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Excitation and De-excitation Pathways of the ^{178}Hf Isomeric State

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Abstract

Nuclear isomers with long-lived high-excitation states are of significant importance in national strategic security and energy storage fields. The key technology lies in mastering the artificial induction of rapid de-excitation of nuclear isomers to the ground state. In recent years, research on this technology has encountered bottlenecks, requiring investigation from the perspective of nuclear structure studies. Through in-depth study of the formation, stimulated excitation, and de-excitation mechanisms of nuclear isomers, these bottlenecks can be broken through, thereby mastering the key technology for inducing rapid de-excitation of nuclear isomers to the ground state. In this work, on the experimental technique front, a white neutron beam was utilized to bombard a Hf target. We innovatively established a triple-coincidence measurement system combining full-energy measurement by a barium fluoride detector array with characteristic gamma-ray measurement by a high-purity germanium detector, along with white neutron time-of-flight windowing. Through analysis of the experimental data, we discovered indications of excitation of the ^{178}Hf isomeric state to a transitional energy level and subsequent rapid release.

Full Text

Preamble

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Study on the Excitation and Deexcitation Paths of the ^{178}Hf Isomeric State

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Abstract

Isomers with long lifetimes and high excitation energies hold significant importance for national strategic security and energy storage applications. The key technological challenge lies in mastering artificial methods to induce rapid deexcitation of isomers to their ground states. Recent progress in this field has encountered bottlenecks, necessitating a fundamental approach from nuclear structure physics. By investigating the formation, stimulated excitation, and deexcitation mechanisms of isomers, we can overcome these obstacles and develop the critical technologies needed for controlled isomer decay. In this work, we employed white neutron beams to bombard a Hf target and innovatively established a triple-coincidence measurement system combining a BaF_2 detector array for total energy measurement with a high-purity germanium (HPGe) detector for characteristic gamma-ray spectroscopy, integrated with neutron time-of-flight gating. Analysis of the experimental data reveals signatures of the ^{178}Hf isomeric state being excited to a transitional level followed by rapid energy release.

Keywords: ^{178}Hf isomer; induced decay; nuclear energy release; multi-probe multi-parameter gamma-ray coincidence measurement system

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Introduction

Isomeric states represent a special class of nuclear excitations whose unique structure inhibits spontaneous decay [1]. Nuclides with long-lived excited states are known as nuclear isomers, which hold important research significance in nuclear structure and astrophysics, as well as promising applications in clean energy, high-capacity energy storage devices, nuclear clocks, medical diagnostics and therapy, and gamma-ray lasers [2]. Over the past two decades, experimental studies on nuclear isomers have grown rapidly [3].

Before Chadwick's discovery of the neutron [4], Soddy proposed the concept of multiple structures within nuclides [5]. In 1921, Hahn first obtained experimental evidence for the existence of ^{234}Pa isomers [6], but it was not until 1936 that von Weizsäcker provided a theoretical explanation [7]. Concurrently, neutron bombardment experiments discovered many new isomers, such as those of Br and In reported by Kurtchatov et al. [8] and Szilard and Chalmers [9]. Investigations revealed numerous high-excitation-energy (>1 MeV) and long-lifetime (>1 ms) isomers in the $A=180$ mass region [10].

Based on their formation and existence mechanisms, isomers are primarily classified into five categories [3]. The first category comprises spin isomers, which arise when gamma-ray emission from nuclear excited states cannot satisfy spin selection rules (including angular momentum conservation) [10], with ^{180}Ta serving as a representative nuclide [11,12]. The second category consists of K-isomers, a variant of spin isomers where decay depends not only on the magnitude but also on the orientation of the nuclear spin vector, exemplified by ^{180}Hf [13]. The third category includes shape isomers, which occur when a nucleus in its ground state is deformed and possesses multiple minima in its potential energy surface; if the secondary minimum is sufficiently deep, shape isomerism may result [3], as demonstrated by ^{72}Kr [14]. The fourth category comprises fission isomers [15,16], a type of shape isomer commonly found in heavy nuclei, where decay competes between gamma-ray deexcitation to the ground state and spontaneous fission, with ^{242}Am as a representative case [17]. The fifth category consists of seniority isomers, where electromagnetic decay is hindered by selection rules related to seniority quantum numbers, with their existence primarily governed by the quadrupole pairing matrix elements of nucleon-nucleon interactions.

Isomers have found established applications in medical imaging [20] and Mössbauer spectroscopy [21]; however, their utilization as energy storage materials requires further investigation. The key technological challenge is identifying a controllable method for the stimulated excitation and deexcitation of long-lived, high-excitation-energy isomers. In 1999, Belic et al. achieved artificially induced deexcitation of ^{180}Ta [19], but its excitation energy of only 77 keV—far below the 1 MeV photon energy required to induce premature decay—limits its practical applications. In contrast, ^{178}Hf represents a more promising candidate,

featuring a 31-year lifetime and 2.4 MeV excitation energy. In 1999, Collins claimed that x-rays with energies below 100 keV could induce premature decay of ^{178}Hf [22], though these results remain controversial [23]. Current research indicates multiple approaches for inducing isomer deexcitation, including photon stimulation, Coulomb excitation, thermal neutron capture [24], and electron-nucleus interactions [25-27].

To date, researchers have yet to identify an effective technique for artificially inducing premature isomer decay, particularly for ^{178}Hf , where studies have reached a bottleneck. Addressing this challenge requires a fundamental approach from nuclear structure research, focusing on deep investigation of isomer formation, stimulated excitation, and deexcitation mechanisms to break through current limitations and master the key technologies for inducing isomer decay. Theoretical studies suggest a viable mechanism wherein isomers are first excited to nearby short-lived transitional states [28] before completing deexcitation and releasing energy.

Theoretical Framework and Experimental Motivation

In 2006, Gao Zaochun, Tuya, and Chen Yongshou introduced the γ degree of freedom into the Projected Shell Model (PSM), establishing a triaxial projected shell model (TPSM) with multi-quasiparticle configurations [29] that successfully reproduced the multi-quasiparticle bands of ^{178}Hf and the experimentally observed γ -vibrational band built on the ground state. If the intrinsic configuration of ^{178}Hf possesses the same γ degree of freedom as the ground state, several rotational bands may exist based on the ^{178}Hf intrinsic configuration. Theoretical predictions place the first 14^+ state approximately 860 keV above ^{178}Hf . Since these rotational bands share the same intrinsic configuration, excitation from the ^{178}Hf (16^+) state to the 14^+ state should be relatively facile. Consequently, this 14^+ state can serve as a short-lived transitional state in the excitation and deexcitation process of ^{178}Hf [30]. Experimental verification of such a transitional level and identification of its excitation and deexcitation pathways hold significant scientific and practical value. This project employs white neutron beams to bombard a Hf target, utilizing a multi-probe multi-parameter gamma-ray coincidence measurement system combining the GTAF-II BaF₂ detector array with an HPGe detector to investigate the isomeric state's excitation and deexcitation paths, aiming to find evidence for the theoretically predicted transitional state and discover its deexcitation pathways to lower excited states.

Experimental Setup

The experiment was conducted at the Back-n white neutron source terminal [31] at the China Spallation Neutron Source. A pulsed white neutron beam spanning more than eight orders of magnitude in energy (1 eV–100 MeV) bombarded a natural Hf target 50 mm in diameter and 1 mm thick, with isotopic composition of 18.60% ^{177}Hf , 27.28% ^{178}Hf , 13.82% ^{179}Hf , and 35.08% ^{180}Hf .

The target material was placed in air for this experiment. The experimental approach involved first populating the ^{178}Hf isomeric state through neutron-induced reactions, then exciting the ^{178}Hf isomer to transitional levels using white neutrons of various energies, followed by rapid deexcitation to lower excited states. Gamma rays emitted from the target nucleus were detected using a combination of the GTAF-II array with 40 BaF_2 detector units [34] and a single HPGe detector. The data acquisition system employed XIA's universal digital acquisition system [35]. By applying triple-coincidence gating between the sum peaks measured by the BaF_2 detectors, characteristic gamma peaks measured by the HPGe detector, and neutron time-of-flight, we searched for and verified the existence of the theoretically predicted transitional level.

Detector Systems and Measurement Principle

The detector array consisted of two setups. "Detector Setup I," shown in Figure 1 [Figure 1: see original paper], comprised the GTAF-II array with an added HPGe detector, along with associated instrumentation and data acquisition systems. This configuration leveraged the high efficiency of the BaF_2 array combined with the high energy resolution of the HPGe detector to initially search for the predicted transitional level. "Detector Setup II," illustrated in Figure 2 [Figure 2: see original paper], consisted of LaBr_3 and HPGe detectors, utilizing the excellent time resolution of LaBr_3 and the high energy resolution of HPGe to precisely identify the deexcitation pathways of the ^{178}Hf isomeric state. The white neutron beam used in this experiment featured a broad energy spectrum in pulsed mode, enabling neutron energy determination through time-of-flight measurements [38]. We employed neutron time-of-flight gating to establish whether excitation of the ^{178}Hf isomer to the transitional state was temporally correlated with specific neutrons.

The experimental data analyzed in this paper primarily originated from Detector Setup I. The fundamental principle of this system's total-energy gamma spectroscopy measurement is as follows: when a nucleus deexcites from an excited state to the ground state, the BaF_2 detector array efficiently measures the cascade gamma rays from various deexcitation pathways. The temporal correlations between cascade gammas are recorded through fast-timing coincidence, and the energies of coincident gamma rays are summed to produce sum peaks corresponding to the excited state's energy. Simultaneously, we used the high-resolution HPGe detector to measure characteristic gamma rays from the deexcitation of the ^{178}Hf isomer to specific lower excited states. By applying coincidence gating between these HPGe measurements and the BaF_2 sum peaks, we could identify whether observed excited states belonged to ^{178}Hf . Building upon this, triple-coincidence gating with neutron time-of-flight was implemented to further verify if the excited state represented the theoretically predicted transitional state and whether it was neutron-induced, as illustrated in Figure 3 [Figure 3: see original paper].

Based on this methodology, we aimed to indirectly determine the existence of

the predicted transitional level. Since this ^{178}Hf transitional state may deexcite through multiple pathways, the total energy of the cascade gamma rays remains constant and equals the transitional state's energy. By using the high-efficiency GTAF-II BaF_2 detector array to collect as many cascade gamma rays as possible from the deexcitation of this ^{178}Hf transitional state, a “sum peak” should appear near the theoretically predicted transitional state energy in the measured spectrum.

Furthermore, the intense beam-related background in the white neutron beam [36,37] interferes with observation of the sum peaks. To reduce this background, we replaced one BaF_2 detector unit in the GTAF-II array with a high-energy-resolution HPGe detector for coincidence gating. Leveraging the nuclear structure characteristics of ^{178}Hf , we used the 213 keV gamma ray emitted in the decay from the 4^+ to 2^+ state to distinguish gamma rays from the ^{178}Hf deexcitation process from beam-induced background. Comparison between Figure 4a [Figure 4: see original paper] and Figure 4b reveals that the unfiltered sum-peak spectrum in Figure 4a is overwhelmed by excessive background, obscuring observable phenomena. In contrast, Figure 4b shows the sum-peak spectrum after filtering with the 213 keV gamma ray detected by the HPGe detector, where the background is dramatically suppressed. The data processing involved gating on the 213 keV peak in the HPGe spectrum to examine coincident sum peaks in GTAF-II, with a coincidence time window of 50 ns.

Data Analysis and Discussion

As shown in Figure 4b, analysis of the experimental data yielded the GTAF-II sum-peak spectrum gated by the 213 keV characteristic gamma ray of ^{178}Hf . The spectrum exhibits two prominent features: a peak at 3.0 MeV and a broad structure between 6 and 9 MeV. The 6–9 MeV peak corresponds to sum peaks from various gamma rays emitted during deexcitation of compound states formed via the $^{178}\text{Hf}(n,\gamma)$ reaction after neutron capture. The 3.0 MeV peak likely represents sum peaks from deexcitation of the theoretically predicted 14^+ transitional state.

Comparison between experimental measurements and theoretical calculations of the ^{178}Hf transitional state energy, shown in Figure 5 [Figure 5: see original paper], demonstrates good agreement. The theoretically predicted transitional level energy is 3.3 MeV, while the experimentally measured sum-peak energy after gating on the 213 keV line is 3.0 MeV, yielding an experimental level energy of 3.2 MeV. This preliminary identification suggests the observed state may be the theoretically predicted transitional state. Further verification through triple-coincidence gating with neutron time-of-flight is required.

Figure 6 [Figure 6: see original paper] presents the white neutron time-of-flight spectrum, showing a significant peak in the 330–340 ns region, suggesting that neutrons of this energy may induce the observed decay. Figure 7 [Figure 7: see original paper] displays the triple-coincidence sum-peak spectrum gated by

both the 213 keV HPGe signal and the 330–340 s neutron time-of-flight window. A clear peak appears at 3.0 MeV, providing preliminary evidence that this transitional state is temporally correlated with neutron-induced excitation.

Based on comprehensive analysis of the data shown in Figures 4b and 7, combined with the theoretical-experimental comparison in Figure 5, we confirm that the 3.0 MeV feature observed in Figures 4b and 7 corresponds to the energy of the theoretically predicted 14^+ transitional state. To further verify that our measurements are not artifacts from background or accidental coincidences, we performed additional gating analyses near the 213 keV energy region and the 330–340 s time-of-flight region.

Figure 8 [Figure 8: see original paper] shows the spectrum gated on 300 keV gamma rays (background region near ^{178}Hf 's 213 keV line) and 330–340 s neutron time-of-flight, revealing no peak in the transitional level energy region. Figure 9 [Figure 9: see original paper] shows the spectrum gated on 100 keV gamma rays (another background region near 213 keV) and 330–340 s neutron time-of-flight, also showing no peak. Figure 10 [Figure 10: see original paper] presents the spectrum gated on 213 keV gamma rays but with neutron time-of-flight in the 350–360 s range (outside the neutron peak region), again showing no peak. Figure 11 [Figure 11: see original paper] shows the spectrum gated on 213 keV gamma rays with neutron time-of-flight in the 310–320 s range (also outside the neutron peak region), with no peak observed. It should be noted that the neutron time-of-flight spectrum in Figure 6 was obtained after gating on the HPGe 213 keV line, displaying multiple peaks corresponding to excitations to 3 MeV and ^{178}Hf resonance absorption. The strong peaks in the 310–320 s and 350–360 s ranges belong to resonance absorption events not temporally correlated with the 3 MeV excitation, which explains the absence of peaks at 3 MeV in Figures 10 and 11.

Therefore, comprehensive analysis of the experimental data reveals signatures of the ^{178}Hf isomeric state being excited to a transitional level with rapid energy release, suggesting this transitional state may indeed be the predicted intermediate level in the excitation and deexcitation process of the ^{178}Hf isomer.

Conclusion

Long-lived, high-excitation-energy isomers hold significant importance for energy applications and national strategic security. However, the key technology for artificially inducing premature isomer decay remains to be developed. Theoretical predictions indicate the existence of a 14^+ transitional state, and excitation of ^{178}Hf to this state could enable rapid nuclear energy release.

In this experiment, we bombarded a Hf target with white neutron beams and combined the high efficiency of the BaF_2 detector array with the high resolution of the HPGe detector to perform triple-coincidence gating of total-energy sum peaks and characteristic gamma rays with neutron time-of-flight. As shown in Figure 5 [Figure 5: see original paper], we observed signatures of the ^{178}Hf

isomeric state being excited to a transitional level with rapid energy release, suggesting this state may be the theoretically predicted intermediate level in the isomer's excitation and deexcitation process. Future work must further confirm the existence of this transitional state under different experimental conditions and identify pathways for accelerated energy release from isomers.

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References

- [1] JAIN A K, MAHESHWARI B, GOEL A. Nuclear isomers: A primer [M]. Springer Nature, 2021.
- [2] WALKER P M, CARROLL J J. *Physics Today*, 2005, 58(6).
- [3] GARG S, MAHESHWARI B, SINGH B, et al. *Atomic Data and Nuclear Data Tables*, 2023, 150: 101546.
- [4] CHADWICK J. *Proceedings of the Royal Society of London Series A, Containing Papers of a Mathematical and Physical Character*, 1932, 136(830): 692.
- [5] SODDY F. *Nature*, 1917, 99(2482): 244.
- [6] HAHN O. *Berichte der deutschen chemischen Gesellschaft (A and B Series)*, 1921, 54(6): 1131.
- [7] V. WEIZSÄCKER C. *Naturwissenschaften*, 1936, 24(51): 813.
- [8] KURCHATOV B, KURCHATOV J, LATYCHEV G. *Comptes Rendus Acad Sci*, 1935, 200: 1199.
- [9] SZILARD L, CHALMERS T. *Nature*, 1935, 135(3403): 98.
- [10] WALKER P, DRACOULIS G. *Nature*, 1999, 399(6731): 35.
- [11] EBERHARDT P, GEISS J, LANG C, et al. *Zeitschrift für Naturforschung A*, 1955, 10(9-10): 796.
- [12] BAUMINGER E, COHEN S. *Physical Review*, 1958, 110(4): 953.
- [13] BURSON S, BLAIR K, KELLER H, et al. *Physical Review*, 1951, 83(1): 62.
- [14] GÖRGEN A, CLEMENT E, BOUCHEZ E, et al. Shape coexistence in light krypton isotopes [C]//AIP Conference Proceedings: volume 802. American Institute of Physics, 2005: 275.
- [15] VANDENBOSCH R. *Annual Review of Nuclear Science*, 1977, 27: 1.
- [16] BJØRNHOLM S, LYNN J. *Reviews of Modern Physics*, 1980, 52(4): 725.
- [17] BARNES R, HENDERSON D, HARKNESS A, et al. *Journal of Inorganic*

- and Nuclear Chemistry, 1959, 9(2): 105.
- [18] VAN ISACKER P. Seniority isomers in nuclei [C]//Journal of Physics: Conference Series: volume 322. IOP Publishing, 2011: 012003.
 - [19] BELIC D, ARLANDINI C, BESSERER J, et al. Phys Rev C, 2002, 65(3): 035801.
 - [20] DRACOU LIS G, WALKER P, KONDEV F. Reports on Progress in Physics, 2016, 79(7): 076301.
 - [21] MÖSSBAUER R L. Zeitschrift für Physik, 1958, 151: 124.
 - [22] COLLINS C, DAVANLOO F, IOSIF M, et al. Phys Rev Lett, 1999, 82(4): 695.
 - [23] LI YANG T, DE ZE R, LONG WU H, et al. Phys Rev C, 2013, 88(1): 014303.
 - [24] CARROLL J, LITZ M, NETHERTON K, et al. Nuclear structure and depletion of nuclear isomers using electron linacs [C]//AIP Conference Proceedings: volume 1525. American Institute of Physics, 2013: 586.
 - [25] ZADERNOVSKY A, CARROLL J. Hyperfine interactions, 2002, 143: 153.
 - [26] HARSTON M, CARROLL J. Laser physics, 2005, 15(4): 487.
 - [27] RZADKIEWICZ J, POLASIK M, SŁABKOWSKA K, et al. Phys Rev Lett, 2021, 127(4): 042501.
 - [28] CARROLL J J. Nucl Instr and Meth B, 2007, 261(1-2): 960.
 - [29] GAO Z C, CHEN Y, SUN Y. Phys Lett B, 2006, 634(2-3): 195.
 - [30] GAO Zao-Chun, Tuya, CHEN Yong-Shou. γ degree of freedom for the 178Hf isomeric state [C]//The 11th National Nuclear Structure Conference and the 7th National Nuclear Structure Symposium. Changchun: China Institute of Atomic Energy, 2006: 109. (in Chinese)
 - [31] CHEN H, WANG X L. Nature materials, 2016, 15(7): 689.
 - [32] KARAMIAN S, CARROLL J, ADAM J, et al. High Energy Density Physics, 2006, 2(1-2): 48.
 - [33] KARAMIAN S, CARROLL J. Phys Rev C, 2011, 83(2): 024604.
 - [34] TANG J Y, AN Q, BAI J B, et al. Nuclear Science and Techniques, 2021, 32: 1.
 - [35] WU H, LI Z, TAN H, et al. Nucl Instr and Meth A, 2020, 975: 164200.
 - [36] ZHANG L, JING H, TANG J, et al. Applied Radiation and Isotopes, 2018, 132: 212.
 - [37] LI Q, JING H, ZHOU B, et al. Nucl Instr and Meth A, 2020, 980: 164200.
 - [38] AN Q, BAI H, BAO J, et al. Journal of Instrumentation, 2017, 12(07): P07022.

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