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Unsafe Coulomb Excitation of $^{240-244}\text{Pu}$

I. Wiedenhöver¹, R. V. F. Janssens¹, G. Hackman¹, I. Ahmad¹, J. P. Greene¹, H. Amro¹,
M. P. Carpenter¹, D. T. Nisius¹, P. Reiter¹, T. Lauritsen¹, C. J. Lister¹, T. L. Khoo¹,
S. Siem^{1,2}, J. Cizewski^{1,3}, D. Seweryniak¹, J. Uusitalo¹, A. O. Macchiavelli⁴
P. Chowdhury⁵, E. H. Seabury⁵, D. Cline⁶, C. Y. Wu⁶

¹Argonne National Laboratory, Argonne, IL 60439

²Lawrence Berkeley National Laboratory, Berkeley, CA 94720

³University of Oslo, Oslo, Norway

⁴Rutgers University, New Brunswick, NJ 08903

⁵University of Massachusetts, Lowell, MA 01854

⁶WSRL, University of Rochester, Rochester, NY 14627

Abstract. The high spin states of ^{240}Pu and ^{244}Pu have been investigated with GAMMASPHERE at ATLAS, using Coulomb excitation with a ^{208}Pb beam at energies above the Coulomb barrier. Data on a transfer channel leading to ^{242}Pu were obtained as well. In the case of ^{244}Pu , the yrast band was extended to $34\hbar$, revealing the completed $\pi i_{13/2}$ alignment, a "first" for actinide nuclei. The yrast sequence of ^{242}Pu was also extended to higher spin and a similar backbend was delineated. In contrast, while the ground state band of ^{240}Pu was measured up to the highest rotational frequencies ever reported in the actinide region (~ 300 keV), no sign of particle alignment was observed. In this case, several observables such as the large $B(E1)/B(E2)$ branching ratios in the negative parity band, and the vanishing energy staggering between the negative and positive parity bands suggest that the strength of octupole correlations increases with rotational frequency. These stronger correlations may well be responsible for delaying or suppressing the $\pi i_{13/2}$ particle alignment.

INTRODUCTION

The stable actinide nuclei remain an interesting source of nuclear structure information as, on the one hand, they offer the possibility to investigate the interplay between collective rotation and octupole degrees of freedom, and on the other provide a unique opportunity to investigate the behavior with frequency of the proton $i_{13/2}$ and neutron $j_{15/2}$ high- j orbitals. Recent progress in the study of these nuclei has come from the work of Ward et al. [1] and Hackman et al. [2] where the power of inelastic scattering at beam energies slightly above the Coulomb barrier on thick targets was used to study collective excitations with high sensitivity. This technique is often referred to as "Unsafe" Coulomb Excitation. Besides the higher than usual beam energy, which optimizes the population of the highest spin levels, the technique is also associated with the use of thick targets and with the selection of the cascades of the highest multiplicity as a means to enhance the γ rays emitted after the excited nucleus has come to rest. In this way most of the transitions in a collective cascade are measured with the intrinsic resolution of the Ge detectors.

EXPERIMENT

The experiments described here represent the first "production" runs with GAMMASPHERE at ATLAS. Targets of isotopically enriched ^{240}Pu and ^{244}Pu were bombarded with a 1300 MeV ^{208}Pb beam. The Pu material was electroplated onto a thick (50 mg/cm^2) Au foil, which served as a backing to stop both the recoiling Pu nuclei and the beam. The targets had a thickness of $\approx 300\mu\text{g/cm}^2$. In addition, a second, thin Au foil ($\approx 50\mu\text{g/cm}^2$) was mounted in front of each target to minimize the danger of contaminating the experimental equipment by the release of radioactive target material. It should be pointed out that the

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relatively low target thickness achievable with electroplating constitutes the main experimental limitation in these measurements.

During three days of beam time, coincidence events of fold three or higher were collected. Most of the subsequent data analysis was performed on the quadruple γ coincidence events, where the data were sorted into a coincidence cube gated on known transitions from the nuclei of interest. Proper subtraction of random coincidence events proved essential in order to remove the contamination of the spectra with the intense ^{197}Au Coulomb excitation γ rays. The data analysis was performed with the programs of the RADWARE package [3].

Fig. 1 presents sample spectra of the yrast bands of ^{244}Pu and ^{242}Pu obtained as sums of the cleanest coincidence gates. These spectra were measured with the ^{244}Pu target. Here ^{242}Pu is produced by a direct two-neutron transfer reaction, as demonstrated by the presence of the ^{210}Pb ground-state transition in the coincidence spectra.

RESULTS

$\pi i_{13/2}$ Quasiparticle Alignment

In ^{244}Pu , the yrast rotational band could be traced up to the 34^+ level. A pronounced backbending is present at a rotational frequency of $\hbar\omega \approx 240$ keV (see figs. 1 and 2). This result represents the first instance in the actinide region where a backbending is now fully delineated. The resulting gain in alignment ($\sim 10\hbar$) is consistent with a $i_{13/2}$ quasi-proton alignment, but is too low for a $j_{15/2}$ quasi-neutron alignment. Thus, the present result settles a long standing debate about the nature of quasiparticle alignments in the actinide region [1,4]. The observed behavior can be reproduced both by cranked shell model and by HFB calculations which find that the quasi-proton alignment occurs at lower frequency than the quasi-neutron one. The behavior with frequency of the ^{242}Pu yrast band is very similar to that seen in ^{244}Pu : a strong backbending is present here as well, starting at a frequency of ~ 250 keV.

Because of the success of microscopic calculations in understanding the yrast sequences of ^{242}Pu and ^{244}Pu , the behavior of the ground-state band of ^{240}Pu appears to be rather puzzling. Being a member of the same isotopic chain, with a deformation essentially identical to that of the heavier even-even Pu nuclei, ^{240}Pu would be expected to exhibit a similar backbend at roughly the same frequency. Yet its yrast sequence, which is extended in the present work up to 32^+ and a rotational frequency of $\hbar\omega \approx 300$ keV, shows no indication of any irregularity. In fact, this ground-state band exhibits all the characteristics of a single, "well-behaved" rotational band, without any sign of a band crossing in either the alignment plot (fig. 2) or the evolution of the dynamic moment of inertia with frequency (not shown). Thus, the alignment process of the $i_{13/2}$ protons and/or of the $j_{15/2}$ neutrons appears to be either suppressed or at least delayed towards higher rotational frequencies. Possible explanations for this observation can be found in the behavior of the negative parity bands discussed below.

Negative-Parity Bands

The presence of low lying rotational bands of negative parity is a characteristic feature of the level structure of all actinide nuclei. These bands have been associated with various components of the octupole phonon [1,2]. In both ^{240}Pu and ^{244}Pu , these octupole bands have now been followed up to high spin (29^- and $\hbar\omega \sim 270$ keV in ^{240}Pu , 27^- and $\hbar\omega \sim 230$ keV in ^{244}Pu). The measured alignments are again quite different for the two isotopes (fig. 2). In the lighter ^{240}Pu isotope, an initial gain in alignment of $3\hbar$ is followed by a value which becomes constant with frequency as would be expected for the $K=0$ component of the octupole phonon [2]. While a similar low frequency rise of the alignment towards the same $3\hbar$ value occurs in ^{244}Pu , it is followed by a further increase at higher frequencies which mirrors that seen in the yrast sequence. Thus, the behavior discussed previously for the yrast sequences is also present in the negative parity bands.

The behavior of the negative parity bands in the two isotopes is also found to be markedly different when the excitation energy of the levels is considered. In fig. 3, the so-called energy staggering $S(I)$ between the odd-spin, negative parity and even-spin, positive parity bands is presented. This staggering is defined as:

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

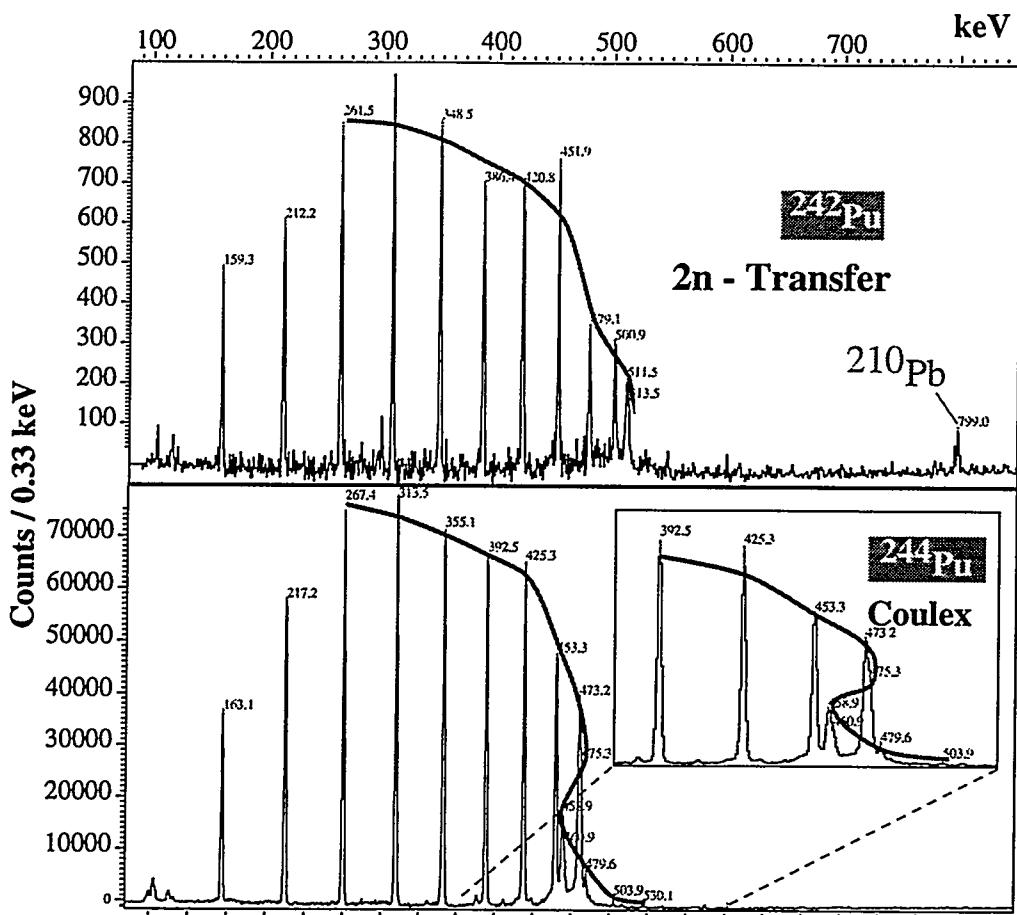


FIGURE 1. Sample coincidence spectra for the $^{242,244}\text{Pu}$ yrast bands. ^{242}Pu was obtained as a direct, two-neutron transfer reaction product from the ^{244}Pu target, as illustrated by the presence of the ^{210}Pb ground-state transition.

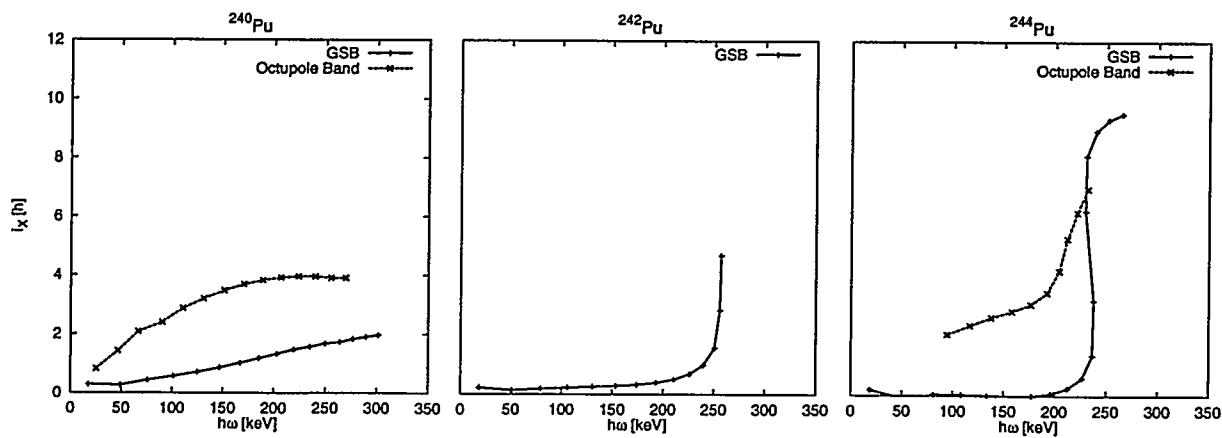


FIGURE 2. Alignment of the ground state and octupole rotational bands in $^{240,242,244}\text{Pu}$. In all cases the same reference is subtracted, with the Harris parameters $J_0 = 65\text{h}^{-2}\text{MeV}^{-1}$, $J_1 = 369\text{MeV}$.

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 level of spin I has an excitation energy located in between those of the two neighboring even spin states with respective spins $I - 1$ and $I + 1$.

The experimental $S(I)$ values for ^{240}Pu , ^{244}Pu , ^{238}U and ^{222}Th are displayed in fig. 3. As expected, the quintessential “octupole-deformed” nucleus ^{222}Th exhibits essentially zero staggering. The staggering observed in the three heavier actinide nuclei is very large at low spin as expected for an octupole vibrational band, but decreases with spin. However, only in ^{240}Pu does the value of $S(I)$ approach zero, while $S(I)$ levels off in ^{238}U and ^{244}Pu . Thus, at first sight, it appears that only in ^{240}Pu the yrast and near-yrast structure evolves from a vibrational character at low spin to a situation approaching octupole rotation at higher frequencies. If this interpretation is correct, ^{240}Pu would represent experimental confirmation of the model for octupole bands proposed by Jolos and von Brentano [5] where an octupole vibration evolves with rotational frequency into an octupole rotation as a result of the Coriolis interaction. This interpretation also would provide a natural explanation for the absence or delay of the backbending along the yrast line as octupole correlations are known to alter significantly alignment patterns seen in axially symmetric nuclei [6,7]. Nevertheless, further experimental evidence is highly desirable. The latter can come, at least in part, from a careful examination of the E1 and E2 branching ratios discussed in the following section.

Branching Ratios

The branching ratios for the E1 transitions linking the negative parity band to the yrast band were analyzed with the formalism of generalized intensity ratios described in ref. [9].

$$\sqrt{B(E\lambda)} = Q_t < I_i K_i \lambda (K_f - K_i) | I_f K_f > (1 + q(E\lambda) [I_f(I_f + 1) - I_i(I_i + 1)]) \quad (2)$$

In the present case, the expressions for the $B(E1)$ and $B(E2)$ transitions probabilities reduce to the following formulae:

$$\begin{aligned} \sqrt{B(E1, I_i \rightarrow I_i - 1)} &= D_0 < I_i 0 10 | (I_i - 1) 0 > (1 + q(E1) [-2I_i]) \\ \sqrt{B(E1, I_i \rightarrow I_i + 1)} &= D_0 < I_i 0 10 | (I_i + 1) 0 > (1 + q(E1) [2(I_i + 1)]) \\ \sqrt{B(E2, I_i \rightarrow I_i - 2)} &= Q_0 < I_i 0 20 | (I_i - 2) 0 > \end{aligned} \quad (3)$$

In these expressions, the effects of Coriolis mixing can be found in the so-called decoupling coefficient $q(E1)$, which can be calculated in models of the intrinsic motion, such as the cranked RPA model [10], for example. In fig. 4, the $\sqrt{B(E1)}$ -branching ratios, corrected with the appropriate Clebsch-Gordan coefficient of equation 3, are displayed for the cases of ^{238}U [1], ^{240}Pu and ^{244}Pu . The data can be described with a very small decoupling coefficient in the ^{240}Pu and ^{238}U cases. In contrast, the data for ^{244}Pu require a much larger value of $q(E1) = 0.065\hbar^{-1}$. This situation agrees nicely with cranked RPA calculations [2,11], which predict that the $K=0$ component of the octupole phonon is lowest in ^{240}Pu (giving rise to a very small $q(E1)$ value), but that the other K -components are much closer in excitation energy in ^{244}Pu , resulting in substantial configuration mixing due to the Coriolis force.

While the E1-branching ratios can be consistently explained by the general intensity relations of eq.3, the branching between the in-band E2 and the out-of-band E1 transitions present a much different picture. In figure 5, the ratios of the E1 matrix elements to the E2 ones are presented as a function of the spin-difference parameter $[I_f(I_f + 1) - I_i(I_i + 1)]$. In this plot, the $I_i^- \rightarrow (I_i - 1)^+$ transitions (e.g. the so-called “downhill” transitions) correspond to negative values of the spin-difference parameter. Conversely, the so-called “uphill” transitions, corresponding to the $I_i^- \rightarrow (I_i + 1)^+$ sequence, are associated with positive values of the same parameter. Again, the data for the measured ratios in ^{244}Pu show a good agreement with the values calculated with the same $q(E1) = 0.065$ decoupling parameter extracted from the E1-branching ratios. Thus, a clear picture appears to emerge in this case.

At the same time, the ^{240}Pu data exhibit E1 matrix elements which increase strongly with spin, i.e. the $B(E1)$ reduced probabilities compete more favorably with the in-band $B(E2)$ probabilities at the highest spins. Under the usual assumption that the transition quadrupole moments Q_0 are constant within a rotational band, this result implies an increase in E1 (i.e octupole) collectivity with angular momentum. This behavior cannot be reproduced with a single value of the $q(E1)$ parameter.

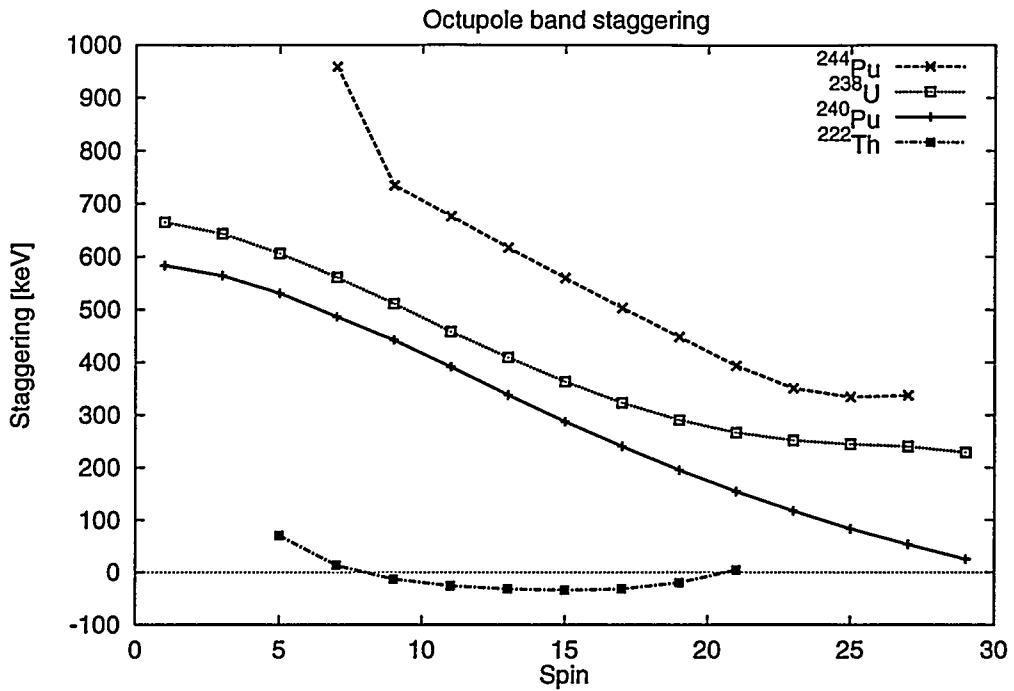


FIGURE 3. staggering parameter (see eq. 1) for negative and positive parity bands in ^{244}Pu , ^{238}U , ^{240}Pu and ^{222}Th .

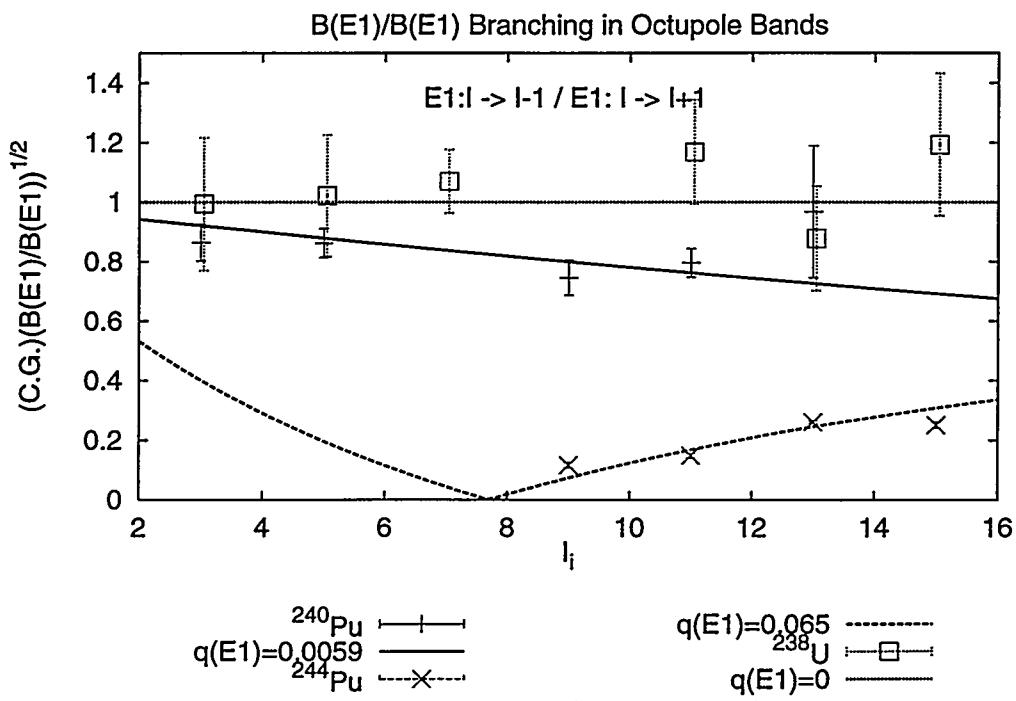


FIGURE 4. Ratio of the E1-matrix elements $I_i^- \rightarrow (I_i - 1)^+ / I_i^- \rightarrow (I_i + 1)^+$ as a function of I_i for the octupole bands of ^{240}Pu , ^{244}Pu and ^{238}U [1]

The right y-axis of fig. 5 is labeled with the transition dipole moment values D_0 derived from the measured transition probability ratios assuming a constant $Q_0 = 25.88eb$ quadrupole moment. The latter value of Q_0 is adopted from the measured $B(E2)$ value of the $2^+ \rightarrow 0^+$ ground state transition [8]. It can be seen from fig. 5 that the D_0 values at high spin become quite large. In fact, they are comparable to the dipole moments observed in ^{222}Th , an octupole deformed nucleus [7]. Although experimental data on absolute lifetimes and/or $B(E2)$ reduced transitions probabilities are highly desirable, the present analysis strongly suggests a substantial increase of the transition dipole moment D_0 with spin, until it reaches values consistent with a sizable octupole deformation.

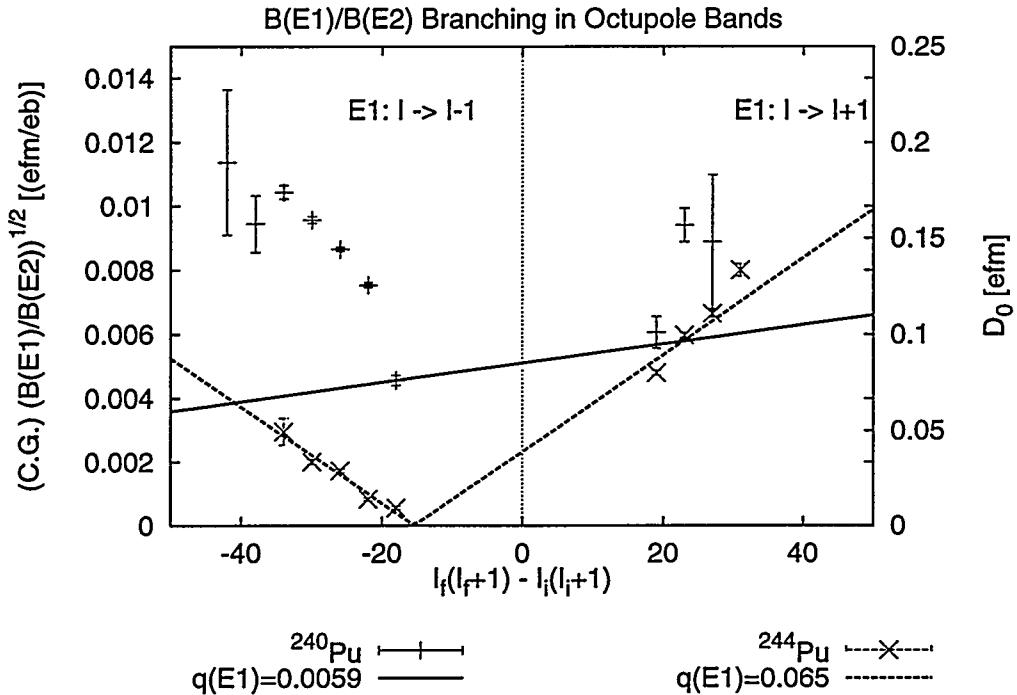


FIGURE 5. Ratio of the E1-matrix elements to the E2-matrix elements $E1 : I_i^- \rightarrow (I_i \pm 1)^+ / E2 : I_i^- \rightarrow (I_i - 1)^-$ as a function of the parameter $I_f(I_f+1) - I_i(I_i+1)$. The $I_i^- \rightarrow (I_i - 1)^+$ decays correspond to the negative x-values, the $I_i^- \rightarrow (I_i + 1)^+$ to the positive values. The values of D_0 given on the right hand side are calculated assuming rotational E2-matrix elements with the Q_0 of the ground state band.

CONCLUSIONS

The method of unsafe Coulomb excitation at GAMMASPHERE has provided a wealth of information on the nuclei ^{244}Pu and ^{240}Pu . The data allowed for the first complete delineation of a quasiparticle alignment in an actinide yrast sequence. This alignment has been understood to be of proton character. In sharp contrast, the yrast band of ^{240}Pu shows no signs of backbending up to the highest rotational frequencies. In the present work this surprising observation has been linked to the presence of octupole correlations increasing in strength at high spin. These correlations may well block the expected alignment or, at least, shift it to higher frequencies. This interpretation is corroborated by the observation of a vanishing energy-staggering between the octupole and ground state bands at the highest frequencies and by E1 matrix elements which also increase strongly with spin. This interpretation, if correct, represents the first experimental evidence for the gradual change of a band associated with an octupole vibration into one of collective, rotational octupole character for a well deformed nucleus.

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