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Reaching the prolate-oblate boundary at N=116 via first fragmentation of a ^{198}Pt beam: Sharp transition to triaxiality in ^{189}Ta

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High-spin isomers in very-neutron-rich $A\approx190$ Hf-Ta-W nuclei were populated via the pioneering fragmentation of a ^{198}Pt primary beam at the National Superconducting Cyclotron Laboratory. The nuclei were implanted in a Si detector stack surrounded by the Gamma-Ray Energy Tracking In-beam Nuclear Array (GRETINA) to detect delayed γ rays, providing first level schemes using γ-γ coincidence data from isomeric decays in this previously inaccessible region of the nuclear chart. A sudden transition to a strong triaxial shape is observed in the very-neutron-rich ^{189}Ta (N=116) nucleus from axially-prolate shapes in lighter Ta isotopes, providing a critical experimental benchmark for competing theoretical predictions of nuclear shape evolution.

The collective model has been a cornerstone of nuclear structure physics from the time it was invoked to explain rotational and vibrational dynamics of deformed nuclei [1]. How shapes of heavy nuclei evolve, from spherical at a shell closure to strongly deformed at mid-shell and then back to sphericity, continues to be a research frontier in the field. Varied theoretical approaches and models have emerged to probe the rich interplay of single-particle and collective degrees of freedom in these strongly-correlated many-body systems [2]. In axially deformed nuclei, the competition between prolate (rugby ball shape) and oblate (discus shape) potential energy minima is strongly influenced by nucleons in shape-driving valence orbitals. To understand the evolution of shape minima as a function of nucleon number and whether the transitions between shapes are gradual or abrupt, one needs spectroscopic data, ideally spanning a complete shell. The highest proton and neutron shells fully identified experimentally, *viz.* $50\leq Z\leq 82$ and $82\leq N\leq 126$, provide the most expansive environment for this exploration. Nuclei near the middle of these Z and N shells, such as the Hf-Ta-W ($72\leq Z\leq 74$) isotopes close to stability, exhibit robust axially-prolate shapes in their ground and excited states up to high angular momenta. With increasing Z from the mid-shell, their ground states evolve from prolate to spherical as the Z=82 shell closure is approached, with experimental data available to constrain theory [3]. With increasing N, however, little high-spin data exist beyond the middle of the neutron shell, a region where theories have been predicting exciting shape dynamics for over four decades [4–11]. This spectroscopic study with emerging techniques tests these long-standing and varied predictions of prolate-oblate shape transitions in neutron-rich nuclei in this region.

Experiments using inelastic and multi-nucleon transfer reactions have made modest inroads into neutron-rich territory [12–14], but have not been able to reach the nuclei where sudden shape changes are predicted. New reaction mechanisms and experimental advances are necessary. We used the fragmentation of a ^{198}Pt beam for the first time, to produce nuclei which were not previously accessible by any other reaction mechanism [15, 16]. In addition to discovering new isotopes, these reactions create very-neutron-rich nuclei in metastable states at high angular momenta. Coupling precise particle-identification with advanced γ-ray detection, the γ-

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decay pathways of these "high-spin isomers" allow nuclear structure information to be extracted from their excited levels, significantly expanding the scope of such studies very far from stability in the $A{\approx}190$ region. Earlier fragmentation studies using ^{208}Pb beams at 1 GeV/nucleon had been able to populate isomers in $73{\leq}Z{\leq}81$ nuclei, but with insufficient γ-γ coincidence statistics for constructing new level schemes, especially for the lowest-Z Ta and W isotopes [17–19]. In this work, the first level structure deduced for the very-neutron-rich ^{189}Ta (N=116) nucleus reveals a sharp transition to a strongly triaxial shape, a radical departure from the axially-prolate shapes observed in the lighter isotopes closer to stability. Reaching this clear marker for a prolate-oblate shape boundary as a function of N provides a rigorous test for theories extending out to this neutron-rich terra incognita.

High-spin isomers in prolate Hf-Ta-W nuclei near the stability line arise from the approximate conservation of the K quantum number, defined as the projection of the total angular momentum of the nucleus onto the symmetry axis. Such "traps" were first observed in Hf isotopes using fusion-evaporation reactions [20, 21]. Early theories using a cranked modified oscillator potential predicted robust prolate K-isomers across the stable Hf isotopic chain [22], where appropriate valence orbitals were available for both neutrons and protons to form low-lying high-K configurations. At the same time, oblate shape competition at high angular momenta was predicted in the heaviest stable ^{180}Hf isotope by self-consistent Hartree-Fock-Bogoliubov (HFB) theories [4], and at even lower excitation energies in neutron-rich ^{182,186}Hf by configuration-constrained calculations using a Woods-Saxon potential including pairing [5]. Constrained HFB models, which have explored ground-state deformations in more neutron-rich nuclei using Gogny and Skyrme interactions [7, 8], predict a rapid prolate-to-oblate transition through triaxial shapes at N=116 for both Hf and W. Other macroscopic-microscopic calculations such as the finite range droplet model predict similar sudden shape changes, but at N=120 [10]. Recent configuration-constrained calculations for Hf predict a dissolving of the prolate-oblate barrier in the ground state at N=118 [11]. All of these predictions have remained untested to date. This work presents first experimental evidence of reaching the prolate-oblate shape boundary in this neutron-rich terrain through the observation of a sudden transition to a strong triaxial shape for ^{189}Ta (Z=73, N=116), which has 8 neutrons more than the stable ^{181}Ta isotope.

Details of the experimental setup and particle identification techniques were discussed in a recent publication of our collaboration [15], which focused on the discovery of new isotopes. Therefore, only key information is summarized here. An 85-MeV/nucleon ^{198}Pt primary beam was incident on Ni and Be production targets at the National Superconducting Cyclotron Laboratory (NSCL) at Michigan State University [23]. The high-Z fragmentation products emerge with a variety of charge states and were momentum-analyzed by the A1900 fragment separator [24]. Different settings were used to transport ^{186}Hf, ^{189}Hf and ^{192}Hf isotopes at different phases of the experiment. A thin plastic scintillator at the A1900 focal plane provided the start signal for time-of-flight

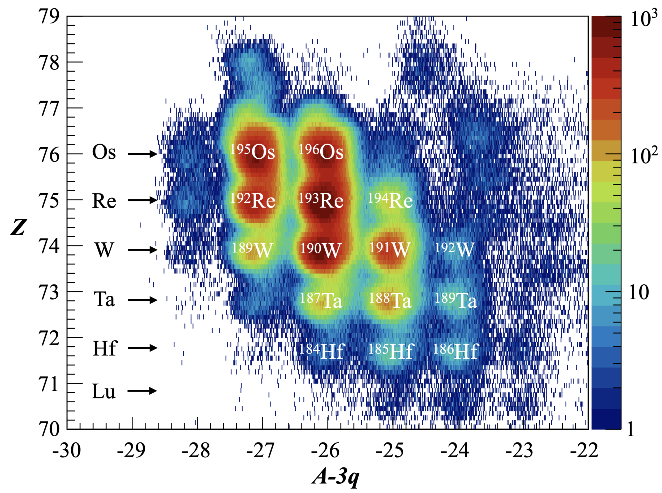

FIG. 1. Particle identification plot for fragments produced with a Be target and A1900 optimized for the ^{186}Hf setting, gated on $Z - q = 2$ (He-like) charge states.

(ToF) measurements for the fragments of interest, which were transported through the S800 analysis beamline and stopped in a stack of Si detectors at the S800 target location [15]. The Si-stack was positioned at the center of the GRETINA array [25, 26], which recorded γ decays of fragments implanted in isomeric states. Isotopic identification was achieved using the Bρ-ΔE-TKE-ToF parameter set to obtain A, Z and q (charge) on an event-by-event basis [15, 27]. The ions from the separator were mainly distributed in q = Z-2 (He-like) or q = Z-3 (Li-like) charge states. The charge-state selection is required to produce a well-resolved Z vs A-3q particle identification (PID) plot (Fig. 1). Each grouping in the PID plot corresponds to a specific isotope implanted either in its ground state or in an excited isomeric state. A range of isomers was identified in the neutron-rich $72{\leq}Z{\leq}{<}76$ region, with many observed for the first time. The ≈400 ns flight time of the fragments to the implantation detectors and an average implantation interval of ≈10 ms defined the approximate half-life limits for isomer identification. A compilation of half-lives and γ rays from the decay of all identified isomers will be presented in a forthcoming publication [28].

Constructing a decay scheme and extracting new physics for a specific isotope in this previously inaccessible neutron-rich region depends critically on available statistics and the power of GRETINA to provide implant-correlated γ-γ coincidence data. We focus on one such example, where we deduce a sudden triaxial shape distortion in the N=116 nucleus ^{189}Ta compared to the predominantly prolate shapes of its less neutron-rich odd-A neighbors. Extracting this physics is only made possible by following the γ decay of the implants from their excited isomeric states and by identifying sequences of collective excitations in the process. The spectroscopic data and logic for constructing the first level scheme for ^{189}Ta is presented below, followed by a discussion of the new physics that emerges for this rare neutron-rich isotope.

A total of approximately 25 000 ^{189}Ta isotopes were implanted, with 10-15% in an isomeric state at high angular momentum, which were then correlated with delayed γ-ray tran-

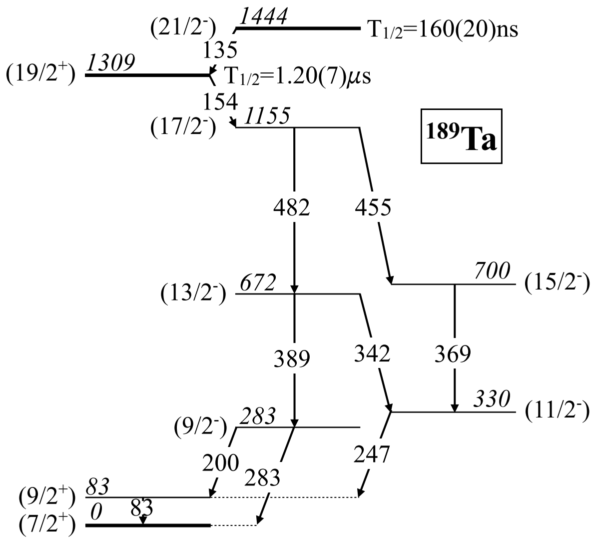

FIG. 2. Level scheme for ^{189}Ta deduced in the present work showing the decay of two isomers via collective structures, with γ-ray energies in keV.

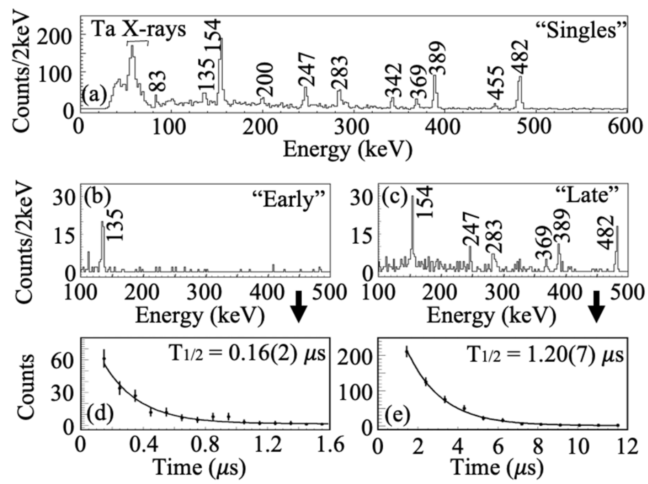

FIG. 3. (a) "Singles" spectrum of ^{189}Ta γ rays emitted between 0.3 and 12.3 μs following implant; (b),(c) Time-correlated spectra, with conditions of 0.2 to 1 μs (early) and 1 to 7.5 μs (late) following implant; (d),(e) γ-gated half-life plots for the 1444-keV and 1309-keV isomers.

sitions from the isomer decay. While many of these γ rays in ^{189}Ta had been reported in earlier fragmentation studies using a ^{208}Pb primary beam [17–19], the statistics had been insufficient for deducing a decay scheme. In the present work, more implants, together with the γ-γ coincidence efficiency of GRETINA, enabled the observation of additional transitions. All observed γ rays were placed in a level scheme for ^{189}Ta (Fig. 2). Two isomers that had been observed in the prior studies [17–19] were identified and their half-lives measured with higher statistics.

To identify isomeric decays, γ ray emission times following an implant were inspected. Fig. 3(a) shows the "singles" spectrum of γ rays emitted between 0.3 and 12.3 μs following a ^{189}Ta implant. A time window closer to the implants ($\Delta t \leq 1$ μs) showed the 135-keV γ ray to have a significantly shorter half-life compared to a common half-life for all the other γ rays. "Early-late" time correlations, shown in Fig. 3 (b),(c), revealed that the shorter-lived isomer feeds the longer-lived isomer with a single 135-keV γ ray, and all the other γ rays originate from the decay of the lower isomer. Half-lives were extracted by fitting appropriate energy-gated time spectra shown in Fig. 3 (d),(e). The rest of the ^{189}Ta level scheme was deduced from γ-γ coincidence spectra shown in Fig. 4. A single exponential fit to the time spectrum of the 135-keV γ ray yields a half-life of 0.16(2) μs for the upper isomer placed at an excitation energy of 1444 keV. For the lower isomer placed at an excitation energy of 1309 keV, a single-exponential fit to the summed time spectrum of strong γ rays (beyond ten half-lives of the upper isomer) yields a half-life of 1.20(7) μs. These results are consistent with the most recently published work where the same ordering of the isomers is proposed, with the half-lives quoted as $\approx$200 ns and $\approx$1.2 μs [19]. Table I lists the properties of all the decay γ rays from the lower isomer. Since intensity balance must be satisfied for each level, this allows the assignment of transition multipolarities by constraining the choices of internal conversion coefficients [29] for each transition.

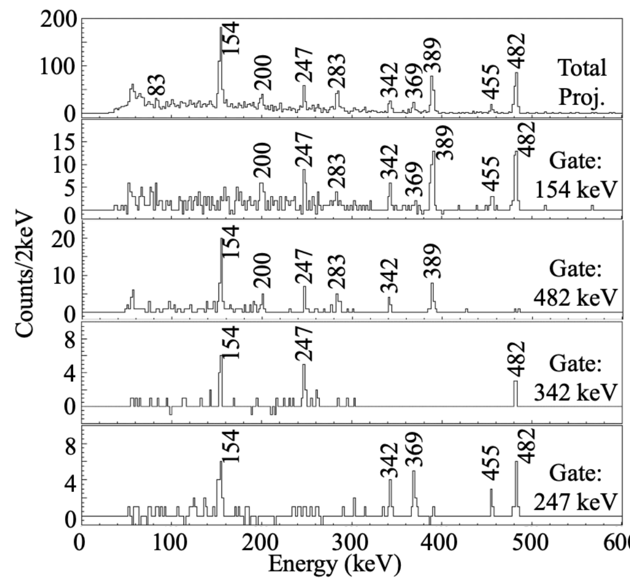

FIG. 4. Spectra projected from a 2D γ-γ coincidence matrix, for decay γ rays emitted between 0.2 μs and 7.7 μs following a ^{189}Ta implant and within 40 ns of each other.

All lighter odd-A Ta nuclei exhibit deformed rotational excitations, including the nearest ^{187}Ta neighbor [30]. Our ^{189}Ta level scheme is consistent with the stronger 389-482-keV cascade being of $E2$ multipolarity, the energies of which follow the moment-of-inertia systematics of the lighter odd-A neighbors. With a ground-state spin-parity assignment of I^{π}=7/2$^+$, where the unpaired proton occupies the valence 7/2$^+$[404] Nilsson orbital as in lighter odd-A $^{177-187}$Ta, other spin-parity assignments follow from transition multipolarities deduced via intensity balance (Table I). These assignments are robust, especially for the rotational structures that are our pri-

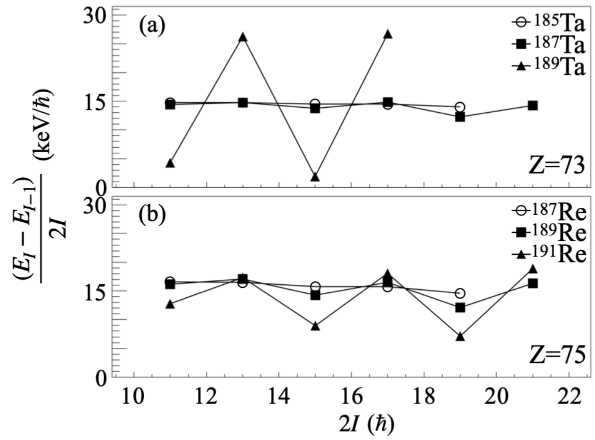

FIG. 5. Staggering of the moment of inertia of rotational bands built on the $\pi 9/2^{-}$[514] Nilsson state in (a) odd-A ^{185,187,189}Ta and (b) their isotones in odd-A Re [32], indicating the sharp transition to a strong triaxial shape in ^{189}Ta.

mary physics focus, but are bracketed in the level scheme as per spectroscopic norms when not directly measured. The smooth increase in excitation energy of the $9/2^{-}$[514] bandhead with N (175-225-283 keV in ^{185,187,189}Ta, respectively), and its similar γ-decay to the $7/2^{+}$[404] band members as compared to ^{185,187}Ta [30, 31], support the proposed level scheme.

The most striking feature that the level scheme reveals is the large staggering between the two signatures of the rotational band populated in the isomeric decay, which is a classic manifestation of a strong triaxial shape. In an odd-A nucleus, the coupling of the angular momentum of the unpaired nucleon to the collective rotation of the even-even core provides a sensitive measure of asymmetries in the underlying nuclear shape. A standard method of quantifying this staggering (or signature splitting) is to plot the quantity $(E_I - E_{I-1})/2I$ as a function of $2I$, as in Fig. 5(a). A softening of axial rigidity towards triaxial shapes is expected as the Z=82 shell closure is approached, as has been observed in the Z=75 isotones of

TABLE I. Transition energies (to the nearest keV), relative γ intensities, (tentative) spin-parity assignments of initial and final states, and relative total intensity (corrected for internal conversion assuming a pure transition multipolarity), for γ rays from the decay of the 1309-keV isomer in ^{189}Ta.

E_γ (keV)	I_γ	$J_i^\pi \rightarrow J_f^\pi$	Mult	$I_{\rm tot}$
83	0.09(3)	$(9/2^+) \rightarrow (7/2^+)$	$M1$	0.65(19)
154	1.00(7)	$(19/2^+) \rightarrow (17/2^-)$	$E1$	1.00(7)
200	0.11(3)	$(9/2^-) \rightarrow (9/2^+)$	$E1$	0.11(3)
247	0.42(4)	$(11/2^-) \rightarrow (9/2^+)$	$E1$	0.39(4)
283	0.54(6)	$(9/2^-) \rightarrow (7/2^+)$	$E1$	0.49(5)
342	0.26(4)	$(13/2^-) \rightarrow (11/2^-)$	$M1$	0.26(4)
369	0.14(3)	$(15/2^-) \rightarrow (11/2^-)$	$E2$	0.14(3)
389	0.62(7)	$(13/2^-) \rightarrow (9/2^-)$	$E2$	0.58(6)
455	0.13(3)	$(17/2^-) \rightarrow (15/2^-)$	$M1$	0.12(2)
482	0.94(8)	$(17/2^-) \rightarrow (13/2^-)$	$E2$	0.86(7)

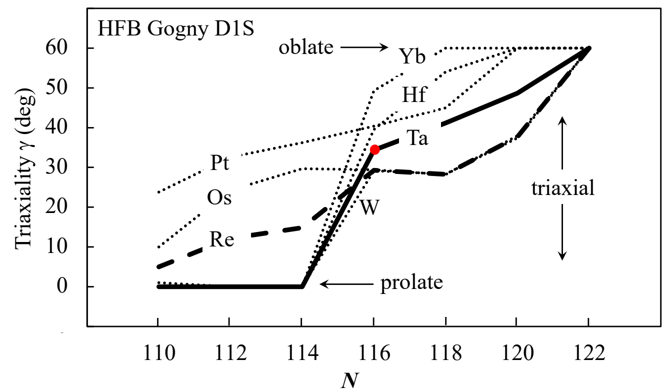

FIG. 6. Theoretical predictions of Robledo *et al.*(dotted) for shape evolution of even-even $70 \leq Z \leq 78$ nuclei as a function of N [7]. The thick lines for the odd-Z Ta (Z=73) (solid) and Re (Z=75) (dashed) isotopes are interpolations between their nearest even-even neighbors. The red dot denotes ^{189}Ta, with maximal axial asymmetry ($\gamma \approx 30°$) at N=116, in agreement with experiment. Recent complementary results in ^{190}W [35] also indicate such a transition at N=116. In contrast, the FRDM model [10] predicts a prolate-to-oblate shape transition at N=120 (see text).

Re nuclei shown in Fig. 5(b), and long known in higher-Z Os and Pt nuclei [3]. Nevertheless, the suddenness of the transition from prolate to strong triaxial shapes in the odd-A Z=73 Ta nuclei at N=116 compared to the odd-A Z=75 Re nuclei is stark (Fig. 5).

The strong departure from axial symmetry is also supported by the reduced hindrances $f_\nu = (F_W)^{1/\nu}$ for the γ decay of K-isomers. The hindrance F_W is the ratio of partial γ-ray half-life to the Weisskopf single-particle estimate for direct decays out of the isomers, and $\nu (= \Delta K - \lambda)$ is the degree of forbiddenness for a transition of multipolarity λ between two different K states. While there is a large spread, typical "good" K-isomers exhibit f_ν values of 20 or higher [33, 34]. The f_ν for K-hindered $E2$ transitions in odd-A Ta nuclei evolve from $f_\nu = 71$ in ^{185}Ta [31] to 27 in ^{187}Ta [30], with axial symmetry still holding. In ^{189}Ta, while the decay of the upper isomer is K-allowed, the K-hindered 154-keV decay yields a f_ν of just 7, which folds in an additional 10^{-4} hindrance factor typically used to compare $E1$ decays with other multipolarities. This heralds a severe erosion of the K quantum number and a breakdown of axial symmetry at N=116.

The new spectroscopic results in ^{189}Ta provide an excellent test for model predictions that have been awaiting experimental data in this difficult-to-access neutron-rich region. Theories with published comprehensive calculations in this region include self-consistent constrained HFB calculations of Robledo *et al.* [7] and Delaroche *et al.* [8] using Gogny and Skyrme interactions, as well as the macroscopic-microscopic finite-range droplet model (FRDM) using the folded-Yukawa potential of Möller *et al.* [9, 10]. While all the models can include triaxiality γ in their parameter space to map the evolution from prolate to oblate through triaxial shapes, the published FRDM results for our specific nuclei only include axial shapes involving quadrupole and higher multipole deformations. We have chosen, therefore, to highlight one of the representative HFB calculations in detail, and discuss the FRDM results in comparison. Fig. 6 compares the predictions for even-even nuclei in the constrained HFB model of Robledo *et al.* [7] using the Gogny D1S interaction. While the published calculations are only for even-even systems, simple interpolations provide an excellent perspective for odd-Z behavior. The abrupt departure from prolate to a strong triaxial shape at N=116 for Ta on the way to oblate shapes is in excellent agreement with experiment, as is the difference in behavior between the Ta and Re isotones, where the latter exhibit a more gradual evolution in γ, as seen in the staggering patterns of the collective band structures in their respective level schemes (Fig. 5). In comparison, the FRDM calculations for Ta predict a prolate shape at N=118 with a transition to oblate at N=120 [10]. Also, Total Routhian Surface calculations using the cranked Woods-Saxon-Strutinsky method [6] show a transition from prolate to oblate in odd-odd Ta (N=115,117) systems at $\hbar\omega$ = 0.1 MeV, again identifying ^{189}Ta as a critical nucleus defining the prolate-oblate boundary at N=116. The experimental signature staggering in the moment of inertia of the rotational band built on the π9/2$^-$[514] Nilsson state in ^{189}Ta (Z=73) is more pronounced compared to the corresponding ^{191}Re (Z=75) isotone, indicating a larger axial asymmetry than $\gamma = 25^\circ$ deduced for ^{191}Re [32]. This observation is not in line with the arguments presented in earlier comparisons using only N=114 data available at the time, where it was claimed that triaxiality plays a reduced role as Z decreases [7, 30]. The isotopes of odd-A Re (Z=75) exhibit a more gradual evolution of triaxial shapes, as evidenced from their observed staggering (Fig. 5), which is also in keeping with theoretical expectations interpolated from their even-even W (Z=74) and Os (Z=76) neighbors. Thus the N=116 ^{189}Ta nucleus seems to mimic a sharp critical-point at a phase transition in the shape landscape as a function of neutron number. Complementary recent work measuring the lifetime of the first-excited 2$^+$ state in the neighboring even-even ^{190}W nucleus populated by fragmenting a 1 GeV/A ^{208}Pb beam also indicate a switch from prolate to oblate deformation at N=116 [35]. Extending the spectroscopy using fragmentation reactions to even lower-Z N=116 systems (^{188}Hf, ^{187}Lu) would allow a comprehensive experimental mapping of prolate-to-oblate shape evolution via intermediate strongly-triaxial shapes with distinctive signatures. How shapes of neutron-rich nuclei in this unexplored region evolve towards the N=126 shell closure are questions that future experiments being proposed at next-generation facilities will address.

In summary, we have experimentally accessed very-neutron-rich $A\approx$190 nuclei using the fragmentation of a ^{198}Pt primary beam for the first time. This work proves that fragmentation reactions, in addition to their potential of discovering new isotopes, can produce nuclei in isomeric states at high angular momenta that survive the flight paths to the experimental end station. With fragment-specific spectroscopy of a chosen range of isotopes implanted in a Si stack surrounded by the GRETINA array, we have identified and measured a wide range of isomers, many for the first time, and used their γ-decay pathways to explore new physics on the evolution of shapes. The level structure deduced for the very-neutron-rich ^{189}Ta (N=116) nucleus reveals a sudden transition to a strongly triaxial shape from the robust prolate axial symmetry observed in lighter isotopes closer to stability, providing a critical experimental test for competing model predictions. With precise particle-identification tools coupled with γ-ray detection with high efficiency, modularity and resolution, such fragmentation experiments open up new landscapes for nuclear structure studies far from stability [15, 16], with eventual access to nuclei in the rapid neutron capture process pathway critical for stellar nucleosynthesis.

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