Contrasting Behavior in Octupole Structures Observed at High Spin in ²²⁰Ra and ²²²Th

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Alternating-parity states connected by strong E1 transitions, characteristic of a reflection-asymmetric rotor, have been observed to high spins in the isotones ²²⁰Ra and ²²²Th. This level structure is observed up to $J^{\pi} = 29^{-} (31^{-})$ in ²²⁰Ra while it cannot be seen beyond $J^{+} = 24^{+} (25^{-})$ in ²²²Th. These observations are consistent with Woods-Saxon-Bogolyubov cranking calculations which predict that the yrast band of ²²²Th will undergo a shape transition at $J = 24\hbar$, in contrast to that of ²²⁰Ra which maintains its reflection asymmetry to higher spins.

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In the last decade the long-standing prediction [1,2] of stable octupole deformation in certain nuclei has gained experimental vindication. An octupole-deformed nucleus has reflection asymmetry or a pear shape and is expected to exhibit some of the spectroscopic characteristics of asymmetric molecules such as HCl. "Quasimolecular" rotational bands of alternating-parity states connected by strong electric-dipole transitions are the experimental signatures of octupole-deformed nuclei, and many of these octupole bands have been observed [3], particularly in nuclei around ²²⁴Th and ¹⁴⁶Ba. It has been shown empirically that rotation can stabilize the octupole shape. With the advent of large germanium-detector arrays designed for high-spin studies, it has become possible to trace the evolution of the octupole shape to the highest observable rotational frequencies.

Much theoretical work has been devoted to the description of octupole nuclei [4-6] and their rotation [7–9]. Cranked Woods-Saxon-Bogolyubov-Strutinsky calculations have been performed by Nazarewicz, Leander, and Dudek [8] to trace the development of the octupole deformation to high spin. A spectroscopic phenomenon observed when these nuclei rotate is the absence of backbending which can be present if the nucleus is reflection symmetric. This is due to the property of the reflection-asymmetric octupole potential, in that it can mix high-*j* intruder orbitals of opposite parity with the low-*i* normal-parity states in a given shell, so that any aligned angular momentum is fragmented over several states. Nuclei, with $Z \simeq 88$ and $N \simeq 134$, have their neutron and proton Fermi levels in close proximity to the octupole-driving $\nu(j_{15/2} \text{ and } g_{9/2})$ and $\pi(i_{13/2} \text{ and } f_{7/2})$ orbitals, rendering these nuclei particularly susceptible to octupole deformation. The content of this Letter focuses on the high-spin behavior of two such isotones, $^{222}\mathrm{Th}$ and $^{220}\mathrm{Ra}.$

Previous studies have shown that both ²²⁰Ra [10–14] and ²²²Th [15-17] exhibit octupole bands up to spins $22^+/23^-$ and $24^+/23^-$, respectively. The absence of a previously predicted [18] backbend at low spin in ²²²Th was explained by the presence of octupole deformation [19]. More detailed calculations [8] predict that, while the reflection-asymmetric shape should persist to high spin in ²²⁰Ra, in ²²²Th a shape transition from a reflectionasymmetric to a reflection-symmetric shape should occur at spin $\sim 24\hbar$. The only experimental observation of such an effect has been reported in ¹⁵⁰Sm [20] where the octupole band develops at $\sim 8\hbar$, but at $\sim 15\hbar$ this crosses a reflection-symmetric band. In the Ra and Th nuclei, the known level schemes do not extend to sufficiently high rotational frequencies to test these predictions. The purpose of this work is to investigate the yrast level structures of both ²²⁰Ra and ²²²Th at high spin where shape changes are expected to occur.

High-spin states were simultaneously populated in the reactions 208 Pb(18 O, $\alpha 2n$) 220 Ra and 208 Pb(18 O, 4n) 222 Th. The 95 MeV 18 O beam was provided by the tandem Van de Graaf accelerator at the Nuclear Structure Facility, Daresbury Laboratory. The target consisted of two stacked foils of 208 Pb, each of thickness 500 $\mu g/cm^2$. Deexcitation gamma rays were collected with the Eurogam (phase I) gamma-ray spectrometer [21–23], consisting of 40 large volume (70% efficiency, relative to a 3 in. \times 3 in. sodium-iodide detector, at 1.33 MeV) escape-suppressed germanium detectors arranged in rings of 9, 4, 3, 9, 10, and 5 detectors at angles of 72.0°,

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85.8°, 94.2°, 108.0°, 133.6°, and 157.6° to the beam axis, respectively. After 48 h of data collection, with the requirement that 5 or more unsuppressed-germanium signals were in coincidence, a total of 1.1×10^9 triple and 0.4×10^9 quadruple escape-suppressed gamma-ray coincidences were recorded. Of the useful coincidence data, corresponding to the evaporation-residue channel (5% of the total reaction), 68% were ²²²Th, 25% were ²²⁰Ra, and 7% were ²¹⁹Ra. The data were analyzed by constructing selected gamma-gamma correlation matrices from fourfold suppressed-germanium coincidences, with the condition that at least two of the four gamma-ray energies in a fourfold event were from the desired reaction channel. Energy and relative efficiency calibration for the Eurogam spectrometer were obtained using ¹⁵²Eu and ¹⁸²Ta sources.

Fourfold coincidence spectra for each nucleus are presented in Figs. 1(a) and 1(b), which serve to illustrate the quality of the data. The level schemes obtained from the coincidence data using energy sums and intensity balance arguments are shown in Figs. 2(a) and 2(b). The spins and parities of the lower levels were taken from previous measurements [10,15] and angular-correlation information has been extracted from the data for the higher spin states. Gamma-gamma correlation matrices were constructed from which it was possible to measure the intensity in the 5 detectors at $\theta = 157.6^{\circ}$ (with azimuthal angles $\phi = 0^{\circ}, \pm 72.0^{\circ}, \pm 144.0^{\circ})$ in coincidence with the 10 detectors at $\theta = \sim 90^{\circ}$ (5 detectors at $\theta = 85.8^{\circ}$ with $\phi = 180.0^\circ, \pm 108.0^\circ, \pm 36.0^\circ$ and 5 detectors at $\theta = 94.2^{\circ}$ with $\phi = 0^{\circ}, \pm 72.0^{\circ}, \pm 144.0^{\circ}), I(157.6, 90),$ and similarly the intensity in the detectors at $\theta \approx 90^{\circ}$ was measured in coincidence with those at $\theta = 157.6^{\circ}$, I(90, 157.6). The angular-intensity ratios $I_{90}^{157.6} =$ [I(157.6,90) - I(90,157.6)]/[I(157.6,90) + I(90,157.6)]were compared for the observed transitions, in order to assist in the assignment of transition multipolarities via the method of directional correlations from oriented states [24]. When the gating transition was one of the transitions previously shown to be a stretched dipole [10,15], the ratio $I_{90}^{157.6}$ was found to have values ranging from -0.03 ± 0.03 to 0.36 ± 0.28 for stretched dipole transitions and from -0.36 ± 0.17 to -0.07 ± 0.02 for stretched quadrupole transitions, with weighted mean values of 0.03 ± 0.03 and -0.22 ± 0.12 , respectively. The results of the directional-correlation analysis support the spin assignment of Fig. 2 where measurable, that is, up to 20^+ in ²²²Th and 24^+ in ²²⁰Ra.

The yrast band in ²²⁰Ra, as shown in Fig. 2(a), has been observed up to $J^{\pi} = 29^{-}$ (possibly 31⁻). These are the highest-spin states reported in such an octupole band, equaled only by those in ²¹⁸Ra [25]. In addition to the intraband *E*2 transitions, the interband *E*1 transitions have also been observed to the highest state. In sharp contrast, only a tentative placement of a state above the 23⁻ state, presumably with $J^{\pi} = 25^{-}$, has been made for



FIG. 1. Spectra showing transitions in (a) ²²⁰Ra and (b) ²²²Th comparing the two reaction channels and illustrating the quality of the data. The spectra are from unfolded quadruple gammaray coincidences where a gamma ray must be in coincidence with three gamma rays from the particular channel before the spectrum is incremented. The gating-transition energies for the ²²⁰Ra spectrum are 128.1, 152.4/153.6 (doublet), 156.7, 231.6, 313.3, and 399.1 keV. The gating-transition energies for the ²²²Th spectrum are 161.1/161.2 (doublet), 170.1, 182.9, 206.4/206.9 (doublet), 211.1, 256.0/256.2/256.3 (triplet), and 310.2 keV. The insets compare the high-energy portions of the spectra. The label "d" indicates a doublet. (c) Plot of the sums of intensities of the depopulating transitions as a function of spin for the states in above $12\hbar$. The intensities are given relative to the 4⁺ to 2⁺ transition in ²²²Th. The horizontal lines at spin values $26\hbar$ and $30\hbar$ indicate the observational limits for ²²²Th are spectruely.

²²²Th [Fig. 2(b)]. The observed octupole band in ²²²Th appears to terminate rather abruptly with the last observed 500.7 keV (24⁺ to 22⁺) transition, as shown in the inset of Fig. 1(b). This is also illustrated by Fig. 1(c), which shows the sums of the intensities of the depopulating transitions for the states above 12⁺ plotted against spin. The intensities are normalized to that of the 4⁺ to 2⁺ in ²²²Th. The horizontal lines at 26 \hbar and 30 \hbar show the observational limits for ²²²Th and ²²⁰Ra, respectively. The intensities in ²²⁰Ra appear to gradually approach the observational limit over the spin range plotted, whereas



FIG. 2. The level schemes for (a) 220 Ra and (b) 222 Th, deduced in this work. The transition energies have errors which range from 0.2 keV for the strongest transitions up to 0.4 keV for transitions between the highest spin states observed. The level energies are taken from a weighted mean of the dipole sums and quadrupole energies, added to the preceding level energy. The numbers in brackets, where given, are the intensities, relative to the 4⁺ to 2⁺, corrected for internal conversion. The error on the intensity ranges from 10% to 30%. This includes a contribution from the systematic error which arises from the nature of the multiple-fold coincidences. The numerous degenerate transitions have larger errors on both energy and intensity values.

those in ²²²Th drop rapidly above 22 \hbar . Despite the cutoff of the yrast octupole band at ~500 keV several strong gamma-ray transitions with energies >500 keV have been identified and assigned to ²²²Th. For example, a 533 keV transition feeds into the octupole band at spin 18 \hbar and a 575 keV transition feeds in at spin 11 \hbar . Both of these transitions are clearly visible on the inset of Fig. 1(b).

An explanation for the sudden nonobservation of states beyond $25\hbar$ in ²²²Th is the predicted shape transition [8] and corresponding loss in population of the octupole band. Another possible explanation is that there are differing fission strengths on the path from the compound nucleus to each evaporation residue. If, on the path to ²²⁰Ra, the alpha particle is emitted before either of the neutrons then the probability of fission is reduced at each of the remaining steps, primarily because the value of Z^2/A is reduced. The increased fission barrier for the smaller Z system will allow it to support more angular momentum. To investigate this effect, a second experiment was performed at the JYFL K-130 cyclotron facility in Jyväskylä, Finland. Under identical reaction conditions, gamma rays were collected by a germanium-detector array, consisting of 12 ($\sim 20\%$ efficiency) escape-suppressed germanium detectors [26] and one 3 in. \times 3 in. sodium-iodide detector. By comparing the number of counts in germanium singles to those in germanium-sodium-iodide coincidences, a value for the multiplicity of the ²²²Th reaction channel was measured to be 12.7 ± 1.6 which is in agreement with the value of 13.0 ± 0.5 for the same reaction at 93 MeV from Ref. [15]. It was not possible to use this method with any accuracy for ²²⁰Ra because of contaminants in the single spectra. In order to accurately compare the multiplicities of the two reaction channels, ratios of the numbers of threefold to twofold germanium coincidence events were determined. The multiplicity M is related to the number of threefold, N_3 , and twofold coincidences, N_2 , via the expression $N_3/N_2 = k(M - 2)$, where k is a constant of proportionality. In this analysis an event was counted only if it contained at least two gamma rays whose energies corresponded to transitions in the nucleus of interest. Using this technique the ratio of multiplicities for the 220 Ra/ 222 Th channels was found to be 1.01 \pm 0.07. This ratio has been corrected for the $\sim 2\%$ effect of evaporation neutrons in the germanium detectors [27]. In contrast to this result, the values of normalized intensity sums for all the observed gamma transitions were measured to be 13.9 ± 0.3 for ²²⁰Ra and 9.9 ± 0.2 for ²²²Th. This suggests that there are more feeding transitions in ²²²Th compared with ²²⁰Ra. These results imply that the probable explanation for the lack of intense transitions above spin $24\hbar$ in ²²²Th is that the yrast octupole band crosses one or more bands and the intensity is shared over several states rendering the depopulating transitions too weak to be observed.

The calculations of Ref. [8] predict that, for ²²²Th, a reflection-asymmetric configuration remains yrast until 24 \hbar when a reflection-symmetric band of aligned $\nu(j_{15/2})$ and $\pi(i_{13/2})$ pairs becomes lowest in energy. For ²²⁰Ra the spherical ground state becomes both octupole and quadrupole deformed under rotation, but in this case the four quasiparticle $\beta_3 = 0$ configuration is not as favored in energy so the octupole band structure continues to very high spin. Figure 3 shows how the experimental angularmomentum alignment varies smoothly with rotational frequency in both nuclei. The spin value at which the band crossing is predicted to occur in ²²²Th is marked by the dashed line and the last experimental point for ²²²Th



FIG. 3. Aligned angular momentum I_x for ²²⁰Ra and ²²²Th plotted as a function of rotational frequency $\hbar \omega$. The dashed line represents the spin value where the shape transition is predicted to occur in ²²²Th [8]

lies just below the position of the predicted band crossing. The data points for 220 Ra can be seen to continue well above the 222 Th band crossing, in agreement with the prediction that the band crossing in 220 Ra should occur at higher spin than in 222 Th.

In summary, interleaving bands of alternating parity states have been seen to high spin in ²²⁰Ra and ²²²Th. The band in ²²⁰Ra has been observed to continue up to spin 31 \hbar in contrast to that in ²²²Th which could not be observed above $25\hbar$, whereas the measured multiplicities suggest that the two nuclei are populated with similar mean angular momenta. The abrupt loss of intensity of the octupole band at spin $25\hbar$ in ²²²Th can be explained by the assumption that a crossing of one or more bands occurs so that the depopulating intensity becomes fragmented. A candidate for such a crossing band would be the 4 quasiparticle reflection-symmetric band predicted by Nazarewicz, Leander, and Dudek [8] to become yrast at comparable spin values. The same theory predicts that such a band crossing occurs at higher spin in ²²⁰Ra, which is consistent with the experimental observation.

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