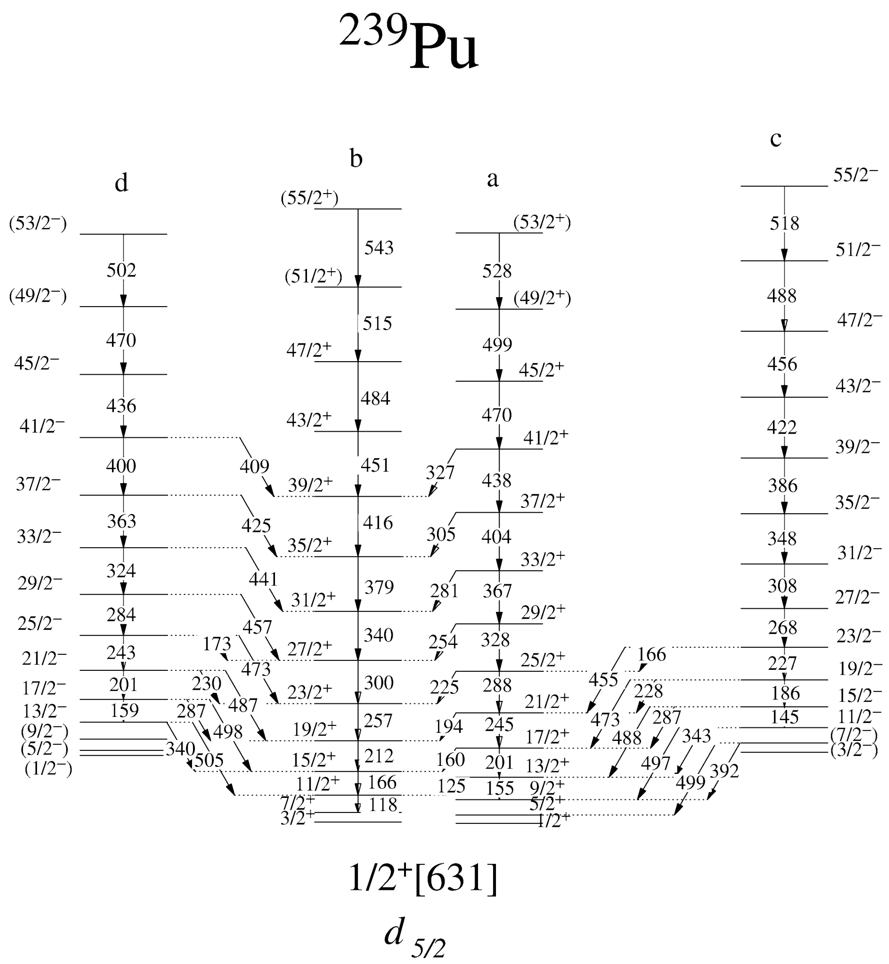

Fig. 2. Proposed ^{239}Pu level scheme with the transition energies given in keV. Bands c and d are the octupole bands under discussion in this Letter.

have now been firmly established and expanded considerably towards higher frequencies (bands c and d in Fig. 2). Moreover, they have been firmly linked to the lower octupole states established earlier through decay studies [20] and the E1 linking transitions have been observed. Sums of coincidence spectra double-gated on in-band transitions in the c and d sequences are shown in panels (a) and (b) of Fig. 1: strong interband transitions depopulating states in band d up to $I^\pi = 41/2^-$ towards band b, and levels up to $23/2^-$ in band c towards band a are clearly visible. In contrast with the case of ^{237}U discussed below, the search for rotational bands built on other known configurations in ^{239}Pu [20], especially those associated with the $\nu j_{15/2}$ intruder orbital, proved unfruitful. This is presumably due to the more limited statistics available in this case as the thickness of the available targets was limited by health safety considerations.

The information on the ^{237}U level structure available prior to the present work results mainly from decay and light-ion transfer measurements. It is summarized in the compilation of Ref. [23], and serves as a starting point for the present study. Representative spectra for ^{237}U are also shown in Fig. 1. Because of the reaction used, coincidence spectra for ^{237}U contain transitions in the binary reaction partner ^{208}Pb. Panel (c) in Fig. 1 was produced from a hypercube by summing coincidence spectra requiring the 583-keV transition in ^{208}Pb together with a number of double gates placed on transitions in a newly discovered,

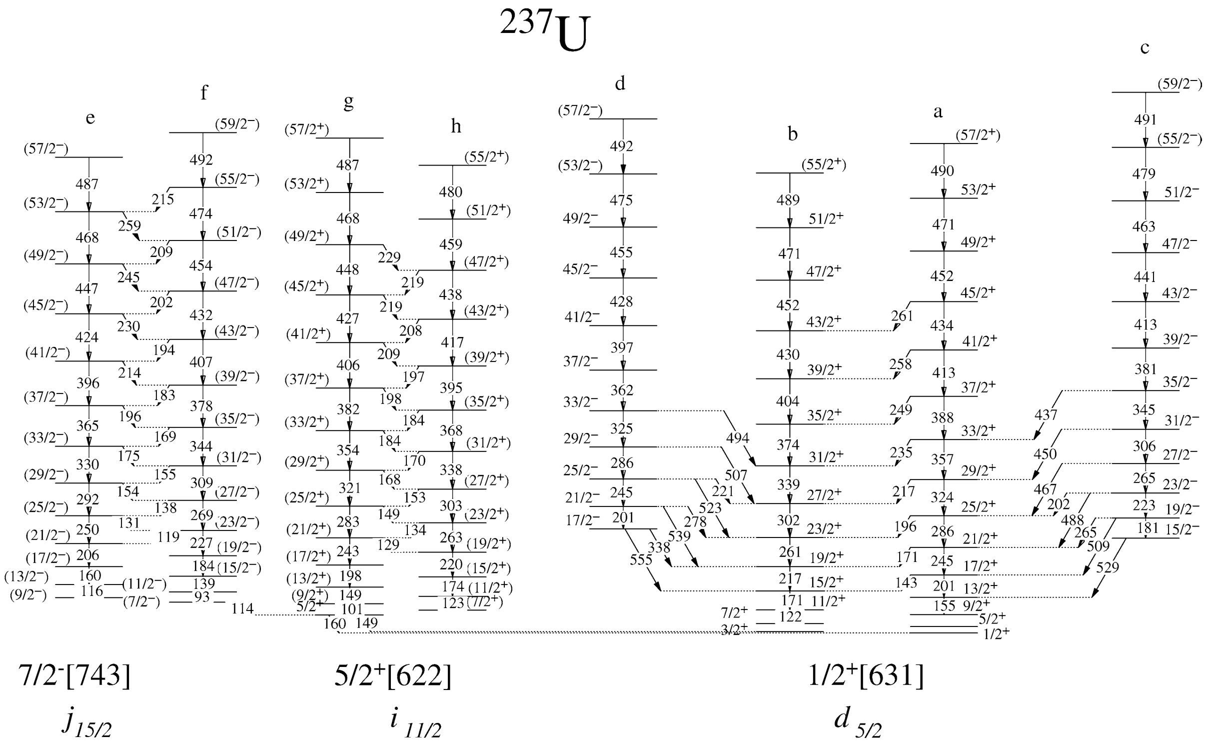

Fig. 3. Proposed level scheme for ^{237}U. The energies of the transitions are given in keV. Bands *c* and *d* are the octupole bands under discussion in the present Letter.

negative-parity band of ^{237}U labeled as band *c* in the level scheme of Fig. 3. Panel (d) was produced in the same manner, but for another negative-parity sequence labeled as band *d* in ^{237}U.[1]

As can be seen in Fig. 3, a total of eight rotational bands, labeled *a* through *h*, are assigned to ^{237}U. All of them were found to be in strong cross-coincidence with the first two ^{208}Pb yrast transitions at 2615 and 583 keV. Taking advantage of the high detection efficiency for low-energy γ rays, it proved possible to link every observed structure to the low-lying levels identified previously [23]. Sequences *e* and *f* form a pair of signature partner bands with negative parity based on the 7/2$^-$[743] configuration associated with the $\nu j_{15/2}$ intruder state. Known previously only up to $(9/2^-)$ and $(15/2^-)$, respectively, these two sequences are now traced to high spin, i.e., $(57/2^-)$ and $(59/2^-)$. In addition, inter-band transitions linking the two sets of levels have been identified all along the two band structures. The signature partner bands *g* and *h*, based on the 5/2$^+$[622] configuration of $i_{11/2}$ parentage, have also been delineated to high frequencies, and here as well most levels are characterized by deexcitations towards both the next in-band member and the state lower by one spin unit in the signature partner sequence. Just as in the ^{239}Pu isotone, the signature partner bands built on the 1/2$^+$[631] ground state configuration were observed to high spin in ^{237}U: band *a* was delineated up to $(57/2^+)$ and its partner, band *b*, up to $(55/2^+)$, while 8 transitions linking the partner sequences were placed as well. Another analogous feature between the two isotones is the observation of two bands *c* and *d* feeding into the ground state signature partner bands *a* and *b* through transitions of E1 character, as established from the angular correlation analysis. In two instances E1 transitions of both types, i.e., $I^- \to (I+1)^+$ and $I^- \to (I-1)^+$ have been

[1] Additional evidence for band assignments to ^{237}U can be found in a separate study of the ^{208}Pb + ^{238}U reaction described in Ref. [14].

observed. As can be seen in Fig. 3, the $21/2^-$ level of band d decays through 278 and 539 keV transitions towards both the $23/2^+$ and $19/2^+$ states of band b, while γ rays of 202 and 488 keV link the $23/2^-$ level in band c with the $25/2^+$ and $21/2^+$ band a members. This observation, together with the noted similarity with the band structures in ^{239}Pu, supports the assignment of bands c and d as rotational bands built on an octupole excitation with $K = 1/2$.

As stated above, a key indicator of strong octupole correlations in odd-mass nuclei is the presence of parity doublets: the stronger the correlations, the smaller the energy difference between quantum states with same spin and opposite parity. Thus, the assumption that ^{239}Pu is a rigid octupole rotor in its $1/2^+$ ground state would translate into an excitation energy for its $1/2^-$ doublet partner of ~ 0 keV. Instead, the experimental energy difference is 470 keV. Even though in ^{237}U the lowest negative parity state identified in the experiment is the $7/2^-$ level, the extrapolated excitation energy of the $1/2^-$ state is of the same order: ~ 496 keV. In both nuclei, these energy differences are twice as large as the value found in ^{223}Th [2], one of the odd-A nuclei exhibiting the strongest octupole correlations. Hence, in the ^{237}U and ^{239}Pu isotones, the negative parity bands appear to be associated with an octupole vibration at low spin. With increasing angular momentum, however, ^{239}Pu levels of same spin and opposite parity come closer and closer in excitation energy: the $45/2^+$ and the $45/2^-$ states are 47 keV apart, the two $49/2$ levels are separated by only 17 keV and the $53/2$ states lie within 8 keV of each other. This is illustrated in Fig. 4(a), where the markedly different behavior at high spin of ^{237}U can be noted as well. In the latter case, the energy differences remain of the order of 200 keV. Fig. 4(a) also indicates that the high spin behavior noted for ^{239}Pu mirrors that exhibited throughout the entire range by the two "octupole deformed" bands (see below) of ^{223}Th [2].

Another way to visualize sizeable octupole strength is to examine the behavior of the energy staggering factor, $S(I)$, which is defined as

$$S(I) = E_I - \frac{(I+1)E_{I-1} + I E_{I+1}}{2I+1}, \qquad (1)$$

and is a measure of the extent to which the sequences of opposite parity are interleaved in spin and can be regarded as a single rotational band of octupole character. As shown in Fig. 4(b), the staggering factor for each of the two bands of intertwined positive and negative parity states in ^{223}Th is close to zero and similar in magnitude to that seen, for example, in ^{220}Ra, one of the best examples of static octupole deformation. The behavior of $S(I)$ for the two sets of bands in ^{237}U is quite different and follows closely that seen for the yrast and the first negative parity band in ^{238}U [5]: the values of $S(I)$ decrease with spin before leveling off, a behavior reflecting the presence of band crossings associated with alignment gains discussed below. Remarkably, the staggering factor in the ^{239,240}Pu isotopes is intermediate between the Th and U cases: the $S(I)$ values are large at low spin, but become small and comparable to those seen in nuclei with octupole deformation for spins $I \geqslant 24\hbar$.

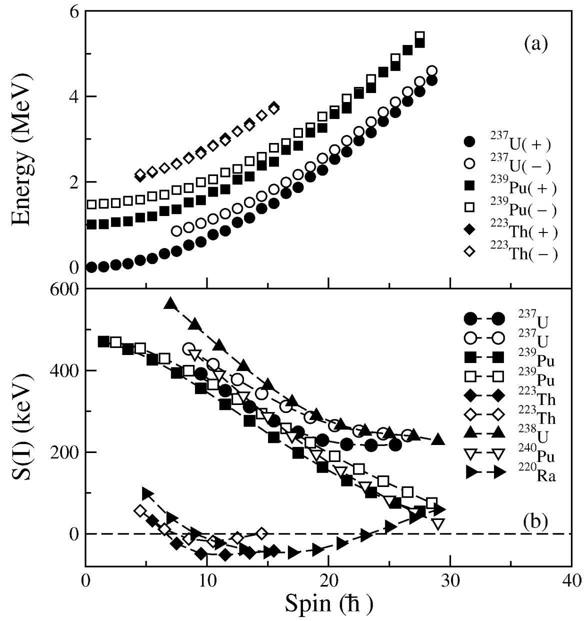

Fig. 4. (a) Excitation energy of levels as a function of spin in the positive- and negative-parity bands of ^{237}U, ^{239}Pu, and ^{223}Th. Band-head energies are displaced by 1 MeV (^{239}Pu) and 2 MeV (^{223}Th) for display purposes. (b) Energy staggering $S(I)$ as a function of spin in odd-A nuclei ^{237}U, ^{239}Pu and ^{223}Th, as well as for the even–even nuclei ^{238}U, ^{240}Pu and ^{220}Ra. Dashed lines are drawn to guide the eye.

As pointed out by several authors [2,3,13,24,25], $K = 1/2$ bands offer a convenient method to identify reflection asymmetry by determining the value of the decoupling parameters a; they are expected to follow the relationship $a(K^\pi = 1/2^+) = -a(K^\pi = 1/2^-)$ in a rigid octupole rotor limit. The strong-coupling formula,

$$E_I = \varepsilon - \frac{1}{2}\frac{\hbar^2}{2\Im} + \frac{\hbar^2}{2\Im}\big[I(I+1) + a(-1)^{(I+1/2)}(I+1/2)\big] - B\big[I(I+1) + a(-1)^{(I+1/2)}(I+1/2)\big]^2, \quad (2)$$

can be used to extract decoupling parameters, a, from a fit to the level energies of $K = 1/2$ bands. The extracted decoupling parameters for the $K = 1/2^+$ and $K = 1/2^-$ sequences in ^{239}Pu are $a = -0.45(2)$ and $a = 0.31(1)$, respectively, in marked contrast to the corresponding values $a = -0.30(1)$ and $a = 0.03(1)$ obtained for ^{237}U.[2] Thus, the decoupling parameters are of the same (small) magnitude, but opposite sign in ^{239}Pu, in line with expectations when strong octupole correlations are present. In ^{237}U, however, the Coriolis effects appear to be negligible in the negative-parity band ($a = 0$), illustrating further the contrasting situation between the two isotones.

In the framework of the rotational model, the ratio of the transition dipole (D_0) and quadrupole moments (Q_0) can be extracted from the experimental E1/E2 branchings using the expressions

$$B(\mathrm{E1}, J_i \to J_f) = \frac{3}{4}\pi D_0^2 \langle J_i K_i 10 | J_f K_f \rangle^2,$$

$$B(\mathrm{E2}, J_i \to J_f) = \frac{5}{16}\pi Q_0^2 \langle J_i K_i 20 | J_f K_f \rangle^2.$$

For the two isotones under discussion, the present data allow an extraction of the E1/E2 intensity ratios only in a limited number of cases because of the presence of doublets and other contaminant transitions in the spectra. Nevertheless, the data are sufficient to compute an average ratio of moments $R = \langle D_0/Q_0 \rangle$; the measured values $R(^{239}\mathrm{Pu}) = 3.1(2) \times 10^{-3}$ efm/eb and $R(^{237}\mathrm{U}) = 2.0(2) \times 10^{-3}$ efm/eb. These R values in turn lead to an estimate of the D_0 moments by adopting Q_0 moments deduced from an interpolation between the quadrupole moments derived from the measured $B(\mathrm{E2}, 0^+ \to 2^+)$ values [26] in the even–even neighbors ^{238,240}Pu and ^{236,238}U, respectively. Since the Q_0 moments are similar, ^{239}Pu has a larger E1 strength which translates into a D_0 moment of 0.035(2) efm ($Q_0 = 11.4(2)$ eb) which is 40% larger than the corresponding quantity in ^{237}U, $D_0 = 0.020(2)$ efm ($Q_0 = 11.0(2)$ eb), but smaller than the average moment $D_0 \sim 0.076(2)$ efm reported in Ref. [7] for ^{238,240}Pu.

All of the observations above clearly point toward octupole correlations of larger strength in ^{239}Pu than in its isotone ^{237}U. In fact, the two odd nuclei appear to follow closely the behavior of their respective even–even neighbors ^{238,240}Pu and ^{236,238}U. It was proposed in Ref. [7] that the strength of the correlations in the two even Pu isotopes may be such that a transition from an octupole vibration to a stable octupole deformation occurs at the highest spins, in agreement with the theoretical description by Jolos and von Brentano [9,10]. It is worth noting that, in both odd nuclei under consideration, the octupole excitations are based on the *same* neutron orbital and the marked difference in behavior then must reflect changes in the respective cores. This in turn points to a significant role for the additional two protons in Pu in polarizing the nuclear shape. However, these protons cannot be solely responsible for the increase in strength; the neutron number has a significant impact as octupole correlations have been shown to be weaker in both the heavier isotopes ^{242,244}Pu [7] and the lighter nucleus ^{236}Pu [27].

As shown in Refs. [11,28], strong octupole correlations or stable octupole deformation impact the magnitude of the alignment gain i_x and its evolution with rotational frequency. The relevant information is provided for the ^{239}Pu and ^{237}U bands in Fig. 5(a) and (b), and for the ^{238}U and ^{240}Pu yrast and octupole bands in Fig. 5(c). In all cases a common reference, suitable for all nuclei in the region [7,27], has been subtracted. The difference between U and Pu isotones is striking here as well. The Pu yrast bands show a small and gradual alignment increase of $\sim 2\hbar$ over the entire frequency range. The negative parity excitations experience the initial (2–3)$\hbar$ alignment increase characteristic of the octupole phonon and no further rise at higher frequency. In contrast, a strong alignment occurs in every

[2] Inertia parameters $\frac{\hbar^2}{2\Im}$ obtained from the best fit for $K = 1/2^\pm$ bands in ^{239}Pu ($\frac{1}{2}^+$: 6.11(2) keV; $\frac{1}{2}^-$: 5.18(1) keV) are very close to the corresponding values in ^{237}U ($\frac{1}{2}^+$: 6.36(3) keV; $\frac{1}{2}^-$: 5.15(3) keV). Note that in ^{237}U the fit included only the low spin levels in order to avoid any potential difficulty arising from changes in the level energies associated with alignments.

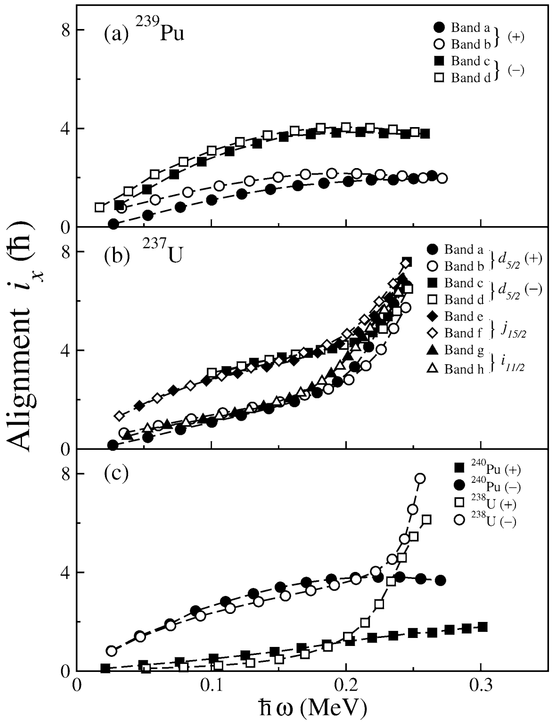

Fig. 5. Aligned spins i_x of the observed bands in ^{239}Pu (a) and ^{237}U (b) as a function of rotational frequency. The ^{240}Pu and ^{238}U alignments are shown in (c) for comparison. In all cases a common reference has been subtracted with the Harris parameters $J_0 = 65\hbar^2$ MeV^{-1} and $J_1 = 369\hbar^4$ MeV^{-3}.

sequence in the two U isotopes, i.e., they *all* experience a noticeable rise at $\hbar\omega \sim 0.25$ MeV. In Ref. [27] it was shown that this rise is due to the alignment of a pair of $i_{13/2}$ protons, in agreement with expectations based on cranked shell model calculations.[3] Thus, the dramatic difference in alignment behavior found originally in the even ^{238,240}Pu isotopes also distinguishes ^{239}Pu from the other actinide nuclei in the region.

To summarize, the present work indicates that the striking difference in behavior between the $A = 238, 240$ even Pu isotopes and other actinide nuclei extends to the odd ^{239}Pu nucleus. All the data available today indicate that strong octupole correlations in these three Pu isotopes have a considerable impact on their structure and that a transition from an octupole vibration to stable octupole deformation may occur at high spin. As pointed out in Ref. [7], several proton and neutron particle–hole configurations obeying the conditions $\Delta l = 3$, $\Delta\Omega = 0$ are close in energy to the Fermi surface ($\sim$0.5 MeV) in the immediate odd–even neighbors of ^{240}Pu. They might be expected to play a role in the ground state configuration and affect the sensitivity to octupole correlations, although the most advanced microscopic calculations, such as those of Ref. [30] using a "beyond mean field" approach, have thus far not revealed a polarization of the shape towards octupole deformation of appreciable magnitude. Clearly, the issue requires further theoretical investigation.

[3] In this context, it is noteworthy that there is no evidence for blocking of a $j_{15/2}$ neutron alignment in ^{237}U. This observation remains puzzling and is a characteristic feature of actinide nuclei in the region. The reader is referred to Refs. [27,29] for further discussion of this point.

Acknowledgements

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