Octupole Correlations in the Pu Isotopes: From Vibration to Static Deformation?

I. Wiedenhöver,[1] R. V. F. Janssens,[1] G. Hackman,[2] I. Ahmad,[1] J. P. Greene,[1] H. Amro,[1,3] P. K. Bhattacharyya,[4]
M. P. Carpenter,[1] P. Chowdhury,[5] J. Cizewski,[1,6] D. Cline,[7] T. L. Khoo,[1] T. Lauritsen,[1] C. J. Lister,[1]
A. O. Macchiavelli,[8] D. T. Nisius,[1] P. Reiter,[1] E. H. Seabury,[5] D. Seweryniak,[1] S. Siem,[1,9] A. Sonzogni,[1]
J. Uusitalo,[1] and C. Y. Wu[7]
[1]*Argonne National Laboratory, Argonne, Illinois 60439*
[2]*University of Kansas, Lawrence, Kansas 66045*
[3]*North Carolina State University, Raleigh, North Carolina 27695,*
and Triangle Universities Nuclear Laboratory, Durham, North Carolina 27708-0308
[4]*Purdue University, West Lafayette, Indiana 47907*
[5]*University of Massachusetts, Lowell, Massachusetts 01854*
[6]*Rutgers University, New Brunswick, New Jersey 08903*
[7]*University of Rochester, Rochester, New York 14627*
[8]*Lawrence Berkeley National Laboratory, Berkeley, California 94720*
[9]*University of Oslo, Oslo, Norway*

(Received 29 April 1999)

In a series of measurements with Gammasphere, striking differences were found between the yrast and negative parity bands in $^{238-240}$Pu and those in $^{241-244}$Pu. These differences can be linked to variations with mass of the strength of octupole correlations. At the highest spins, $^{238-240}$Pu are found to exhibit properties associated with stable octupole deformation, suggesting that a transition with spin from a vibration to stable deformation may have occurred.

PACS numbers: 21.10.Re, 23.20.Lv, 25.70.De, 27.90.+b

Octupole correlations have attracted much interest over the years, especially in deformed nuclei, because they are often associated with some of the lowest collective modes being observed [1,2]. Specifically, in most nuclei of the rare earth and actinide regions, rotational bands of states with odd spins and negative parity appear at excitation energies of ≤1 MeV which are interpreted as structures based on one-phonon octupole vibrations. 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, represent the best experimental evidence for the rotation of octupole-deformed nuclei. It has been shown empirically that rotation can stabilize the octupole shape [1–3].

The energy displacement between the positive and negative parity states 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 [4,5] who showed that it depends strongly on angular momentum. Furthermore, these authors also suggested that nuclei exhibiting dynamical octupole effects at low spins might develop static octupole deformation at sufficiently high angular momentum.

The present Letter examines the properties of the yrast and lowest bands of negative parity in a number of Pu isotopes. It is shown that in the $^{238-240}$Pu isotopes, where the octupole correlations are the strongest, levels of alternate spin and parity become interleaved at the highest spins ($I \sim 30\hbar$), as in the octupole bands of ^{146}Ba and ^{224}Th discussed above. Furthermore, particle alignments and transition rates are found to be affected by the strong correlations. Thus, the available experimental evidence suggests that the evolution from an octupole vibration to an octupole-deformed rotation may have been observed.

The data discussed below have been obtained in a series of so-called “unsafe” Coulomb excitation measurements, with thick targets at beam energies above the barrier, where the feeding of the highest spin states is enhanced. This technique was pioneered in Ref. [6] and was successfully used in earlier work on Cm and Pu nuclei [7]. In the present case, beams of ^{208}Pb ions at an energy of 1300 MeV from the ATLAS accelerator bombarded targets consisting of $\sim$0.3 mg/cm^2 layers of ^{240,242,244}Pu (98% enriched) electroplated onto $\sim$50 mg/cm^2 Au backing foils. These three targets provided information not only on the even Pu nuclei, but also on the odd isotopes ^{241,243}Pu produced via single-neutron transfer. Such a reaction was used to gather data on ^{238}Pu as well. In this case, a ^{239}Pu target with characteristics similar to those just described was used in conjunction with an odd-neutron ^{207}Pb beam at 1300 MeV. In all cases gamma rays were detected with the Gammasphere array, comprised of 101 Compton-suppressed Ge spectrometers. Events were written to tape when three or more suppressed Ge detectors fired in prompt coincidence. In excess of 10^8 events were collected with each target. Most of the subsequent data analysis was performed on the quadruple γ coincidence events, where the data were sorted into cubes gated on known transitions from the nuclei of interest. Additional gating conditions were placed on the γ-ray sum energy and multiplicity detected in Gammasphere. Proper subtraction of random coincidence events proved essential in order to remove the contamination of the spectra by the intense γ rays from ^{197}Au Coulomb excitation. The data analysis was performed with the programs of the RADWARE package [8]. A detailed discussion of all the level schemes and other experimental results obtained in the course of these studies is beyond the scope of the present Letter and can be found in Ref. [9].

Figure 1 compares the aligned spins i_x as a function of rotational frequency $\hbar\omega$ for the bands of interest here, i.e., for the yrast sequences and for the lowest negative parity cascades in $^{238-244}$Pu. A number of interesting features clearly stand out: (i) all the Pu isotopes with mass $A \geq 241$ exhibit a strong alignment in their respective yrast bands at $\hbar\omega \sim 0.25$ MeV, (ii) this alignment is not present at all up to the highest frequencies observed in ^{239}Pu and ^{240}Pu and is delayed at least up to $\hbar\omega \geq 0.28$ MeV in ^{238}Pu, (iii) the behavior of the alignment curve of ^{240}Pu is distinctly different from that of all the other even-even Pu isotopes, (iv) all negative parity excitations show the $\sim(2-3)\hbar$ alignment characteristic of the octupole phonon, and (v) an additional gain in alignment occurs only in the negative parity bands of the isotopes where a drastic upbend occurs along the yrast line, while, in contrast, (vi) the relative alignment between the two bands decreases in ^{240}Pu.

The observation of sudden gains in i_x of $(9-10)\hbar$ in ^{242}Pu and ^{244}Pu complements the original findings of Ref. [10] by delineating completely, for the first time, a backbending in an actinide nucleus. This i_x value is consistent with the alignment of a pair of $i_{13/2}$ protons, as predicted in Refs. [10–12]. To the best of our knowledge, all the available calculations [10–12] indicate that the same strong proton alignment should occur around $\hbar\omega \sim 0.25$ MeV in the lighter Pu isotopes, yet this expected alignment gain is not seen in ^{239,240}Pu and is delayed in ^{238}Pu. The effect is particularly striking in ^{240}Pu: only a small, smooth increase in alignment is observed over a range of four to six transitions beyond the point where the backbending occurs in the heavier Pu nuclei (Fig. 1). There is no sign of a band interaction either in the yrast or in the octupole band [13]. The remainder of this paper discusses a possible reason for this striking observation.

In all the even-even Pu nuclei, the first excited band is of negative parity and is associated with an octupole vibration [7]. It is noteworthy that the first 1^- level is the lowest in excitation energy (597 keV) in ^{240}Pu. It is located at 605 keV in ^{238}Pu, at 781 keV in ^{242}Pu and around 950 keV in ^{244}Pu [14]. This observation, together with the fact that octupole correlations are known to significantly alter alignment patterns seen in reflection symmetric nuclei [2,15], makes it worthwhile to search the data for additional indications of stronger octupole correlations near $A = 240$. Figure 2 presents the energy staggering, $S(I)$, between the odd-spin, negative parity and even-spin, positive parity bands. This staggering is defined as

$$S(I) = E(I) - \frac{E(I-1)(I+1) + E(I+1)I}{2I+1} \qquad (1)$$

and is a measure of the extent to which the two bands of opposite parity can actually be regarded as a single, rotational octupole excitation, i.e., the degree to which the odd-spin, negative parity level of spin I has an excitation energy located in between those of the two neighboring even spin, positive parity states with respective spins $I-1$ and $I+1$. In the Pu isotopes with $A \geq 242$, the values of $S(I)$ decrease with spin before leveling off, a behavior characteristic of many well deformed actinide nuclei, e.g., ^{236,238}U (not shown), which reflects the

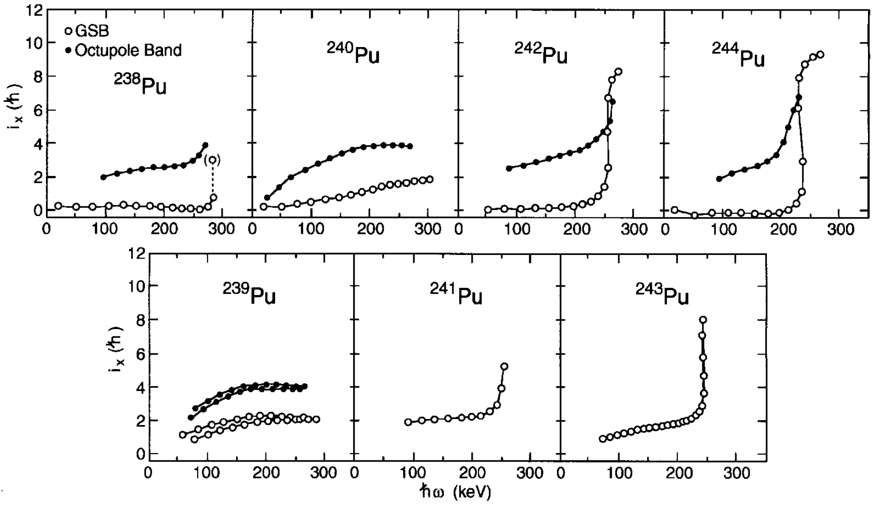

FIG. 1. Aligned spins i_x of the yrast and octupole rotational bands in the Pu isotopes. In all cases the same reference is subtracted, with the Harris parameters $J_0 = 65\hbar^2$ MeV^{-1} and $J_1 = 369\hbar^4$ MeV^{-3}.

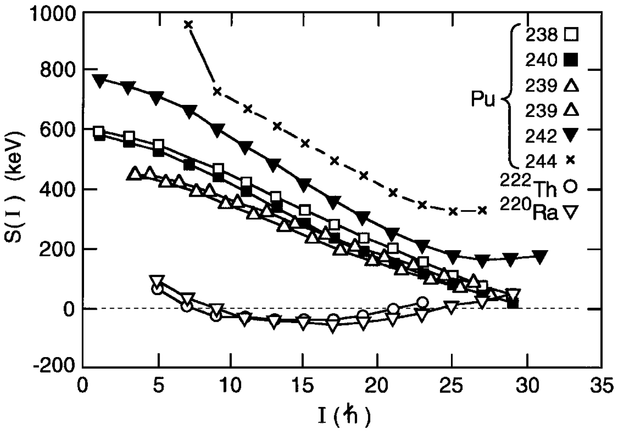

FIG. 2. Comparison of the energy staggering $S(I)$ as a function of spin I in the Pu isotopes and in ^{220}Ra and ^{222}Th (Ref. [16]), two of the best examples of octupole deformed nuclei.

presence of band crossings associated with the alignment gains visible in Fig. 1. Remarkably, the $S(I)$ values for ^{238,240}Pu continue to decrease up to the highest spins (Fig. 2), where they become small and comparable to those seen in ^{222}Th and ^{220}Ra [16], two of the best examples of nuclei with static octupole deformation. Thus, at spin $I \geq 25\hbar$ the level sequences that are seen in the two lightest even Pu isotopes display the energy spacings characteristic of an octupole rotational band. Moreover, the interleaving at high spin of states with opposite parity is also realized in the odd ^{239}Pu nucleus. The octupole vibrational bands built on the two signatures of the [631]1/2 orbital, originally proposed in Ref. [17], have been firmly established and extended in the present work. For both sequences (Fig. 2) the $S(I)$ values mirror the trend seen in the two even neighbors. Also, levels with the same spin but opposite parity are located close in energy: the $49/2^+$ and the $49/2^-$ levels are 17 keV apart, the two 53/2 states are separated by only 8 keV, and the 57/2 levels are within 26 keV. Hence, these states appear to form so-called parity doublets, as would be expected for odd nuclei with static octupole deformation [1,2]. Thus, at least from the point of view of the level energies, the three lightest Pu isotopes behave like octupole deformed rotors at the highest spins. It is worth pointing out that the decrease in relative alignment between the two bands in ^{240}Pu noted above is consistent with this interpretation.

Further indications about the importance of the octupole correlations come from the ratio of the transition dipole (D_0) and quadrupole (Q_0) moments shown in Fig. 3 for the even Pu isotopes. This ratio is extracted from the experimental $E1/E2$ branchings with the following expression:

$$\frac{D_0}{Q_0} = \sqrt{\frac{5}{12}}\,\frac{\langle I020\,|\,I-2\;0\rangle}{\langle I010\,|\,I-1\;0\rangle}\sqrt{\frac{B(E1)}{B(E2)}}\,. \tag{2}$$

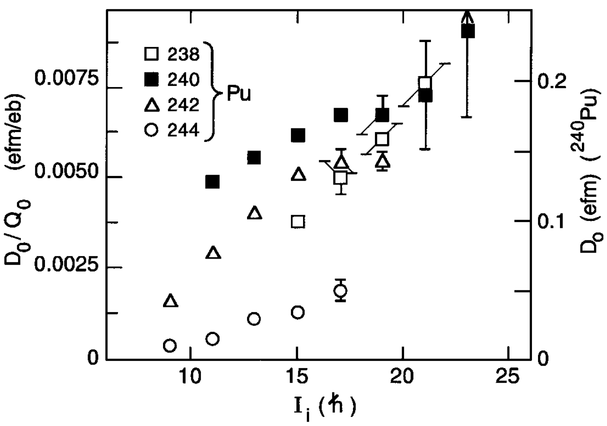

FIG. 3. Ratio of transition dipole and quadrupole moments extracted from the $E1$ and $E2$ branchings $E1{:}I_i^- \rightarrow (I_i-1)^+/E2{:}I_i^- \rightarrow (I_i-2)^-$ as a function of the spin I_i with the expression given in the text. The values of the transition dipole moment D_0 given on the right hand side are for ^{240}Pu only. They have been calculated assuming rotational $E2$-matrix elements with the Q_0 moment of the ground state band.

From the figure, it is clear that the strength of the $E1$ transitions grows with smaller mass number, as expected for the increase in the strength of octupole correlations noted above. Furthermore, the out-of-band $E1$ decays become increasingly competitive with the in-band $E2$ decays as the spin in the negative parity band increases. The effect is most pronounced for ^{240}Pu, and perhaps also in ^{238}Pu, but the data are less precise in this case. Under the assumption that the $B(E2)$ values are constant within a rotational band, this result suggests an increase in $E1$ (i.e., octupole) collectivity with angular momentum. It is also possible to derive values at each spin for the induced intrinsic dipole moment D_0 in ^{240}Pu, assuming a constant quadrupole moment $Q_0 = 25.9\ e$ b adopted from the measured $B(E2)$ value of the $2^+ \rightarrow 0^+$ ground state transition [18]. It can be seen from Fig. 3 (right hand vertical axis) that the D_0 values at high spin become quite large; $D_0 \sim 0.2\ e$ fm for $I \geq 21\hbar$ in ^{240}Pu. Such values are of the same order as the dipole moments observed in the light Th nuclei [19] ($D_0 \sim 0.2$–$0.3\ e$ fm), which are among the best examples of octupole-deformed nuclei [2].

While the experimental evidence discussed above points towards the importance of octupole correlations and, possibly, of octupole deformation in the understanding of the level sequences of $^{238-240}$Pu, some caution remains in order. First, at the highest spins where octupole deformation may have set in (Fig. 2), the level energies display the expected pattern of interleaved states with odd and even spins, but the $E1$ transitions connecting levels of opposite parity have so far not been seen. For example, the observation of the $E1$ strengths of the $27^- \rightarrow 26^+$ *and* the $26^+ \rightarrow 25^-$ transitions would constitute further support for the interpretation presented here. Unfortunately, as the positive and negative parity levels become interleaved, the $E1$ transitions are strongly suppressed by the energy factor. The expected ratio of probabilities $B(E1; 26^+ \rightarrow 25^-)/B(E2; 26^+ \rightarrow 24^+)$ is $\sim 0.016\ e\,\mathrm{fm}/e\,\mathrm{b}$ while the measured upper limit for this ratio at spin 26 is $0.05\ e\,\mathrm{fm}/e\,\mathrm{b}$; i.e., the detection of these transitions is beyond the statistical accuracy of the present data. Also, it should be recognized that while the ratios of the reduced $E1$ and $E2$ transition rates are very suggestive of octupole deformation, they are perhaps not as conclusive as one would like. Figure 3 shows that the ratios in ^{244}Pu are substantially smaller than in ^{238,240}Pu, but that those of ^{242}Pu come closer to the ^{240}Pu values. Ratios similar to those seen in ^{242}Pu have also been reported in other cases of octupole vibrations such as ^{238}U [6]. Presumably, the increase in $B(E1)$ strength with spin seen in most octupole bands reflects the importance of the dynamical effects discussed by Jolos and von Brentano [4,5] and, hence, should be expected.

To summarize, the role of octupole correlations has been investigated in a number of unsafe Coulomb excitation experiments on Pu nuclei. In the lightest isotopes, where the correlations are the strongest, their impact translates in the absence (^{239,240}Pu) or delay in frequency (^{238}Pu) of the strong proton alignment seen in the heavier Pu nuclei. Furthermore, at the highest spins, the correlations are such that the yrast and the octupole bands merge into a single sequence of levels with alternating spin and parity, large intrinsic dipole moments can be inferred from the $B(E1)/B(E2)$ ratios, and so-called parity doublets occur in ^{239}Pu. Thus, the experimental evidence suggests that a transition from an octupole vibration to stable octupole deformation may have occurred. Detailed microscopic calculations are needed to fully account for the enhanced importance of octupole correlations near ^{240}Pu. In particular, the role of the octupole-driving orbitals needs to be fully explored. In this context, it is striking that in the immediate odd-even neighbors of ^{240}Pu the $\Delta l = 3$, $\Delta\Omega = 0$ particle-hole configurations $\pi\{3/2^-[521]3/2^+[651]^{-1}\}$ and $\nu\{7/2^+[624]7/2^-[743]^{-1}\}$ [20] come close in energy to the Fermi surface (within 0.5 MeV) and are expected to play a role in the ground state configuration.

Stimulating discussions with L. Egido, F. Iachello, P.-H. Heenen, and W. Nazarewicz are gratefully acknowledged. This work is supported by the U.S. Department of Energy, Nuclear Physics Division, under Contracts No. W-31-109-ENG-38, No. DE-FG02-94ER40848, No. DE-AC03-76SF00098, and by the National Science Foundation. The authors are indebted to the office of Basic Energy Sciences, U.S. Department of Energy, through the transplutonium element production facilities at the Oak Ridge National Laboratory, for the use of Pu isotopes. One of us (S. S.) acknowledges support from a NATO grant through the Research Council of Norway.

[1] I. Ahmad and P. A. Butler, Annu. Rev. Nucl. Part. Sci. **43**, 71 (1993), and references therein.
[2] P. A. Butler and W. Nazarewicz, Rev. Mod. Phys. **68**, 349 (1996), and references therein.
[3] J. F. C. Cocks *et al.*, Phys. Rev. Lett. **78**, 2920 (1997).
[4] R. V. Jolos and P. von Brentano, Phys. Rev. C **49**, R2301 (1994).
[5] R. V. Jolos and P. von Brentano, Nucl. Phys. **A587**, 377 (1995).
[6] D. Ward *et al.*, Nucl. Phys. **A600**, 88 (1996).
[7] G. Hackman *et al.*, Phys. Rev. C **57**, R1056 (1998).
[8] D. C. Radford, Nucl. Instrum. Methods Phys. Res., Sect. A **361**, 306 (1995).
[9] I. Wiedenhöver *et al.*, in *Proceedings of the International Conference on Nuclear Structure '98, Gatlinburg, TN*, AIP Conf. Proc. No. 481 (AIP, New York, 1999), p. 527; I. Wiedenhöver *et al.* (to be published).
[10] W. Spreng *et al.*, Phys. Rev. Lett. **51**, 1522 (1983).
[11] J. Dudek, W. Nazarewicz, and Z. Szymanski, Phys. Scr. **T5**, 171 (1983), and references therein.
[12] J. L. Egido and P. Ring, Nucl. Phys. **A423**, 93 (1984).
[13] It could be argued that the aligned band is not observed due to an accidentally small mixing at the band crossing resulting in the population of the ground band only in Coulomb excitation. This would, however, not explain the absence of alignment in the negative parity band of ^{240}Pu, the reduction in relative alignment between the two bands, nor the absence of alignment in all the bands of ^{239}Pu.
[14] R. Firestone and V. S. Shirley, *Table of Isotopes* (John Wiley and Sons, Inc., New York, 1996), 8th ed., Vol. II.
[15] S. Frauendorf and V. V. Pashkevich, Phys. Lett. **141B**, 23 (1984).
[16] J. F. Smith *et al.*, Phys. Rev. Lett. **75**, 1050 (1995).
[17] M. Devlin *et al.*, Phys. Rev. C **47**, 2178 (1993).
[18] C. E. Bemis, Jr., F. K. McGowan, J. L. C. Ford, Jr., W. T. Milner, P. H. Stelson, and R. L. Robinson, Phys. Rev. C **8**, 1466 (1973).
[19] P. Schuler *et al.*, Phys. Lett. **174**, 241 (1986).
[20] It should be realized that the configurations suggested here are mixed. For example, the $\nu\{7/2^+[624]\}$ Nilsson state is mixed with the $\nu\{7/2^+[613]\}$ orbital at the deformations of interest.