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Spectroscopy of heavy fissionable nuclei

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Abstract. Structural studies of heavy nuclei are quite challenging due to increased competition from fission, particularly at high spins. Nuclei in the actinide region exhibit a variety of interesting phenomena. Recent advances in instrumentation and analysis techniques have made feasible sensitive measurements of nuclei populated with quite low cross-sections. These include K isomers and rotational band structures in isotopes of Pu ($Z = 94$) to Rf ($Z = 104$), and octupole correlations in the Th ($Z = 90$) region. The obtained experimental data have provided insights on various aspects like moments of inertia and nucleon alignments at high spins, quasiparticle energies and evolution of quadrupole and octupole collectivity, among others. An overview of some of these results is presented.

Keywords. Fission; spectroscopy; heavy nuclei; K isomers; octupole correlations.

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1. Introduction

Nuclei in the actinide chain and beyond are prone to fission owing to considerable Coulomb repulsion, yet many isotopes are relatively stable due to quantal shell effects. The delicate balance between these two opposing effects is pivotal in determining the extent of stability of specific nuclei, and also the boundaries of the periodic chart with respect to superheavy elements. Spectroscopic studies of such nuclei offer excellent insight into the underlying shell structure and its evolution as a function of atomic number, isospin and excitation. This region is characterized by rich and diverse structure phenomena like octupole correlations in lighter actinides [1–3], well-deformed rotational structures [4–6], superdeformed fission isomers and recently discovered K isomerism [7,8].

Spectroscopy of such nuclei, particularly in-beam studies, are quite challenging due to the low cross-sections and overwhelming competition from fission. The development of new instrumentation and analysis techniques has recently enabled highly sensitive measurements for isolating quite weakly populated reaction products. In-beam, prompt spectroscopic studies have focussed on rotational properties of quadrupole-deformed nuclei, the emergence of strong octupole correlations and the possible role of even higher-order deformations. Investigations of K isomers have employed fusion–evaporation and recoil–decay tagging technique in the case of $Z \geq 100$ elements, while for $Z = 94$–98, multistep Coulomb excitation and transfer reactions with standalone large γ-detector arrays have been utilized. These studies have been performed at various laboratories across the world, most notably ANL, JYFL, LBNL, GSI and more recently at BARC–TIFR.

The existence of K isomers is revealed in several nuclei viz., ^{244}Pu ($Z = 94$), ^{246,248}Cm ($Z = 96$), ^{250}Fm ($Z = 100$), ^{252,254}No ($Z = 102$) and ^{256}Rf ($Z = 104$) [7,9–13]. Isomer lifetimes in the μs–s range have been measured, and the underlying nucleonic configurations identified. The information obtained from experiments has placed stringent constraints on theoretical models, like self-consistent mean-field approaches, which predict the properties of superheavy nuclei and the location of the next island of stability beyond ^{208}Pb.

High-spin studies of prompt rotational structures have shown that many nuclei in this region can accommodate remarkable amount of angular momentum despite their tendency to undergo fission. Moments of inertia and nucleon alignments induced by Coriolis antipairing, were investigated in a number of even–even and odd-A isotopes viz., $^{244-246}$Pu, $^{246-250}$Cm, $^{248-251}$Cf and beyond. While an improved description of several structural aspects is obtained, there is limited understanding of some features like the rotational alignment of $j_{15/2}$ neutrons and the role of higher multipole deformations.

Nuclei around the Ra–Th region show evidence of enhanced octupole strength and reflection-asymmetric shapes. Octupole correlations vary from vibration-like to the appearance of near-static octupole deformation. Nuclei with neutron number $N \approx 130$ exhibit evidence of transitional behaviour arising from the effects of shell gap at $N = 126$, weak quadrupole collectivity and enhanced octupole correlations.

Selected results highlighting some of the phenomena of interest described above are outlined in the subsequent sections.

2. High-spin rotational structures in odd-A Cm and Cf isotopes

Prompt collective structures are established in a number of doubly-even nuclei ranging from Pu and Cm to No and Rf isotopes [4,6,11,14]. Experimental investigations of odd-mass nuclei are typically more difficult, because the intensity is fragmented over multiple decay pathways. However, such studies yield complementary information on specific nucleonic configurations. Rotational structures are identified in odd-A Cm and Cf isotopes, built on some of the highest-lying neutron single-particle levels, including the $\nu[620]1/2$ orbital of $2g_{7/2}$ parentage, from above the $N = 164$ spherical subshell gap, in ^{249}Cm ($N = 153$). Further, in isotones ^{247}Cm and ^{249}Cf ($N = 151$), rotational bands built on the $\nu[734]9/2$ orbital of $j_{15/2}$ parentage are established. In all the cases, states up to $\approx 25\hbar$ are identified.

The data on prompt rotational structures and K isomers described in the subsequent sections, were obtained from several experiments performed at the Argonne National Laboratory. For the results described in this section, beams of ^{209}Bi and ^{207}Pb, with energies ranging between 1430 and 1450 MeV (about 10–15% above the Coulomb barrier) from the ATLAS superconducting linear accelerator were made incident on radioactive targets of ^{248}Cm and ^{249}Cf, respectively. The targets used had a thick ($\approx$50 mg/cm^2) Au backing. Gamma-ray coincidence data were recorded by the Gammasphere array [15,16] consisting of about 100 Compton-suppressed Ge detectors, for these experiments, and analysed using the RADWARE suite of programs [17].

Only a few low-spin levels in ^{247,249}Cm and ^{249}Cf were known from earlier work, and transitions from these states were at or below the threshold of detection in our experiments. Therefore, coincidences with known γ-rays could not be used to assign new transitions. The situation was further complicated by the presence of substantial background arising from fission and relatively long-lived decays. Techniques like x–γ and cross coincidences for element and isotope identification, in addition to band search routines, were employed, to obtain desired selectivity and enable unambiguous identification.

Excited states up to $\approx 25\hbar$ were established in isotones ^{247}Cm and ^{249}Cf ($N = 151$), and in ^{249}Cm ($N = 153$) [18]. Coincidence spectra and decay schemes for ^{249}Cf and ^{249}Cm are illustrated in figures 1 and 2. Unambiguous configuration assignments were obtained, using $(g_K - g_R)/Q_0$ values obtained from the branching ratios of $\Delta I = 1$ to $\Delta I = 2$ transitions. These were compared with the expected values for lowest-lying quasineutron configurations in ^{249}Cf and ^{249}Cm (figures 1 and 2), and it is evident that experimental

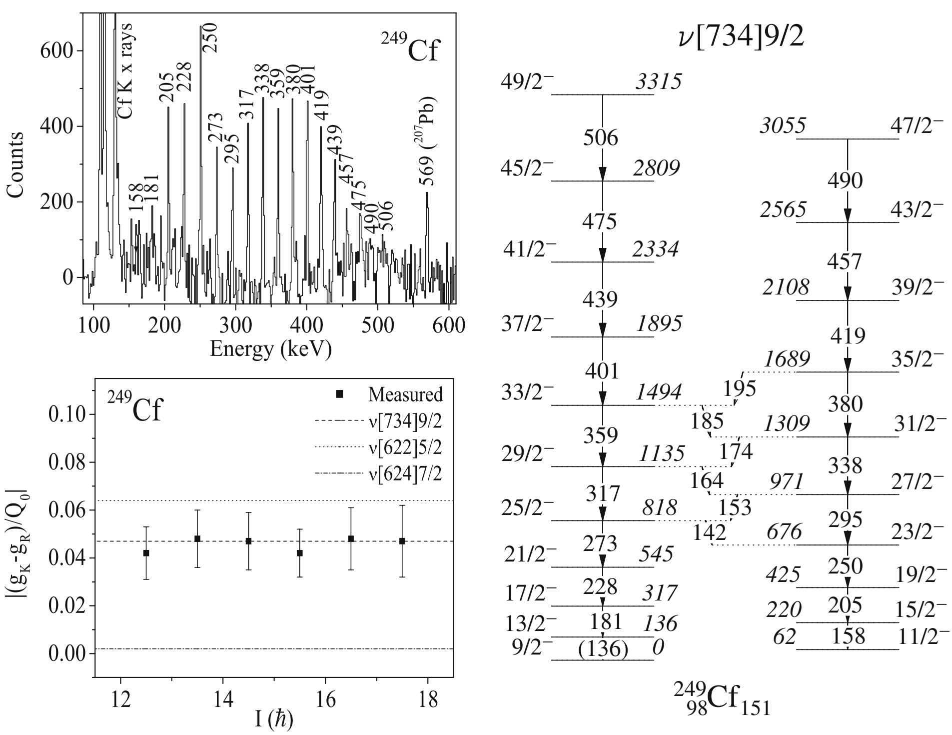

Figure 1. Gamma-ray coincidence spectrum and decay scheme illustrating the observed rotational sequences in ^{249}Cf built on the ν[734]9/2 ground state. The configuration assignment is based on $(g_K - g_R)/Q_0$ values inferred from the data.

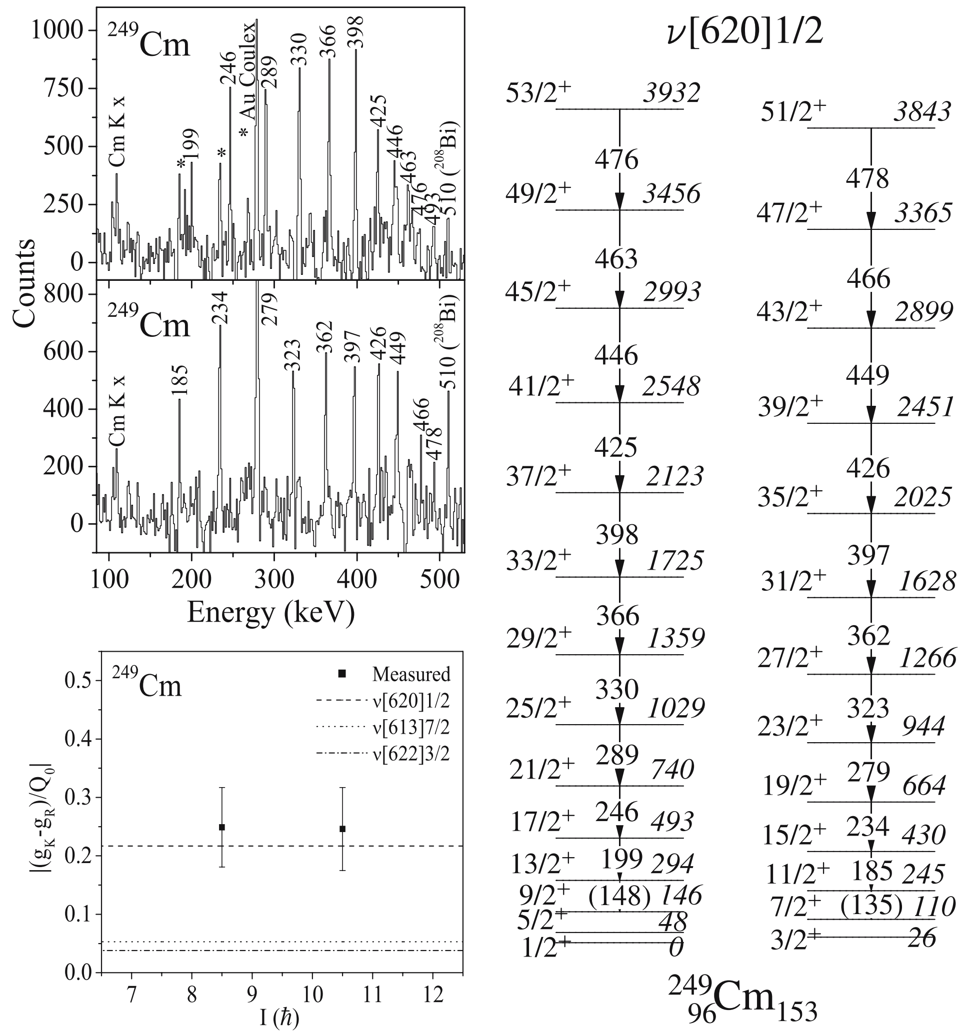

Figure 2. Gamma-ray coincidence spectra and partial decay scheme for ^{249}Cm. The observed rotational band is built on the $\nu[620]1/2$ ground state. Though $\Delta I = 1$ transitions were not observed, $(g_K - g_R)/Q_0$ values for the two states could be deduced from the observed coincidences between signature partners.

values favour $\nu[734]9/2$ and $\nu[620]1/2$ assignments in ^{249}Cf and ^{249}Cm, respectively. Rotational states built on the $\nu[734]9/2$ configuration have also been established in ^{247}Cm. With the identification of excited states up to high spins in isotones (^{247}Cm and ^{249}Cf) and isotopes (^{247,249}Cm), it is possible to explore the role of both $i_{13/2}$ proton and $j_{15/2}$ neutron alignments in a region where shell stablization effects are crucial.

Previous work in this region has revealed that the contribution to aligned angular momentum from $j_{15/2}$ neutrons is either quite small or absent. On the contrary, several instances of $i_{13/2}$ proton alignments were observed, up to the expected ($\approx 12\hbar$) contribution. Significant insight into this aspect could be obtained from this work. The $j_{15/2}$

neutron alignment is blocked for the $\nu[734]9/2$ structures in ^{247}Cm and ^{249}Cf, while the $i_{13/2}$ proton alignment is allowed. In ^{249}Cm, for the $\nu[620]1/2$ structure, both alignments are possible. A comparison of aligned angular momentum in these nuclei over the range of observed frequencies (up to $\hbar\omega \approx 0.25$ MeV) reveals a possible, small but significant contribution from $j_{15/2}$ neutrons in ^{249}Cm, as evidenced from the higher degree of alignment compared to ^{247}Cm over the observed range of frequencies (figure 3). More recent work [9], wherein rotational bands built on excited states in ^{247}Cm and ^{249}Cf are identified, seems to support this interpretation. Additionally, the possible role of higher multipole deformations has also been revealed.

The decreased alignment in ^{249}Cf ($Z = 98$) compared to its isotone ^{247}Cm ($Z = 96$) may be understood in terms of the valence protons originating from the higher-Ω, $\nu[633]7/2$ orbital in ^{249}Cf as compared to the $\nu[642]5/2$ level in ^{247}Cm. Orbitals with lower values of Ω are expected to be more strongly downsloping than those with higher Ω over a similar range of frequencies, leading to increased alignment in the former case. Woods–Saxon (WS) cranking calculations were performed using the universal parametrization [19]. These provide a satisfactory account of the observed properties, with the quasiparticle energies as a function of rotational frequency for the $\nu[734]9/2$ level in ^{249}Cf (figure 3), being a good example.

These data constitute crucial input for improving the description of nuclei within not only the microscopic–macroscopic treatment, but also the more modern density functional approaches. There are very few predictions in the literature regarding high-spin properties like moments of inertia and rotation alignment frequencies of shell-stablized, odd-*A* nuclei. With more data on odd-*A* systems expected in the near future, existing theoretical approaches and interactions can be refined further.

3. *K* isomers from Pu to Rf

The extension of the periodic chart to ever higher values of atomic number (Z) has been an ongoing quest for the past many decades. Nuclei with $Z \geq 100$ would be unstable

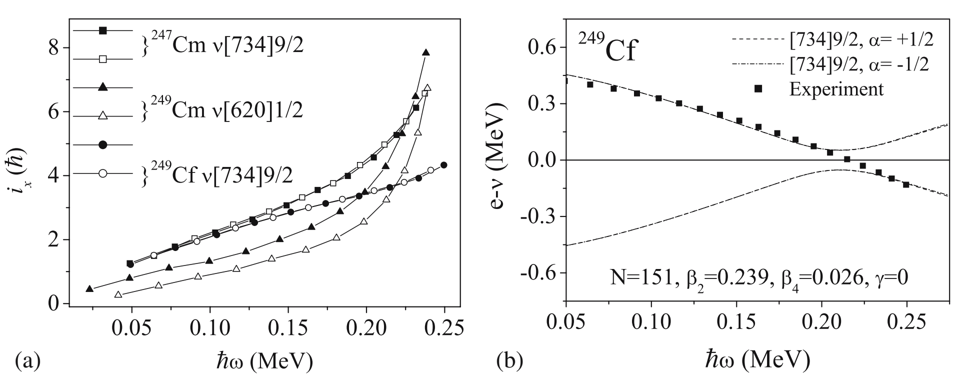

Figure 3. (**a**) Alignments for ^{247,249}Cm and ^{249}Cf and (**b**) the calculated quasiparticle energies in ^{249}Cf, as a function of rotational frequency. The experimental quasiparticle energy in (**b**) has been offset by an arbitrary amount to enable better comparison with the predictions.

against fission due to the large Coulomb repulsion were it not for the stabilizing influence of the shell-correction energy. The very existence of superheavy nuclei is therefore critically dependent on the location and magnitude of shell gaps. For most of the superheavy elements between Z = 113 and 118, only a small number of nuclei have been synthesized, which is insufficient to reveal information on single-particle energies in this region. The direction for future work aimed at reaching the superheavy 'island of stability' is not well defined because existing theoretical approaches do not agree on its precise location. The spherical magic gaps may be at $Z = 114$, 120 or 126 and $N = 172$ or 184, depending on whether microscopic–macroscopic or specific density functional approaches employing a variety of interactions provide a more accurate description. One possible way of making more reliable predictions of shell gaps in the superheavy region is to validate the predictions of these models for properties of nuclei around $Z = 100$, where spectroscopic information is recently being obtained.

K isomers are intrinsic states in deformed nuclei, and constitute an excellent probe of quasiparticle energies. As nuclei in the $A \approx 240$–250 region are characterized by substantial deformation and high-Ω valence orbitals are present near the Fermi surface, K-isomeric states are expected to be realized. Initial investigations were focussed on ^{254}No [7,8], with experiments being performed at both ANL and JYFL. Later experiments explored the nuclei ^{252}No [12], ^{250}Fm [11] and ^{256}Rf [13,20]. The nuclei were populated through fusion–evaporation reactions using ^{48}Ca beams and thin ^{208}Pb, ^{206}Pb and ^{204}HgS targets, respectively, for the No and Fm nuclei. In the case of ^{256}Rf, a ^{50}Ti beam was made incident on ^{208}Pb targets; the targets in all cases were mounted on a target wheel, the rotation of which was synchronized with the radio frequency from the particle accelerator. The fragment mass analyser (FMA) at ANL, and the gas-filled separator RITU at JYFL were employed, along with a range of focal plane detectors, including double-sided silicon strip detectors (DSSD) and clover Ge detectors. Isomer decay products, including conversion electrons and γ-rays, were recorded in these detectors.

The cross-sections for evaporation residues in the above fusion reactions ranged from $\approx$1–2 μb for ^{254}No to around 10 nb for ^{256}Rf, owing to the overwhelming competition from fission. This factor limited the amount of information that could be obtained from these experiments. Therefore, a programme was undertaken at ANL to study K isomers in Pu Cm and Cf isotopes through deep-inelastic excitation using heavy beams like ^{209}Bi and ^{207}Pb at above-barrier energies. Radioactive targets of ^{244}Pu, ^{248}Cm and ^{249}Cf were utilized. The most notable advantage in these cases, in contrast to the fusion–evaporation route, was that much higher cross-sections (at the mb level) could be realized, allowing for relatively easier and more sensitive identification of isomer decays. The Gammasphere array [15,16] was used to record γ-ray coincidence data; no auxiliary detectors were employed.

Isomers with $K^\pi = 8^-$ were identified in ^{244}Pu, ^{246}Cm, ^{252}No, and ^{254}No, with half-lives 1.75(12) s, 1.12(24) s, 109(6) ms and 266(10) ms, respectively. The isomers in $N = 150$ isotones ^{244}Pu, ^{246}Cm and ^{252}No have identical ν^2([624]7/2, [734]9/2) configurations and a similar decay pattern, with branches to both octupole vibrational structures and the ground-state rotational band. In the case of ^{254}No, a π^2([514]7/2, [624]9/2) configuration is most likely responsible. However, recently, a two-quasineutron configuration has also been suggested. There are conflicting reports [13,20] about the observation of K

isomers in ^{256}Rf, and more data are required to reach a final conclusion about the number of isomers present and their lifetimes.

Two-quasiparticle energies for K isomers were calculated [21] using the 'universal' WS potential and Lipkin–Nogami formalism for pairing, which accounts for reduced pairing due to blocking. Overall, it is found that the WS predictions are consistent with observed energies to within 200–250 keV in all cases (figure 4), including the low-lying $K^{\pi} = 3^{+}$ state in ^{254}No, indicating that both ordering and relative energies of single-particle levels are reasonably accurate. Density functional predictions using the Skyrme SLy4 and Gogny D1S interactions give a good account of the two-quasiparticle energies in several cases. However, the disagreement is as much as 1 MeV in some instances. The situation is similar to the relativistic mean field (RMF) calculations. More accurate predictions will be required to correctly pinpoint the location of superheavy island of stability, where the spherical magic gaps are expected to be ≈2 MeV in magnitude. Improved predictions are expected from the refinement of theoretical approaches and interactions using the data obtained from these measurements.

4. Octupole correlations in ^{221}Th

Proton-rich nuclei in the $Z \approx 90$, $A \approx 220$ region are characterized by enhanced octupole collectivity due to coherent contributions from Δj, $l = 3$ valence protons and neutrons. The nuclear shapes are reflection-asymmetric, and in odd-A nuclei, the so-called parity-doublet structures, with near degenerate states having identical spins and opposite parities, being present. Nuclei in the transitional region ($N \approx 130$) are expected to have similar magnitudes of quadrupole and octupole deformations (β_2, $\beta_3 \approx 0.1$) near the ground state [22,23].

High-spin states in ^{221}Th ($Z = 90$, $N = 131$) were explored through fusion–evaporation using an 86 MeV ^{16}O beam from the BARC–TIFR Pelletron in Mumbai, India, incident on a ^{208}Pb target [24]. About 80% of the compound nuclei formed underwent fission. Gamma rays from the reaction products were detected using the Indian National Gamma

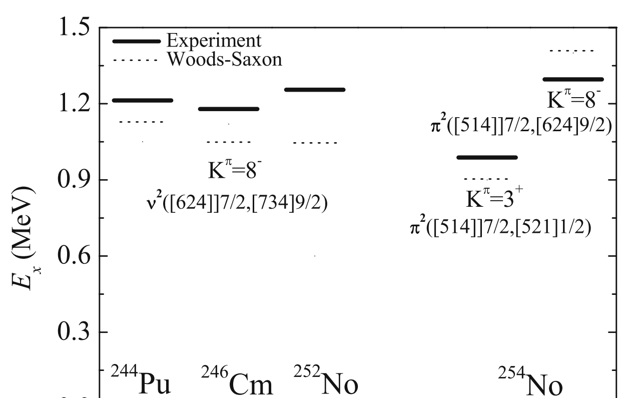

Figure 4. Excitation energies of K isomers established in Pu, Cm and No isotopes, and their comparison with predictions of WS calculations.

Array (INGA), consisting of 19 Compton-suppressed clover Ge detectors at the time of the experiment. A triggerless digital data acquisition system [25] was used to record γ–γ coincidence data (figure 5).

One sequence each with positive- and negative-parity states had been determined through previous work [26], with some transitions at intermediate spins being tentatively assigned. The yrast sequences were firmly established with new transitions up to $(39/2)\hbar$ identified, and a few transitions being modified [24]. Parity doublets are not evident in ^{221}Th. However, new non-yrast sequences of both positive and negative parities are established. Considerable contribution from a $K = 1/2$ component in the ground-state configuration is most likely responsible for the observed staggering between states belonging to sequences of the same parity, and the consequent absence of parity doublets. A similar situation is realized in the isotone ^{219}Ra. The displacement in energy (δE) between interleaved positive- and negative-parity states is close to zero in nuclei with near-static octupole deformation, a good example being ^{223}Th [26]. In ^{221}Th, δE fluctuates around zero, indicating variation in the underlying octupole deformation. Another measure of changing octupole collectivity is the intrinsic electric dipole moment (D_0), as determined from the measured intensities of $E1$ and $E2$ transitions, and its variation as a function of angular momentum (figure 6).

Kinematic moments of inertia of both positive- and negative-parity states in ^{221}Th are plotted in figure 6. While those for the negative-parity sequences remain almost constant with rotational frequency, for the positive-parity states, an increase is evident around $\hbar\omega = 0.2$ MeV. In nuclei with considerable octupole deformation, Ω is no longer a good quantum number, leading to mixing between states corresponding to different values of Ω in the axially-symmetric case. As a result, a discontinuous change in the moment of inertia due to a large contribution to aligned angular momentum from a high-j orbital is not expected in these nuclei. Further, WS cranking calculations of quasiparticle levels in ^{221}Th, including octupole deformation, indicate that the $i_{11/2}$ neutron crossing around $\hbar\omega = 0.2$ MeV is blocked, while the $i_{13/2}$ proton alignment is predicted beyond 0.25 MeV, outside the range of frequencies observed in the experiment. The increase in moment of inertia for the positive-parity states in ^{221}Th can most likely be correlated with the enhanced quadrupole collectivity at higher spins.

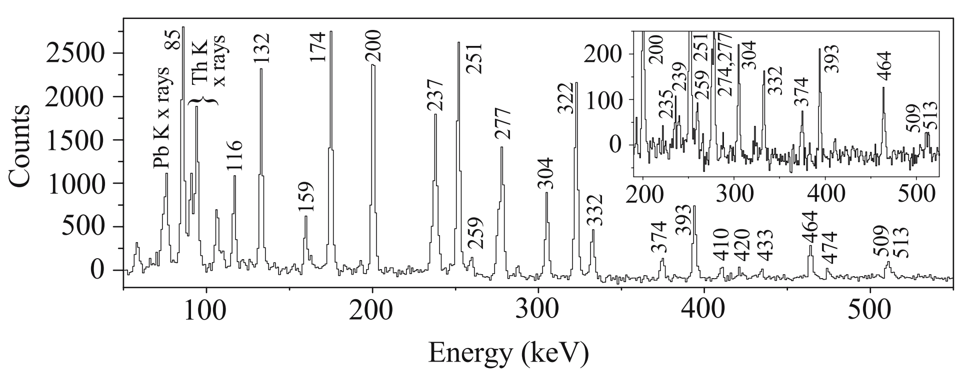

Figure 5. Gamma-ray transitions from positive- and negative-parity states in the yrast octupole structure of ^{221}Th.

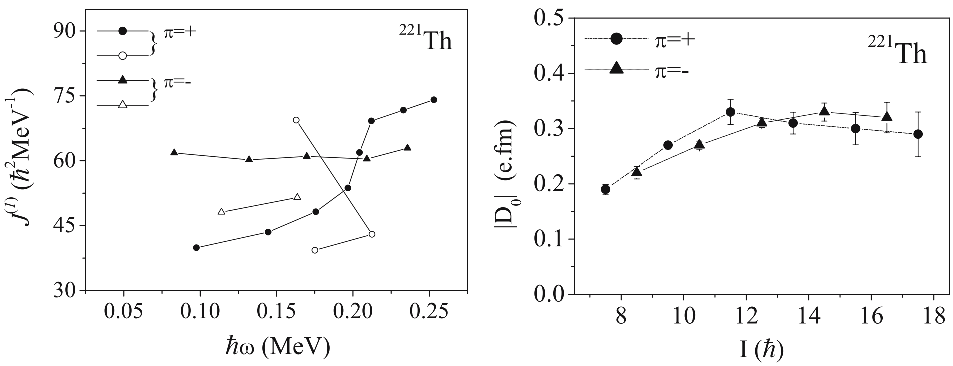

Figure 6. Kinematic moments of inertia and intrinsic electric dipole moments as a function of rotational frequency and angular momentum, respectively, in ^{221}Th. Details are given in the text.

5. Summary and outlook

Recent experimental advances have revealed a rich variety of phenomena in heavy, fissionable nuclei. It has been demonstrated that these nuclei can accommodate substantial angular momentum (25–30$\hbar$ and beyond) without undergoing fission. Moments of inertia and rotation-induced nucleon alignments at high spins were studied. Systematic studies of K isomers have provided data on two-quasiparticle energies in the highest proton and neutron shells, and provided critical validation of theoretical models which predict properties of superheavy nuclei. The dynamic variation of both octupole and quadrupole collectivity in the transitional region was observed. There is limited information on several aspects, viz., K isomers and high-spin structures built on quasiproton configurations, the role of higher-multipole deformations, the contribution to aligned angular momentum from the $j_{15/2}$ neutrons and octupole correlations beyond $Z = 90$. Experiments to explore each of these aspects in greater detail are planned in the near future.

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