



$N = 151$ Pu, Cm and Cf nuclei under rotational stress: Role of higher-order deformations



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ABSTRACT

Fast-rotating $N = 151$ isotones ^{245}Pu , ^{247}Cm and ^{249}Cf have been studied through inelastic excitation and transfer reactions with radioactive targets. While all have a ground-state band built on a $\nu j_{15/2}[734]9/2^-$ Nilsson configuration, new excited bands have also been observed in each isotope. These odd- N excited bands allow a comparison of the alignment behavior for two different configurations, where the $\nu j_{15/2}$ alignment is either blocked or allowed. The effect of higher order deformations is explored through cranking calculations, which help clarify the elusive nature of $\nu j_{15/2}$ alignments.

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The spectrum of single-particle energies in very heavy and superheavy nuclei is a key to understanding their surprising stability and robustness. The exact sequence of levels and the gaps between them are responsible for both static properties, such as ground-state deformation, moments and spins, as well as dynamic properties like rotational inertia and pair fields. While the static aspects of very heavy nuclei close to their deformed ground states have been the primary focus for decades, studied through α - and β -decay and light-ion transfer reactions, recent techniques have allowed us to explore the nuclear dynamics and study nuclei under significant rotational stress. New experimental data in deformed heavy actinides near $Z \approx 100$ and $A \approx 250$ have provided many surprises and challenges to theory. Part of the stimulation stems from the fact that deformation and rotation draw levels from above the next magic spherical shell gaps thought to stabilize superheavy nuclei into the experimentally accessible domain. Lacking a true ab initio theory, there has been intense effort in empirical modeling. These include macroscopic-microscopic approaches with cranked mean field potentials [1,2], self-consistent

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Hartree–Fock–Bogoliubov calculations with Skyrme [3] or Gogny [4] forces, relativistic density functional theories [5,6], and projected shell models [7].

One unresolved issue in these very heavy systems under rotational stress surrounds neutron alignment. In general, rotations built on states with the highest intrinsic angular momentum exhibit the most dramatic variation due to the Coriolis force. For example, the alignment of protons occupying the $i_{13/2}$ orbit in plutonium isotopes is well known [8]. One might expect that for neutrons, which fill the next major shell, the occupancy of the $j_{15/2}$ orbit would be similarly dramatic and clear. Surprisingly, at present, there is no clear evidence for this alignment, despite it being anticipated in “standard” cranking calculations [9,10]. Obtaining new information on neutron alignments is the goal of the current research. Conceptually, studying odd- N systems is the best way to investigate such effects for neutrons, as the presence of the odd particle in different orbits can either allow or block alignment. Such studies turn out to be extremely difficult experimentally, as the cross sections for high-spin excitations are low, and the very fact that odd- A nuclei have many bands means that the population is distributed into several rotational sequences with near-identical level spacings, and, consequently, identical de-exciting γ decays at low energies. This, combined with the need to use radioactive targets, and the fact that internal conversion suppresses low-energy γ -ray emission, presents a daunting challenge which can only be approached with the most sophisticated instruments in our arsenal.

Prior to the present work, only ground-state bands were known up to high spins for the $N \geq 150$ odd- A nuclei $^{247,249}\text{Cm}$ ($Z = 96$) [10], ^{249}Cf ($Z = 98$) [10], ^{253}No ($Z = 102$) [11,12], ^{251}Md ($Z = 101$) [13] and ^{255}Lr ($Z = 103$) [14]. For populating high-spin states in transfermium ($Z \geq 100$) nuclei, fusion–evaporation reactions are the only choice, where typical cross sections are of the order of sub-microbarns to nanobarns. For $(94 \leq Z \leq 98)$ transplutonium nuclei, inelastic and transfer reactions with radioactive targets and heavy-ion beams provide a viable alternative, with cross sections of the order of tens of millibarns for the inelastic channels, with roughly an order of magnitude drop for each neutron transfer. The relatively higher cross sections compared to fusion reactions are somewhat offset by signal-to-noise challenges from the intense γ -ray background from the radioactive targets themselves, fission competition, highly fragmented population of multiple channels, as well as strong excitations of the substrate on which the radioactive targets are deposited (such as ^{197}Au). Nevertheless, inelastic and transfer reactions provide a complementary and more comprehensive exploration of the physics of the highest neutron orbitals.

For $N = 151$ nuclei, limited spectroscopic information exists for ^{245}Pu ($Z = 94$) [15], although no well-developed rotational bands had been reported, and while multiple quasi-particle level energies in ^{247}Cm and ^{249}Cf [16,17] have been extracted from decay studies, only ground-state bands have been observed to date [10]. In the present work, we report new results on rotational bands in ^{245}Pu built on the $\nu[734]9/2^-$ and $\nu[624]7/2^+$ single-neutron configurations of $j_{15/2}$ and $2g_{9/2}$ parentage, respectively, as well as new rotational bands in ^{247}Cm and ^{249}Cf built on the $\nu[622]5/2^+$ configuration of $i_{11/2}$ parentage (Fig. 1). This is the first time high-spin rotational bands that are *not* built on the $j_{15/2}$ neutron orbital have been observed in these even- Z , $N = 151$ systems and unambiguously identified from M1/E2 branching ratios.

The experimental data were obtained in three experiments using radioactive ^{244}Pu , ^{248}Cm and ^{249}Cf targets. A ^{208}Pb beam was used with the ^{244}Pu and ^{249}Cf targets, and a ^{209}Bi beam with ^{248}Cm , at energies $\sim 15\%$ above the Coulomb barrier. High-spin states were populated in $^{242-246}\text{Pu}$, $^{246-250}\text{Cm}$ and $^{248-250}\text{Cf}$ [18]. All experiments were performed at Argonne National Laboratory,

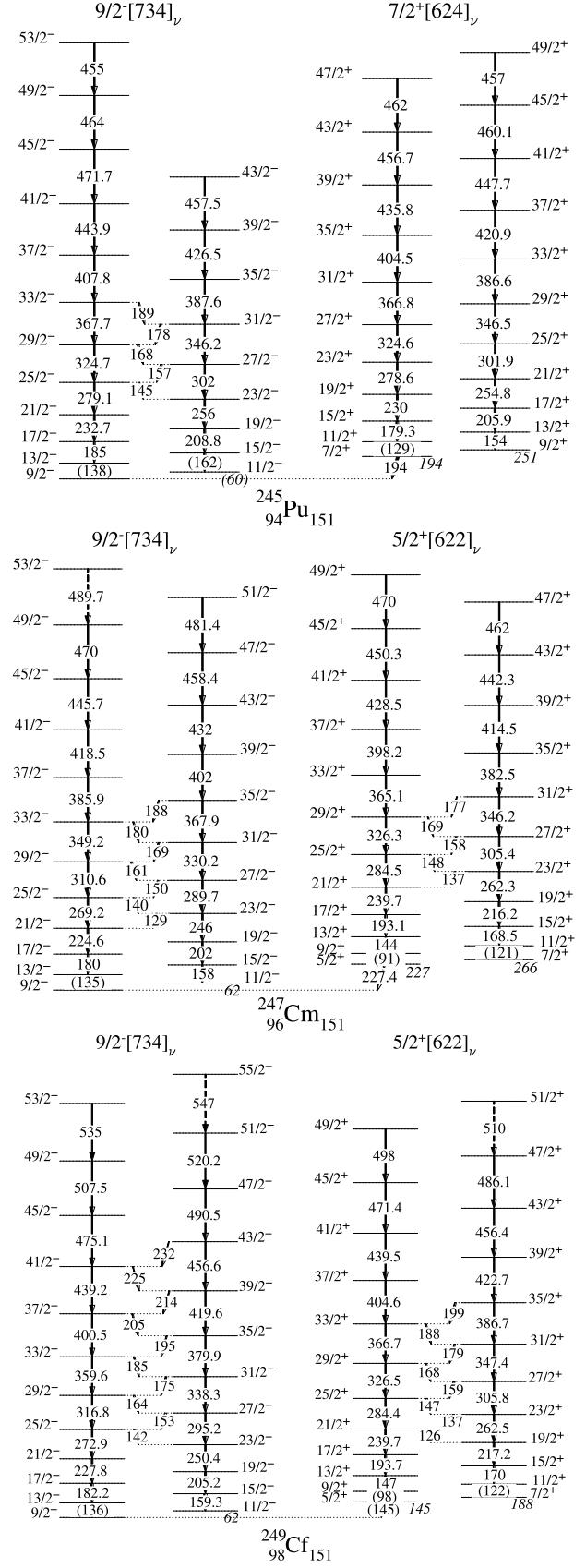


Fig. 1. Partial level schemes obtained in the present work of $N = 151$ isotones ^{245}Pu (top) and ^{247}Cm (middle) and ^{249}Cf (bottom). The ground-state bands in ^{247}Cm and ^{249}Cf were observed previously [10]. Brackets denote estimated energies for unobserved γ rays (see text).

where heavy-ion beams from the ATLAS accelerator were used to bombard radioactive targets sandwiched between layers of ^{197}Au ($\approx 150 \mu\text{g}/\text{cm}^2$ in front and $\approx 50 \text{ mg}/\text{cm}^2$ in the back) to prevent contamination of the target chamber. The γ rays emitted from both target-like and projectile-like fragments were detected by the 101 Compton-suppressed, high-purity germanium detectors of the Gammasphere spectrometer [19,20]. Events within the $\approx 1 \mu\text{s}$ standard coincidence time window of Gammasphere were recorded with a three-fold trigger condition. Details of the ^{248}Cm experiment can be found in Ref. [21]. The data were sorted into symmetric $\gamma-\gamma-\gamma$ energy coincidence cubes, using a prompt time gate of $\approx \pm 25 \text{ ns}$ as well as optimized gates on multiplicity and γ -ray sum energy, which significantly improved the channel selection. The data were analyzed subsequently using the RADWARE suite of codes [22]. Proper background subtraction of random coincidences was critical for the removal of contamination from the spectra, which were dominated by intense peaks from inelastic excitation or transfer on ^{197}Au . Cross-coincidence relationships with binary reaction partners, $x-\gamma$ coincidences, and band search techniques were used to construct the level schemes shown in Fig. 1. The ground-state bands in ^{247}Cm and ^{249}Cf were observed previously [10]. In all three nuclei, the lowest E2 γ rays in each sequence are highly converted and have not been observed. These unobserved transition energies are estimated to within $\approx 2 \text{ keV}$ using a conventional quadratic fit to an E vs $I(I+1)$ plot.

Two rotational bands, with both signature partners, were identified in ^{245}Pu . Cross-coincidence relationships with Pu x rays as well as with γ rays from the one-neutron-transfer partner ^{207}Pb corroborate the assignment of both band structures to ^{245}Pu . The stronger one is consistent with the expected properties of the ground-state band in this nucleus (Fig. 2a). Extracted $|(g_K - g_R)/Q_0|$ values from measured M1/E2 branching ratios, using values of $g_R = 0.8(Z/A) \approx 0.31$ and $Q_0 = 12 \text{ eb}$ typical for the region [23], agree well with expectations for a sequence based on a $\nu[734]9/2^-$ configuration (Fig. 3). The $15/2^-$ state, placed at an excitation energy of 222 keV in an earlier transfer reaction [15], is used to anchor both signatures of the band. The systematics of ground-state bands in $N = 151$ nuclei, built on the $\nu[734]9/2^-$ orbital, previously observed in ^{247}Cm ($Z = 96$), ^{249}Cf ($Z = 98$) [10] and ^{253}No ($Z = 102$) (an assignment in ^{253}No [11] was later revised [12]), is now extended to ^{245}Pu ($Z = 94$). A second weaker sequence is observed in ^{245}Pu (Fig. 2b), with stretched-E2 transitions very similar to those of the ground-state band, except for the lowest and highest γ -ray energies observed. Coincidence with a 194-keV transition for both signatures differentiates this weaker band from the ground-state one. A first-excited $\nu[624]7/2^+$ level with a 194-keV E1 decay to the $\nu[734]9/2^-$ ground state, as well as a $9/2^+$ state in this sequence at 251 keV, observed in the earlier study [15], is used to assign a $\nu[624]7/2^+$ configuration for the bandhead, and anchor the unfavored signature partner. No statistically significant M1 transitions were observed between the two signature partners: this is also consistent with expectations for a $\nu[624]7/2^+$ configuration.

From the $^{209}\text{Bi} + ^{248}\text{Cm}$ data, a new band structure is observed in ^{247}Cm , coincident with excitations in the one-neutron transfer reaction partner ^{210}Bi (Fig. 4a). Similarly, from the $^{208}\text{Pb} + ^{249}\text{Cf}$ data, a new band structure is also seen in ^{249}Cf , coincident with excitations of the ^{208}Pb beam (Fig. 4b). In both nuclei, previous work had established only the ground-state bands up to high spin [10]. In the new sequences, stretched-E2 transitions from both signature partners, as well as the inter-connecting M1 transitions, are observed, with multiple near-identical transition energies in the two nuclei. Extracted $|(g_K - g_R)/Q_0|$ values from M1/E2 branching ratios strongly suggest that both bands are built on known isomeric $\nu[622]5/2^+$ configurations (Fig. 2). In previous

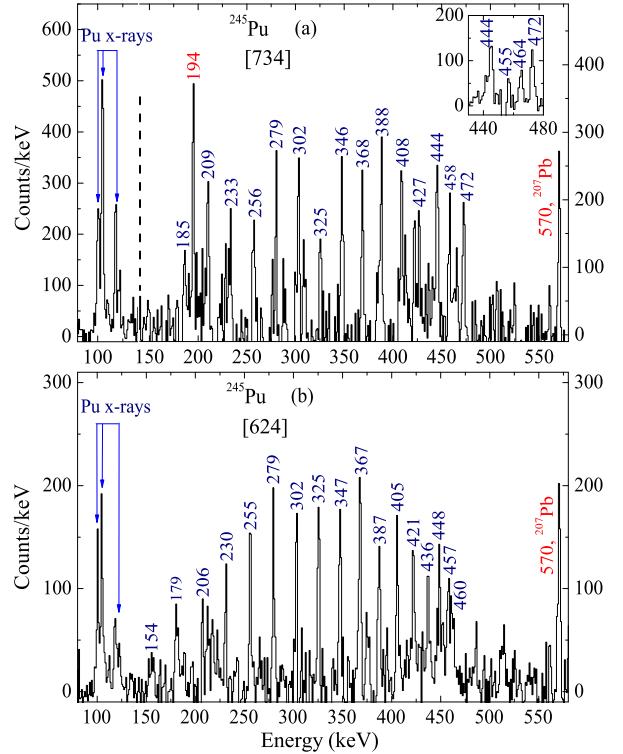


Fig. 2. (Color online.) (a) The ground-state band spectrum of ^{245}Pu , from sums of coincidence gates on E2 band transitions with Pu x rays (the inset shows the highest transitions in the favored signature). The different y-scales on the left and right correspond to the regions separated by the vertical dashed line at $\approx 140 \text{ keV}$. (b) Spectrum of the excited band in ^{245}Pu , from double gates on Pu x rays and the 194-keV transition (see Fig. 1 and text). Both bands are coincident with the 570-keV transition of the ^{207}Pb reaction partner. The 194-keV peak in (a) is a contaminant that arises from multiple near-identical transitions in the ground (a) and excited (b) bands.

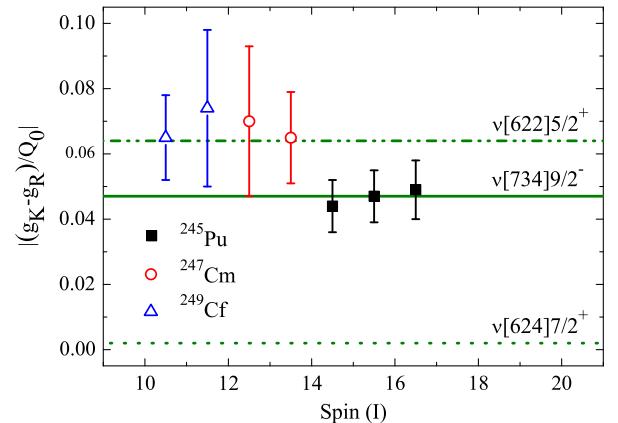


Fig. 3. (Color online.) Measured $|(g_K - g_R)/Q_0|$ ratios for the ground-state band in ^{245}Pu , and excited bands in ^{247}Cm and ^{249}Cf . Expected values are shown for the three lowest neutron configurations $\nu[734]9/2^-$, $\nu[624]7/2^+$ and $\nu[622]5/2^+$ (see text for details).

decay studies, these states were observed in ^{247}Cm and ^{249}Cf at excitation energies of 227 keV ($t_{1/2} = 26 \mu\text{s}$) [16] and 145 keV ($t_{1/2} = 45 \mu\text{s}$) [17,24], respectively. The same studies place the $9/2^+ \rightarrow 5/2^+$ transition at 91 keV in ^{247}Cm [16], and 98 keV in ^{249}Cf [17]. The $7/2^+$ states, also observed in the previous work [16,17], anchor the placement of the unfavored signature partner. The isomeric decays of the bandheads are not observed in the present prompt spectroscopy.

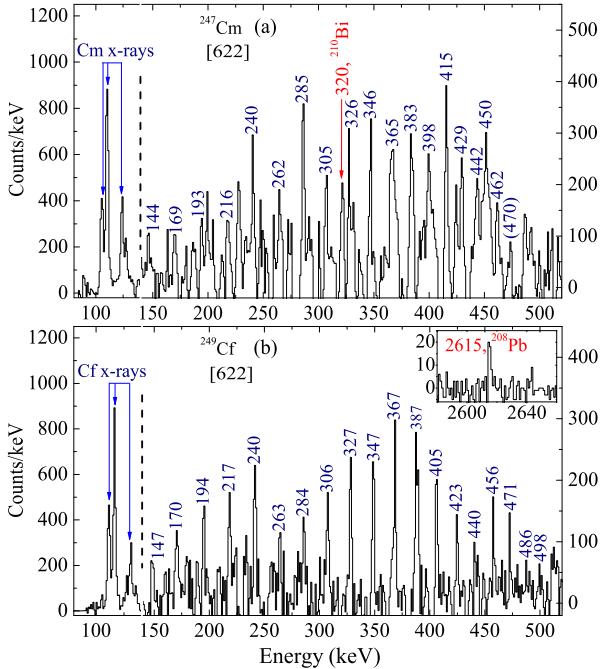


Fig. 4. (Color online.) Spectra of new bands in (a) ^{247}Cm and (b) ^{249}Cf , from sums of coincidence gates on E2 band transitions with corresponding x rays. The bands are coincident with transitions in the respective reaction partners ^{210}Bi and ^{208}Pb . The different y-scales on the left and right correspond to the regions separated by the vertical dashed line at ≈ 140 keV.

The measured energies are used to plot the angular momentum contributions along the collective rotation axis from unpaired nucleons (Fig. 5), and to compare these with their respective $N = 150$ even–even cores. Individual Harris parameters [25] are used, chosen such that zero alignment characterizes the even cores at low frequencies. For the $N = 151$ nuclei, an average of the parameter values of the even- N neighbors is adopted. The following key observations emerge from the data. In ^{245}Pu , by the time rotation frequencies of ≈ 0.2 MeV are reached, the alignment in the ground-state band, built on a $\nu j_{15/2}$ neutron configuration, saturates at $\approx 2\hbar$, while that in the $\nu[624]7/2^+$ band reaches $\approx 1\hbar$ (Fig. 5a). Both bands, built on different neutron orbitals, exhibit strong upbends at exactly the same frequency (≈ 0.23 MeV), where the even–even ^{244}Pu core also undergoes a full alignment gain of $\approx 10\hbar$, strongly suggesting a common proton pair alignment in all three cases. In ^{247}Cm , the low frequency behavior is similar to that in ^{245}Pu , and both the $\nu j_{15/2}$ ground-state band and the new $\nu[622]5/2^+$ sequence show alignment gains beyond a frequency of 0.2 MeV, with a somewhat sharper upbend for the latter, again tracking approximately the strong, but more gradual upbend observed in the even–even ^{246}Cm core (Fig. 5b). Although the full alignment is not mapped out in the Cm isotopes, the ^{246}Cm data suggest an alignment frequency of ≈ 0.24 MeV. In ^{249}Cf , the low-frequency behavior for the $\nu j_{15/2}$ ground-state band is again similar to that of the even–even ^{248}Cf core [18], where the sharp backbend and upbend observed in the lighter isotopes is replaced by a less than $\approx 1\hbar$ alignment gain up to a frequency of 0.27 MeV. The $\nu[622]5/2^+$ band exhibits a much more gradual rise compared to the saturations observed in the Pu and Cm isotones (Fig. 5c), but overtakes the flatter alignment curve of the $\nu[734]9/2^-$ ground-state band at a frequency around 0.23 MeV.

In $N = 151$ isotones, the $j_{15/2}$ neutron crossing is blocked in the ground-state band by the odd neutron in the $\nu[734]9/2^-$ state, but is allowed for the $\nu[624]7/2^+$ and $\nu[622]5/2^+$ bands, which are of $2g_{9/2}$ and $1i_{11/2}$ parentage. The $i_{13/2}$ proton crossing, how-

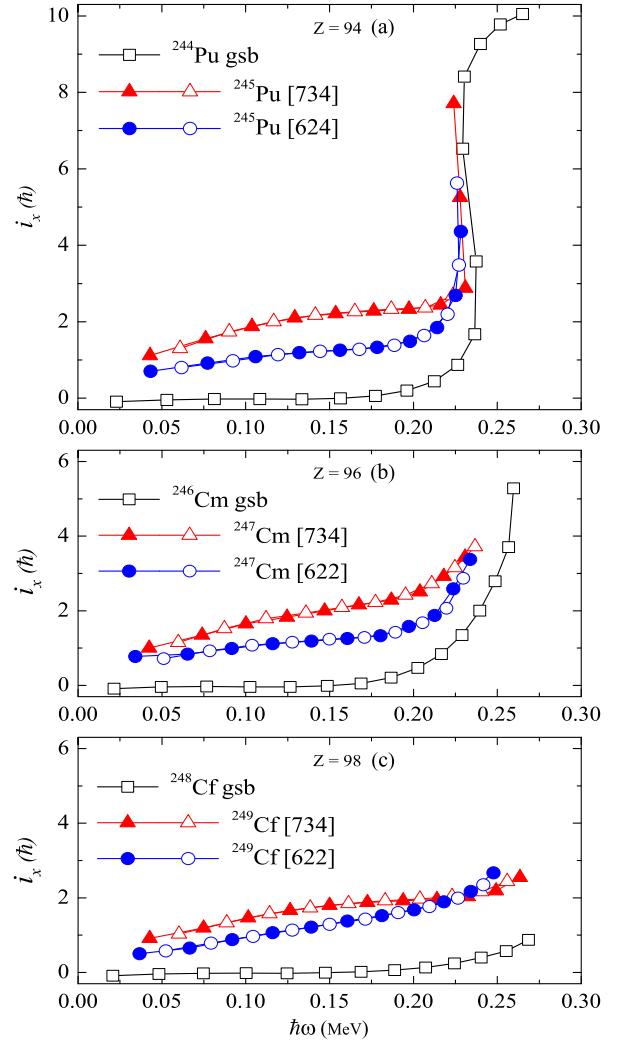


Fig. 5. (Color online.) Experimental alignment for the new bands observed in (a) ^{245}Pu , (b) ^{247}Cm and (c) ^{249}Cf , compared with ground-state bands of even–even neighbors. Level schemes for the even–even nuclei constructed from present data [18] either reproduced or extended earlier work in ^{244}Pu [8], ^{246}Cm [26], and ^{248}Cf [27]. Individual Harris parameters used are $J_0 = 65, 65, 70, 69, 72, 71$ $\hbar^2 \text{ MeV}^{-1}$ and $J_1 = 329, 302, 348, 336, 271, 337$ $\hbar^4 \text{ MeV}^{-3}$ for $^{244,245}\text{Pu}$, $^{246,247}\text{Cm}$ and $^{248,249}\text{Cf}$, respectively.

ever, is possible for all $N = 151$ bands, irrespective of the neutron configuration. Comparing the excited band behavior to that of the ground band suggests a common alignment of $i_{13/2}$ protons in ^{245}Pu and ^{247}Cm . Against the strong “background” of early proton alignments in ^{245}Pu and ^{247}Cm , it would be difficult to extract the weak signal of any underlying neutron alignment. In ^{249}Cf , however, the $\nu j_{15/2}$ trajectory continues to remain flat up to the highest observed frequency of 0.27 MeV, suggesting a delayed proton alignment. Here, the noticeable uptick of the $\nu[622]5/2^+$ band, in contrast to the flat behavior of the $\nu[734]9/2^-$ band where the $\nu j_{15/2}$ alignment is blocked, could herald the onset of $j_{15/2}$ neutron alignment contributions in ^{249}Cf at a frequency of ≈ 0.23 MeV.

To test the efficacy of available theory, we compare these new results with cranked Woods–Saxon (WS) predictions. While WS calculations reproduce single-particle level energies in $A \approx 250$ nuclei reasonably well, their prediction that $j_{15/2}$ neutrons align first, contrary to experimental observations, remains a long-standing puzzle [9]. Two sets of cranking calculations have been performed using the universal parameterization of a deformed,

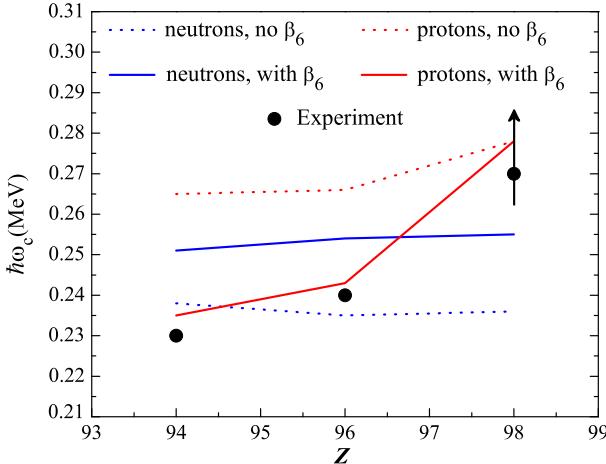


Fig. 6. (Color online.) WS predictions of alignment frequencies for neutrons (blue) and protons (red), with β_6 (solid) and without β_6 (dashed) deformation. Dots denote experimental alignment frequencies for protons.

axially-symmetric WS potential [28], one with β_2 and β_4 deformations only, and the other including the higher order β_6 deformation, which has been shown to be significant in this mass region [29]. In each case, the axial deformation parameters were fixed at the values that minimized the ground-state energy [29], and the pair gaps were fixed at the value obtained from five point odd–even mass differences [30,31].

The predicted alignment frequencies with and without β_6 deformation are compared to experimental data in Fig. 6. The inclusion of β_6 lowers the predicted proton alignment frequencies by ≈ 0.03 MeV in ^{245}Pu , while raising the neutron alignment ones by ≈ 0.015 MeV. Without β_6 , the proton alignment is predicted to occur after the neutron one for all three nuclei, in conflict with experimental observations. The inclusion of β_6 deformation effectively switches the order of the two alignments in both ^{245}Pu and ^{247}Cm , and brings the predictions in line with experimental observations. The “earlier” proton alignment would effectively mask the onset of any neutron alignment in the current data. The qualitative results are found to not be very sensitive to reductions of up to 20% in the pairing strengths. In ^{249}Cf , the inclusion of β_6 seems to have less impact on the proton alignment, and the $j_{15/2}$ neutrons are predicted to align earlier than the $i_{13/2}$ protons, which appears to be consistent with the trends observed in the present data. For a more comprehensive understanding of the high-spin behavior of ^{249}Cf , other well-known effects, such as octupole correlations [32] and increased interaction strengths predicted in WS calculations between ground and aligned bands, would need to be included. It should be noted that the inclusion of the higher order β_6 deformation has provided improved agreements with other experimental data in this region for $Z > 100$ nuclei including: high-K isomer energies in ^{254}No [33–35], and differences in rotational behaviors of ^{252}No and ^{254}No in Total Routhian Surface calculations [36]. Our present work confirms that, even in the $94 \leq Z \leq 98$ nuclei, inclusion of β_6 deformation helps to resolve the longstanding puzzle that WS calculations always predict neutrons to align before protons in this region, contrary to experimental evidence.

Recent calculations of pairing and alignments in heavy actinides using cranked relativistic Hartree–Bogoliubov theory with Lipkin–Nogami pairing [5], as well as cranked Nilsson potentials with a particle-number-conserving treatment of pairing correlations [2], have been tested against previously available rotational excitations built on ground-state band configurations in ^{247}Cm and ^{249}Cf . The theories concur that the alignment contributions in the ground-state bands come primarily from $i_{13/2}$ protons. This is expected,

as the $j_{15/2}$ neutron contributions are blocked for this configuration. It would be of interest to test the predictions of these models against these new data on excited state configurations, where the $j_{15/2}$ neutron contributions are not blocked.

In conclusion, four new rotational bands have been identified up to high spins in odd- A , $N = 151$ ^{245}Pu , ^{247}Cm and ^{249}Cf nuclei. The ground-state band in ^{245}Pu , built on the expected $\nu[734]9/2^-$ configuration, and rotational bands built on excited state configurations, $\nu[624]7/2^+$ in ^{245}Pu and $\nu[622]5/2^+$ in ^{247}Cm and ^{249}Cf , have been observed to high spins for the first time, with measured branching ratios confirming the configuration assignments. In each of the three $N = 151$ nuclei, the high-spin behavior of two configurations is mapped, where $j_{15/2}$ neutron alignments are either blocked or allowed. The (Cranked) WS model accounts for our high-spin observations, consolidating its general ability to satisfactorily explain a broad range of data. In particular, with the inclusion of β_6 deformation, it resolves a longstanding puzzle by explaining that protons align at lower frequencies compared to neutrons in ^{245}Pu and ^{247}Cm . However, the experimental elusiveness of predicted $\nu j_{15/2}$ alignments needs further investigation. Mapping complete alignments to higher rotational frequencies would provide further constraints for the competing theories that attempt a comprehensive theoretical description of the heaviest nuclei.

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