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Population and decay of a $K^{\pi} = 8^{-}$ two-quasineutron isomer in ^{244}Pu

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The decay of a $K^{\pi} = 8^{-}$ isomer in ^{244}Pu and the collective band structures populating the isomer were studied using deep inelastic excitations with ^{47}Ti and ^{208}Pb beams, respectively. Precise measurements of $M1/E2$ branching ratios in the band confirm a $9/2^{-}[734]_{\nu}\otimes 7/2^{+}[624]_{\nu}$ configuration assignment for the isomer, validating the systematics of $K^{\pi} = 8^{-}$, two-quasineutron isomers observed in even-Z, $N = 150$ isotones. These isomers around the deformed shell gap at $N = 152$ provide critical benchmarks for theoretical predictions of single-particle energies in this gateway region to superheavy nuclei.

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A current frontier in nuclear structure research is the exploration of superheavy nuclei (SHN), whose stability depends almost exclusively on quantum shell effects. Macroscopic-microscopic (MM) models of SHN treat gaps in the single-particle energy spectrum of the nucleus as a shell-correction energy added to a smooth liquid-drop term, which lowers the ground-state energy and creates a sizable fission barrier. A rigorous mapping of single-particle energies and resulting shell gaps in nuclei approaching SHN is, therefore, necessary to address the physics of SHN. With direct spectroscopy not possible due to extremely low production cross sections [1], current theoretical models for SHN [2–6], with varied approaches, both MM and self-consistent, lack adequate constraints to validate their assumptions and evaluate their applicability. In the $Z \approx 100$, $A \approx 250$ region of deformed actinides, however, production cross sections range from nanobarns to tens of millibarns, and a growing body of spectroscopic data is becoming available to constrain theory [7]. While quasiparticle (*qp*) energies in odd-A nuclei are essential for mapping the single-particle levels, 2-*qp* and 4-*qp* energies in even-even nuclei near the $N = 152$ sub-shell gap provide additional insight on pair gaps in this region. In addition, axial symmetry and the availability of low-lying high-Ω orbitals near the Fermi surface lead to the presence of K-isomers [8–11], which provide clean experimental tags for extracting information on both deformed single-particle energies and pair gaps. Excitations built on broken-pair configurations also encode information on reduced pairing correlations. Finally, comparing moments of inertia of rotational bands built on high-K isomers to those built on the ground state can provide details on the mixing of wave functions associated with closely-spaced configurations.

Two- and 4-*qp* K-isomers have been observed with low cross sections via fusion-evaporation reactions in Fm ($Z = 100$) [12], No ($Z = 102$) [9, 10, 13], and Rf ($Z = 104$) [14–16] nuclei. Similar 2-*qp* K-isomers are also identified in lower-Z, but more neutron-rich, Pu ($Z = 94$) [17, 18] and Cm ($Z = 96$) [13, 19] isotopes. The energy of the 2-*qp* K-isomers over a range of Z (94 to 102) [13] provides a discriminating test of nuclear models. While MM calculations using the Woods-Saxon (WS) potential [20] reproduce experimental energies fairly well [13], predictions of self-consistent mean-field (SCMF) theories vary [9]. WS calculations consistently favor a low-lying, $K^{\pi} = 8^{-}$ neutron 2-*qp* isomer in the $N = 150$ isotones. This is supported by the measured level energy of the isomer in ^{246}Cm, ^{250}Fm, and ^{252}No [12, 13, 21], which show remarkable similarity. In addition, a proton 2-*qp* isomer was predicted and observed experimentally in ^{254}No ($N = 152$) [9, 10, 22]. However, the discrepancies between the WS and SCMF theories require further experimental evidence to unambiguously identify the 2-*qp* configurations. Such data were absent until very recently, when the bands built on top of the 2-*qp* isomer were identified in $N = 150$, ^{250}Fm [12] and ^{252}No [23]. The suggested neutron 2-*qp* configuration for the previously known isomer in ^{250}Fm [22] was identified as $K^{\pi} = 8^{-}\{9/2^{-}[734]_{\nu}\otimes 7/2^{+}[624]_{\nu}\}$ [12].

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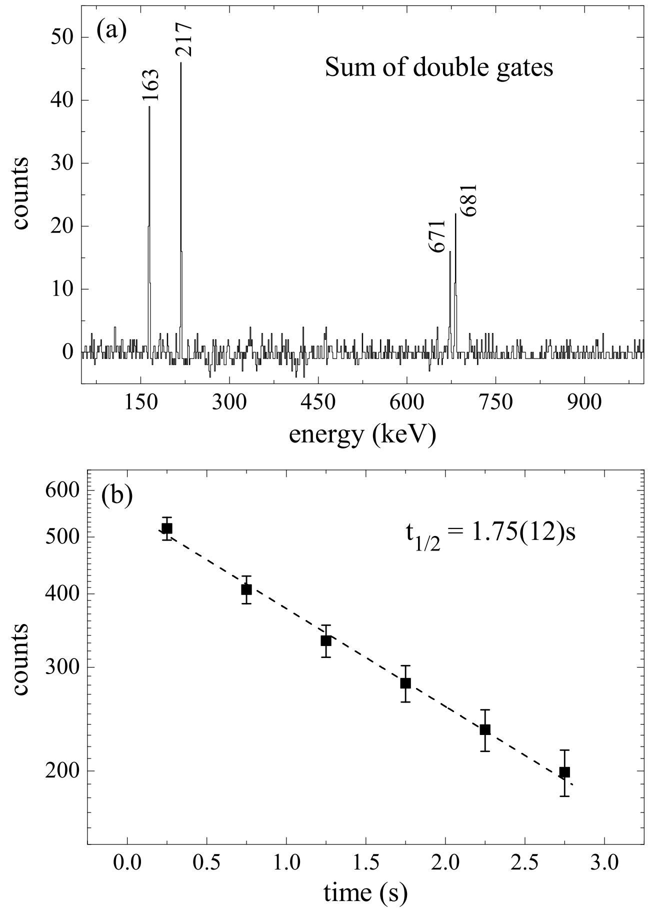

FIG. 1. (a) Sum of double gates (163-217, 163-671, 163-681, 217-671 and 217-681 keV) from events collected in the beam-off periods, showing decay transitions from the ^{244}Pu isomer; (b) the decay curve of the isomer.

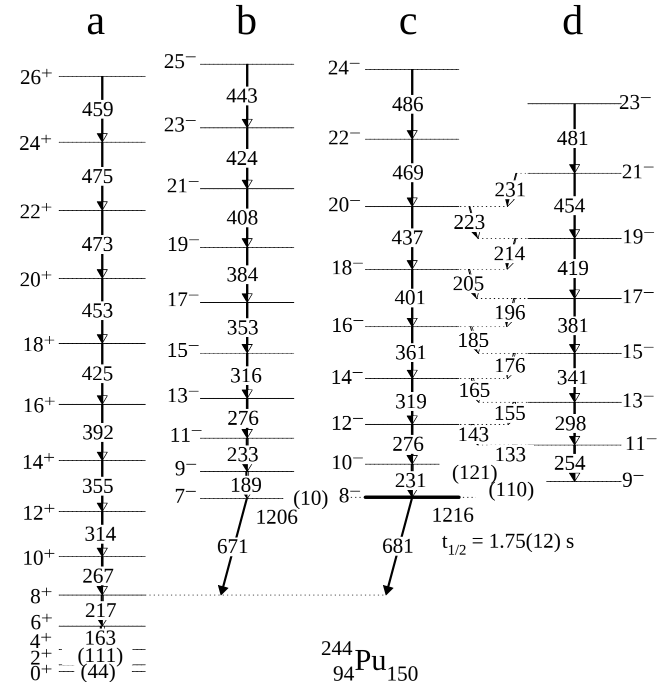

FIG. 2. Partial level scheme of ^{244}Pu, showing the ground-state band (a), the octupole vibrational band (b) and a rotational band (signature partners c,d) built on the suggested $K^\pi = 8^-$ isomer. An unobserved 10-keV transition connects the isomer to the 7^- level of the octupole band.

The present work reports on a measurement of the decay path and half-life of the $K^\pi = 8^-$ isomer in ^{244}Pu, and observation of signature partners of a rotational band built on the isomer, with strong $M1$ transitions between the signature partners. Precise measurements of $M1/E2$ branching ratios in the band provide a firm configuration assignment for the isomer, and extend the systematics of $K^\pi = 8^-$, two-quasineutron isomers observed in even-Z, $N = 150$ isotones.

Two experiments were carried out at the ATLAS facility at Argonne National Laboratory using the Gammasphere array [24, 25]. In the first, the isomeric state was populated using a 305-MeV ^{47}Ti beam incident on a ^{244}Pu target. Beam sweeping was utilized and coincident γ-γ data were collected in the beam-off periods. Since the half-lives in the neighboring $N = 150$ isotones ^{246}Cm [19] and ^{250}Fm [12] had been determined to be $\approx$ 1s, beam sweeping ranges of 1s ON/3s OFF and 3s ON/9s OFF were used, where most of the data was collected with the first time range. Based on the variation in intensity with time for the decay transitions from time-gated γ-γ coincidence matrices, the half-life of the isomeric state was deduced to be 1.75(12)s (Fig.1). In the second experiment, a 1430-MeV ^{208}Pb beam was incident on a ^{244}Pu target ($\approx$ 15% above the Coulomb barrier), where only prompt γ rays from high-spin states were recorded with a data acquisition trigger of at least three γ rays in coincidence. The prompt data were sorted into a symmetric γ-γ-γ cube and a partial level scheme of ^{244}Pu deduced (Fig.2), including the band on top of the isomer in ^{244}Pu observed up to a spin-parity of 24^-.

Four primary transitions are observed in the decay of the isomer from events collected in the beam-off intervals [Fig.1(a)], with γ-ray energies of 163, 217, 671 and 681 keV. The first two correspond to the $6^+ \to 4^+$ and $8^+ \to 6^+$ transitions, respectively, in the ground-state rotational band of ^{244}Pu (Fig.2) [26]. The $4^+ \to 2^+$ and $2^+ \to 0^+$ transitions are not observed, and their accepted values from the evaluated database [27] are used in the level scheme. The states at E_x = 1206 keV and 1216 keV had been observed from β decay [17] and Coulomb excitation [26] studies, however the isomeric character of the 1216-keV state had not been recognized. The 1206-keV and 1216-keV states have tentative spin assignments of 7 and 8, respectively, in the evaluated database [27], and are connected to the 8^+ state in the ground-state band by 671- and 681-keV transitions, respectively. The presence of an unobserved 10-keV transition between the isomeric $K^\pi = 8^-$ state and the $J^\pi = 7^-$ state in the octupole vibrational band is inferred from the data.

For the prompt data, coincidence relationships with

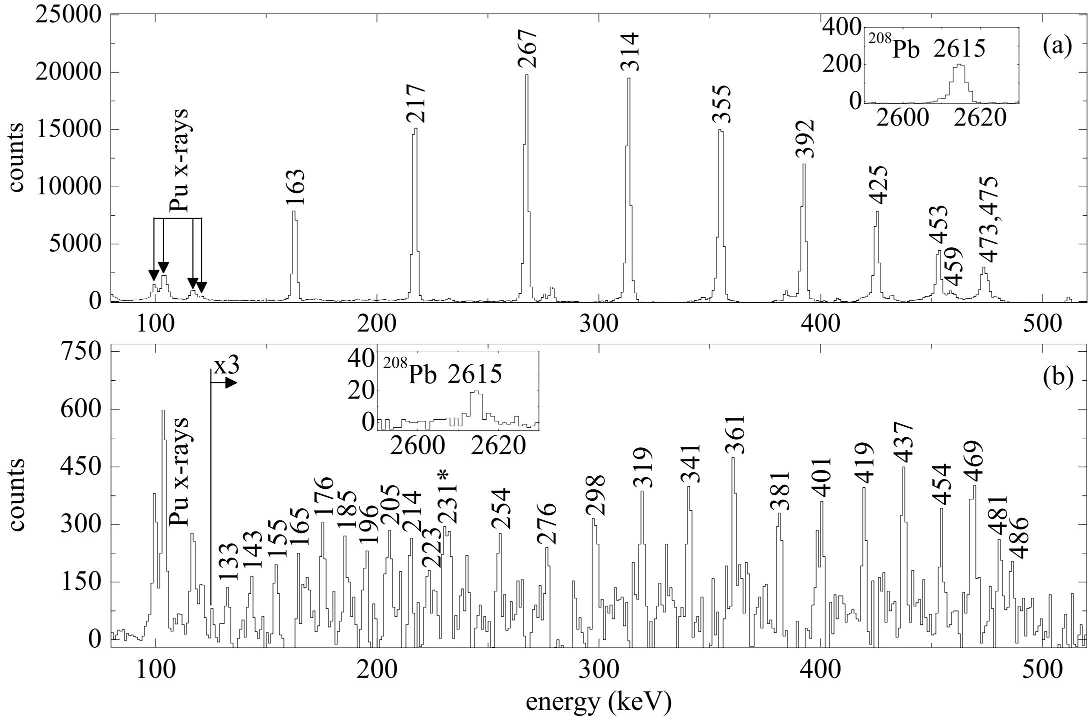

FIG. 3. (a) Spectrum in coincidence with 163- to 473-keV ground-state-band transitions in ^{244}Pu and Pu x rays. (b) Spectrum in coincidence with Pu x rays and selected gates of $M1$ and $E2$ transitions in the band built on the isomer. The 231-keV peak, indicated with an asterisk, has contributions from both an $M1$ and an $E2$ transition in the band. Insets in both panels show the coincident 2615-keV transition from the first excited state in ^{208}Pb (reaction partner).

Pu x rays as well as with the 2615-keV transition in ^{208}Pb, the binary reaction partner of ^{244}Pu for the inelastic channel, were utilized to isolate transitions in ^{244}Pu. In addition to the strongly populated ground-state band [Fig.3(a)], weak members of the band built on the $K^\pi = 8^-$ isomer were extracted [Fig. 3(b)]. Although the long half-life of the 8^- isomer precludes observation of direct coincidence relationships between the feeding and decay gamma rays, the well-tested "cross-coincidence" technique with the binary reaction partner places the band firmly in ^{244}Pu. An additional way to verify the bandhead spin assignment is to extract $|(g_K - g_R)/Q_0|$ values from in-band $M1/E2$ branching ratios (where g_K is the nucleon g factor, g_R is the rotational g factor, and Q_0 is the intrinsic quadrupole moment), and compare them with expected values for specific configurations [28].

Experimentally extracted $|(g_K - g_R)/Q_0|$ values, using $Q_0 = 12$ eb [29] and an attenuation of 0.6 for the intrinsic spin g factors, are compared with expectations for three different low-lying 2-*qp* configurations (Fig.4). In the literature, g_R values used in this region vary, from 0.31 for the odd-A Cm and Cf isotopes [29], to $g_R = Z/A = 0.4$ for ^{250}Fm [12]. However, previous measurements using transient magnetic fields [30] have shown that the value of g_R is $\approx 0.8{*}Z/A$ ($\sim$0.31) for ^{232}Th and ^{238}U, in agreement with the choice in Ref.[29]. Although similar measurements are not available for heavier actinide nuclei, a range of values for g_R have been explored,

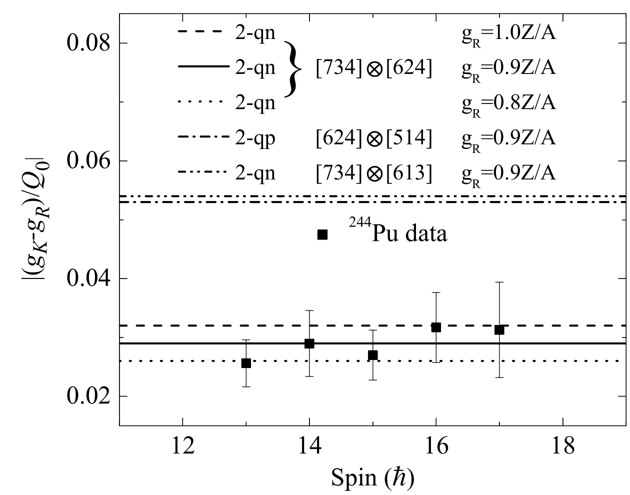

FIG. 4. Measured $|(g_K - g_R)/Q_0|$ for transitions in the band built on the isomer, clearly favoring the $9/2^-[734]_\nu \otimes 7/2^+[624]_\nu$ configuration (see text). Expected values for other low-lying $9/2^+[624]_\pi \otimes 7/2^-[514]_\pi$ and $9/2^-[734]_\nu \otimes 7/2^+[613]_\nu$ configurations are shown as well.

which do not affect the conclusions. The data unambiguously corroborate the assignment of a two-quasineutron $9/2^-[734]_\nu \otimes 7/2^+[624]_\nu$ configuration for the bandhead.

The similarity in the excitation energy of the $K^\pi = 8^-$ isomeric state for $N = 150$ isotones ranging from Pu

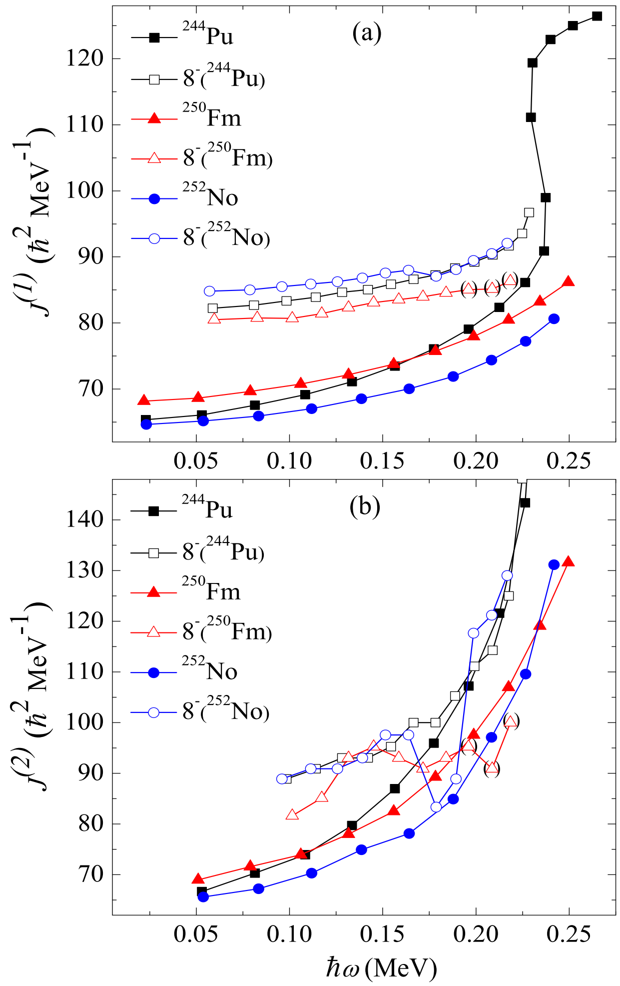

FIG. 5. (Color online) (a) Kinematic and (b) dynamic moments of inertia for the ground-state (solid symbols) and isomeric (open symbols) bands in $N = 150$ nuclei as a function of rotational frequency $\hbar\omega$, calculated as $E_\gamma/[\sqrt{(I+1)(I+2)-K^2} - \sqrt{(I-1)I-K^2}]$, where the transition is between states with $I_i = I+1$ and $I_f = I-1$, and K is the bandhead spin. $J^{(1)} = (2I+1)/E_\gamma$ and $J^{(2)} = 4/\Delta E_\gamma$.

(Z = 94) to No (Z = 102) [13] is a clear indication of a pure neutron contribution. The goodness of the K-quantum number can also be examined via the reduced hindrance factor of the isomer decays, defined as $f_\nu = (\mathrm{T}^\gamma_{1/2}/\mathrm{T}^W_{1/2})^{1/\nu}$, where $\mathrm{T}^\gamma_{1/2}$ is the partial γ-ray half-life and $\mathrm{T}^W_{1/2}$ the corresponding Weisskopf single-particle estimate. The degree of forbiddenness ν is defined as ΔK - λ , where λ is the transition multipolarity. From the measured intensities of the decay transitions, the reduced hindrance factor for the $\Delta K = 8$, $E1$ decay to the 8^+ member of the ground-state band of ^{244}Pu is determined to be $f_\nu = 177$, in line with f_ν values for the $\Delta K = 8$, $E1$ decays for the $K^\pi = 8^-$ isomers in $N = 150$ isotones ^{246}Cm (f_ν = 166) [11, 19], ^{250}Fm (f_ν = 213) [12], and ^{252}No (f_ν = 178) [23]. A two-quasiproton $7/2^+[633]_\pi \otimes 7/2^-[514]_\pi$ configuration had also been originally suggested for the ^{250}Fm isomer, resulting in a 7^- state [31], but was discarded based on measured branching ratios [12]. For ^{244}Pu, an 8^- assignment was based on similar log ft values for both ^{244}Np $\rightarrow$ ^{244}Pu and ^{246}Am $\rightarrow$ ^{246}Cm β^- decays, where it was proposed that both decays proceed via the transition $7^-\{5/2^+[642]_\pi \otimes 9/2^-[734]_\nu\} \rightarrow 8^-\{7/2^+[624]_\nu \otimes 9/2^-[734]_\nu\}$[13]. The large values of reduced hindrance signify that K is a good quantum number for these axially symmetric, deformed actinide nuclei.

The kinematic ($J^{(1)}$) and dynamic ($J^{(2)}$) moments of inertia (MOI) for ^{244}Pu are compared with those of the neighboring isotones ^{250}Fm and ^{252}No (Fig.5). At low $\hbar\omega$ values, $J^{(1)}$ moments of the isomeric bands are $\approx$ 20% higher than those of the ground-state bands, primarily due to the "alignment" effect of two high-j orbitals involved in the configuration. The $J^{(1)}$ moments of the isomeric band in ^{244}Pu parallels that of the ground-state band, and exhibits a slight upbend at essentially the frequency where the latter undergoes a full alignment [Fig.5(a)]. This is the only one out of the three isotones where the band is observed to high enough frequencies to observe this behavior. Since the yrast band alignment in ^{244}Pu is attributed to a pair of $i_{13/2}$ protons [26, 32], the isomer band is expected to mirror it, as the two unpaired quasineutrons coupling to 8^- do not block the proton alignment. For 2-qp K-isomers, a broken pair of nucleons occupies different orbitals with large projections of total angular momentum along the symmetry axis. Since pairing tends to decrease the MOIs from rigid-body values by introducing irrotational components, broken pairs also increase the MOI, as the unpaired nucleons block the scattering of pairs into the occupied orbitals and reduce neutron pairing.

While the $J^{(1)}$ moment for the isomeric band in ^{244}Pu displays a smooth behavior, the isomeric bands in ^{250}Fm and ^{252}No exhibit a slight variation in slope at $\hbar\omega \sim 0.17$ MeV. The $J^{(2)}$ plots [Fig.5(b)] enhance these features. A dip with a minimum at $\hbar\omega \sim 0.20$ MeV is visible for ^{250}Fm, and an even more pronounced one is observed in ^{252}No with a minimum at $\hbar\omega \sim 0.18$ MeV, although the last three transitions in the isomeric bands in ^{250}Fm are tentative [12]. In ^{252}No, a crossing with a side band built on a low-lying 2^- state was put forward as a possible explanation for the anomaly. A more recent theoretical study using a configuration-constrained total-Routhian-surface model suggests that the prominent dips in $J^{(2)}$ moments for both ^{250}Fm and ^{252}No are due to configuration mixing with a proton $7/2^+[633]_\pi \otimes 7/2^-[514]_\pi$ $K^\pi = 7^-$ band [33]. The smooth behavior of the $J^{(2)}$ moment for the 2-qp band in ^{244}Pu is consistent with this interpretation, as the $7/2^+[633]_\pi$ configuration moves to higher excitation energies with decreasing Z and would thus be less likely to mix with the two-quasineutron isomer configuration. It would be instructive to corroborate this interpretation with a full calculation within the model of Ref.[33].

Various theoretical approaches are currently focused on benchmarking their predictive power for excitations in the $N \geq 150$ region. As is obvious from the discussions above, multiple combinations of valence neutron and proton orbitals can combine to form 8^- isomers in trans-plutonium nuclei. Multi-qp calculations with a Woods-Saxon potential and the Lipkin-Nogami pairing formalism, for example, do remarkably well in reproducing level energies in both $N = 150$ and $N = 152$ isotones around the $N = 152$ deformed shell gap [9, 15, 19, 34, 35]. In particular, for the 8^- states in even-even $94 \leq Z \leq 102$ nuclei, where the experimental level energies stay constant to ≈ 75 keV, Woods-Saxon calculations reproduce this constancy, lying on average ≈ 150 keV below the experimental data points, and well within the model uncertainties of ≈ 300 keV [13]. SCMF calculations using the Hartree-Fock-Bogoliubov (HFB) method and a D1S Gogny interaction [5] reproduce the 8^- energies of the $N = 150$ isotones [15], but fail to reproduce the proton 2-*qp*, $K^\pi = 8^-$ energy in ^{254}No [9]. SCMF calculations with the SLy4 parameterization of the Skyrme interaction predict the neutron sub-shell gap at $N = 150$ [36], in contradiction with experiment. The present data, together with the systematics of the region, suggest the need for refining interactions in SCMF theories.

In conclusion, the decay paths and half-life of a new isomer in ^{244}Pu are measured and rotational bands built on the long-lived state have been observed up to high spins. Extracted g_K values from the measured $M1/E2$ branching ratios indicate a $K^\pi = 8^-\{9/2^-[734]_\nu \otimes 7/2^+[624]_\nu\}$ configuration for the bandhead. While 8^- isomers with this suggested two-quasineutron configuration have been observed in a series of $N = 150$ isotones ranging from $Z = 94$ to $Z = 102$, rotational bands built on the isomer have only been previously reported in fusion-evaporation studies of ^{250}Fm and ^{252}No, where confidence in configuration assignments was limited due to poor statistics as well as possible admixtures. The present work confirms a pure two-quasineutron configuration in ^{244}Pu with significantly better statistics using the higher cross-sections for inelastic excitation. Multi-*qp* calculations using a Woods-Saxon potential and Lipkin-Nogami pairing favor the experimental configuration assignment. The extended and improved experimental systematics for 2-*qp* isomers and rotational band structures built on them for $N = 150$ isotones provide important benchmarks for theoretical predictions of single-particle energies in this mass region, that would eventually extrapolate to predicting properties of superheavy nuclei.

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