Structural Evolution in the Neutron-Rich Nuclei ^{106}Zr and ^{108}Zr

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(Received 20 December 2010; published 16 May 2011)

The low-lying states in ^{106}Zr and ^{108}Zr have been investigated by means of β-γ and isomer spectroscopy at the radioactive isotope beam factory (RIBF), respectively. A new isomer with a half-life of 620 ± 150 ns has been identified in ^{108}Zr. For the sequence of even-even Zr isotopes, the excitation energies of the first 2^+ states reach a minimum at $N = 64$ and gradually increase as the neutron number increases up to $N = 68$, suggesting a deformed subshell closure at $N = 64$. The deformed ground state of ^{108}Zr indicates that a spherical subshell gap predicted at $N = 70$ is not large enough to change the ground state of ^{108}Zr to the spherical shape. The possibility of a tetrahedral shape isomer in ^{108}Zr is also discussed.

DOI: 10.1103/PhysRevLett.106.202501 PACS numbers: 23.20.Lv, 23.35.+g, 27.60.+j

Atomic nuclei, which are strongly interacting finite quantum systems, have a spherical or deformed shape. The shape evolution as a function of proton and neutron numbers is related to the shell structure for a spherical or deformed shape. At proton number $Z = 40$, the shell structures for a wide variety of shapes, i.e., spherical, prolate, oblate, and more exotic tetrahedral shapes, are closed [1,2]. So shape transitions and maximum deformation at deformed subshell closure are expected for Zr isotopes ($Z = 40$).

The shape of neutron-rich Zr isotopes changes drastically from spherical to deformed at neutron number $N = 60$ [3]. The quadrupole deformation is known to increase toward $N = 64$ from half-life measurements of the first $J^\pi = 2^+$ (2_1^+) state of even-even Zr isotopes [4,5]. However, the evolution of the deformation beyond $N = 64$ is unknown because there is no spectroscopic information. Several authors have predicted different types of shape transition in the more neutron-rich region around $N = 70$. Specifically, a ground-state shape transition from prolate to oblate is predicted at $N = 72$ [6] or at $N = 74$ [1]. In contrast, a spherical subshell gap at $N = 70$ and a spherical shape of ^{110}Zr are predicted by the Skyrme energy density functional with a tensor force and a reduced spin-orbit term [7]. A spherical shape might be expected to appear also in Zr isotopes approaching ^{110}Zr. Furthermore, an exotic shape with tetrahedral symmetry is predicted to be stabilized around ^{110}Zr, which has the tetrahedral magic numbers, $Z = 40$ and $N = 70$ [2]. An excited state with the tetrahedral shape, predicted at ^{108}Zr, may become an isomeric state. The tetrahedral shape is still hypothetical in atomic nuclei, in spite of many recent theoretical and experimental works [8].

In addition to these striking shape effects, very neutron-rich Zr isotopes are important for understanding nucleosynthesis through the rapid neutron-capture process (r process). The estimated r-process abundances between $A = 110$ and $A = 125$ depend on nuclear mass models [9]. Here, the astrophysical model of prompt supernova explosions with O-Ne-Mg cores is applied. The abundance deficiencies by 1 order of magnitude have been reduced when mass models including shell quenching at $N = 82$ are used. A more drastic shell evolution, accompanied by not only the $N = 82$ shell quenching but also the new $N = 70$ subshell, may influence the r-process path as predicted by self-consistent mean-field calculations [7,10]. Thus, in order to predict nuclear properties reliably for $A \approx 110$–125 on the r-process path (i.e., $N = 70$–82), it is crucial to know whether the subshell gap at $N = 70$ is large or not. The $N = 70$ subshell effect can be investigated through the structural evolution of Zr isotopes near $N = 70$.

β-decay half-lives can be used as a probe of shape transitions even though their values depend mainly on the β-decay Q values. For example, calculations using the quasiparticle random-phase approximation predict shorter β-decay half-lives for a spherical shape than for a deformed one [11]. The half-lives measured previously, including ^{110}Zr [12,13], however, show a gradual decrease as a function of the neutron number, with no indication of the transition to spherical shape.

The purpose of this Letter is to study the structural evolution caused by spherical and deformed shell structures in the neutron-rich Zr isotopes through the low-lying states obtained from β-γ and isomer spectroscopy of ^{106}Zr and ^{108}Zr, and to discuss a new isomeric state of ^{108}Zr in the context of the possibility of a tetrahedral shape isomer. The 2_1^+ state energy, $E(2_1^+)$, and the ratio of the first 4^+ to 2^+ state energies, $R_{4/2} = E(4_1^+)/E(2_1^+)$, for even-even nuclei serve as valuable indicators of deformation and shell closure [14].

The experimental studies of ^{106}Zr and ^{108}Zr were performed at the radioactive isotope beam factory (RIBF) operated by the RIKEN Nishina Center and the CNS of the University of Tokyo. Secondary beams were produced using in-flight fission of ^{238}U beams having an energy of 345 MeV/nucleon. The Be production target was 3 mm thick. Fission fragments were separated using the RI-beam separator, BigRIPS [15,16]. For isotopic separation, an aluminum wedge-shaped energy degrader having a median thickness of 5.8 mm was placed at the momentum dispersive focus of the first stage of BigRIPS. An additional degrader was placed at the momentum dispersive focus of the second stage of BigRIPS in order to eliminate the fragments that were not fully stripped before reaching the second degrader. The shape of the degrader was designed to satisfy the momentum achromatic condition at the next achromatic focus. The thickness along the center of the beam line was 2.1 mm. The separated particles were transported through the ZeroDegree spectrometer [16] to the final focal plane of ZeroDegree.

Beam particles were identified using the magnetic rigidity, $B\rho$, time-of-flight, and energy loss, ΔE, determined using the focal plane detectors of BigRIPS and ZeroDegree [17]. The particle identification spectrum for the same data set is shown in Ref. [17]. The resolution of the atomic number Z and the mass-to-charge ratio A/Q for ^{108}Zr was 0.4 and 0.005 (full width at half maximum), respectively. The identified particles were implanted into nine stacked double-sided silicon strip detectors (DSSD). Each DSSD had a thickness of 1 mm with an active area of 50×50 mm^2 segmented into 16×16 strips. The implanted particles and subsequent β rays were detected by the DSSDs. The DSSDs were surrounded by four Compton-suppressed clover-type Ge detectors and two LaBr$_3$ detectors, which were used to detect γ rays following β and isomeric decays. A plastic scintillation detector was placed in front of each clover detector. By taking an anticoincidence between the plastic and clover detector, the background arising from β-ray events in the γ-ray spectrum can be eliminated.

β-decay events were selected using the position and time correlations between the implanted particle and the β ray. The relative position was restricted to the same pixel of the DSSD. The β-decay half-life of ^{106}Y was measured to be 62^{+25}_{-14} ms [13] in the same data set. Figure 1(a) shows a γ-ray spectrum measured in coincidence with β rays following the implantation of ^{106}Y within 200 ms, which is about 3 times longer than the β-decay half-life. The γ ray with 140 keV was also measured in coincidence with the β decay of ^{104}Y and ^{105}Y, which were included in the cocktail beam [17]. This γ ray was emitted from the 2_1^+ state in ^{104}Zr after β-delayed two-neutron emission of ^{106}Y. The γ ray with 169 keV was also measured in coincidence with the β decay of ^{105}Y and results from ^{105}Zr after β-delayed one-neutron emission. The most intense γ ray at 152 keV was assigned as the transition from the 2_1^+ state to the ground state in ^{106}Zr. Figure 2 shows the ground-state bands of the even-even Zr isotopes with $N \geq 60$. The gradual evolution of $E(2_1^+)$ supports the ^{106}Zr assignment.

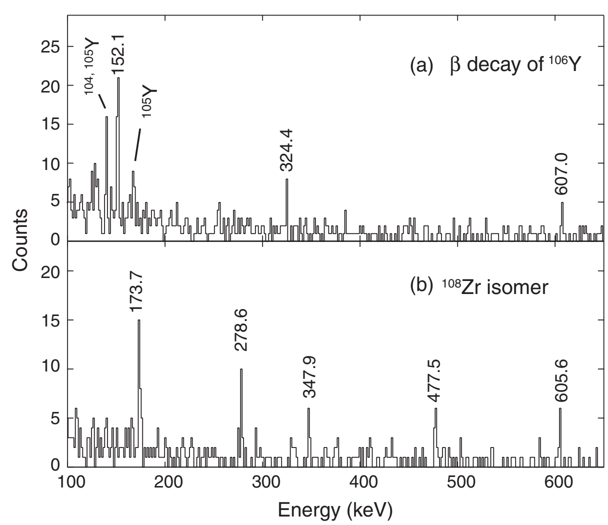

FIG. 1. Gamma-ray spectra measured (a) in coincidence with β rays detected within 200 ms after implantation of ^{106}Y and (b) with a particle gate on ^{108}Zr within 4 μs. Peaks marked with the nucleus name indicate ones measured also in coincidence with the β decay of its nucleus.

The spin and parity of the parent nucleus ${}^{106}_{39}Y_{67}$ are possibly 2^+ or 3^+, because the ground states of ${}^{99,101}Y$ are indicated to have the same proton configuration, $5/2^+[422]$, as ${}^{101,103,105}Nb$ [18–20] and the spin and parity of ${}^{108}_{41}Nb_{67}$ is suggested to be 2^+ or 3^+ [21]. The 4_1^+ and the second 2^+ (2_2^+) states of ${}^{106}Zr$ are likely to be populated in the β decay of ${}^{106}Y$ by comparison with the population of the 4_1^+ and 2_2^+ states of ${}^{108}Mo$ in the β decay of ${}^{108}Nb$ [21]. If the 324 keV γ ray is the transition to the 2_1^+ state, the excited-state energy is 477 keV. Since $E(2_1^+)$ of ${}^{106}Zr$ is slightly larger than that of ${}^{104}Zr$ (Fig. 2), $E(4_1^+)$ is expected to increase gradually and to be 450–500 keV. The energies of the 4_1^+ and 2_2^+ states of ${}^{106}Zr$ are predicted by using the interacting boson model [22]. The parameters of the interacting boson model are obtained from a least-squares fit to the known level energies of ${}^{108}Mo$, ${}^{110}Ru$, and ${}^{112}Pd$ along the isotonic chain ($N = 66$). The largest deviations between the experimental and theoretical $E(4_1^+)$ and $E(2_2^+)$ are 34 keV and 76 keV, respectively. The $E(4_1^+)$ and $E(2_2^+)$ of ${}^{106}Zr$ are extrapolated to be 455 keV and 618 keV, respectively. Therefore, the excited states at 477 keV and 607 keV were tentatively assigned as the 4_1^+ and 2_2^+ states in ${}^{106}Zr$, respectively. The transition from the 2_2^+ state to the 2_1^+ state is expected, but no γ-ray peak at 455 keV was observed due to the low statistics.

The γ rays emitted from a new isomeric state of ${}^{108}Zr$ were observed within 4 μs after the implantation of ${}^{108}Zr$ as shown in Fig. 1(b). Five γ-ray peaks at energies of 174, 279, 348, 478, and 606 keV were unambiguously measured. A half-life of 620 ± 150 ns was derived from the sum of time spectra for these five γ rays. Some low-intensity γ-ray peaks from the ${}^{108}Zr$ isomer might not have been identified, and no information on γ-γ coincidences was obtained due to the low statistics. Nevertheless, it can be estimated that the energy of the isomeric state is likely more than 1 MeV. The ground-state band is populated up to 4^+; thus, the spin is likely more than or equal to 4. Before discussing possible structures of the observed isomer, low-lying states of ${}^{108}Zr$ are discussed.

If a spherical ground state would appear around ${}^{110}Zr$ due to the predicted $N = 70$ subshell gap [7], then $E(2_1^+)$ would have to suddenly increase and $R_{4/2}$ drop to ≈ 2. However, $E(2_1^+)$ of ${}^{106}Zr$ is similar to that of Zr isotopes with $A = 100$–104, which are well deformed with $\beta_2 = 0.355(10)$, 0.43(4), and 0.47(7) for $A = 100$, 102, and 104, respectively [4,5]. Because the γ-ray energies of 174 and 348 keV in ${}^{108}Zr$ are slightly larger than those of 152 and 324 keV in ${}^{106}Zr$ and the relevant energies smoothly change from ${}^{100}Zr$ to ${}^{108}Zr$ (Fig. 2), the 174 and 348 keV γ rays were tentatively assigned as the transitions from the 2_1^+ state to the ground state and from the 4_1^+ state to the 2_1^+ state, respectively. $R_{4/2}$ gradually changes with values of 2.57, 3.15, 3.25, 3.13, and 3.00 for $A = 100$, 102, 104, 106, and 108, respectively. Values of $R_{4/2}$, which is close to 3.3 for a rigid rotor, indicate the rotational character of a deformed nucleus. The ground state of ${}^{108}Zr$ is most likely as deformed as ${}^{106}Zr$. Therefore, the spherical subshell gap at $N = 70$ seems not to be large enough to change the ground state of ${}^{108}Zr$ to spherical shape.

The structural evolution around the neutron-rich Zr isotopes can be visualized using $1/E(2_1^+)$ [14]. Figure 3 shows $1/E(2_1^+)$ as a function of the neutron number. The values of $1/E(2_1^+)$ suddenly increase at $N = 60$ for Kr, Sr, Zr, and Mo isotopes because of the onset of deformation. $1/E(2_1^+)$ reaches a maximum at $N = 64$ for both Zr and Mo isotopes. Another remarkable behavior at $N = 64$ has been observed for Mo isotopes. Hua *et al.* observed a band crossing due to the rotation alignment of an $h_{11/2}$ neutron pair [23]. The shift of the band crossing to higher rotational frequency in ${}^{106}Mo$ is interpreted as a consequence of the deformed subshell closure at $N = 64$. The maximum of $1/E(2_1^+)$ at $N = 64$ can also be interpreted as being due to the deformed subshell closure at $N = 64$ with $\beta_2 \approx 0.47(7)$ [5] for ${}^{104}Zr$.

The r-process path between $A = 110$ and $A = 125$ may be affected by the weakening of the spin-orbit force, which is associated with the neutron skin [24]. The harmonic-oscillator-like doubly magic nucleus of ${}^{110}Zr$ [24] or the

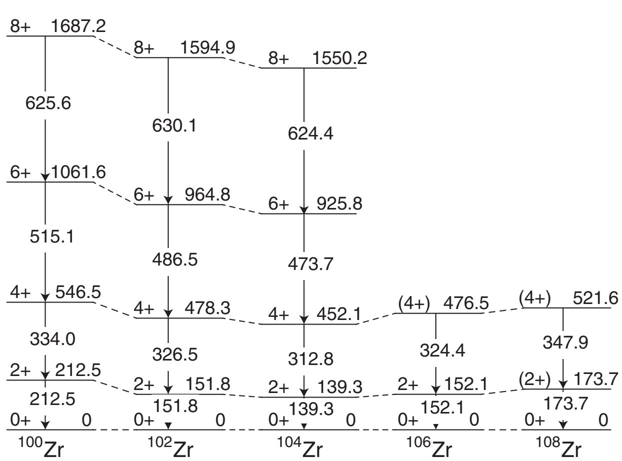

FIG. 2. Ground-state bands of neutron-rich even-even Zr isotopes with $N \geq 60$. The energies of ${}^{100-104}Zr$ are taken from the ENSDF database [27].

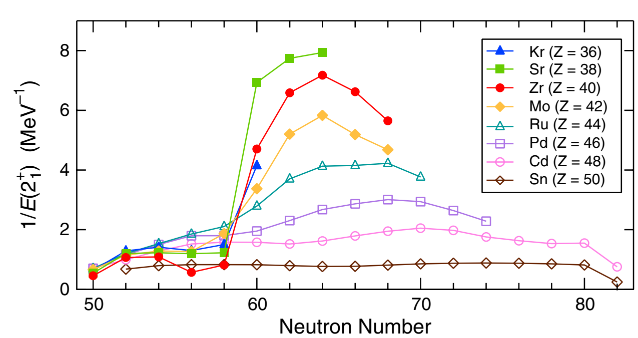

FIG. 3 (color online). $1/E(2_1^+)$ as a function of the neutron number. The present results are for Zr isotopes with $N = 66$ and 68. Others are taken from the ENSDF database [27].

shell closure at $N = 70$ [7] is predicted by using an artificially reduced spin-orbit force. However, the shape of ^{108}Zr as deformed as lighter Zr isotopes indicates no drastic evolution. One may conclude that the deficiencies of the estimated r-process abundances around $A = 110$ are not caused by the drastic weakening of the spin-orbit force.

The existence of a long-enough-lived ($T_{1/2} > 100$ ns) isomer gives access to the excited-state structure. The ^{108}Zr isomer is the only isomer discovered in even-even nuclei in the present work, although the production yield of the isotopes ^{104,106}Zr and the isotone ^{110}Mo was higher than ^{108}Zr. It seems that the structure of the ^{108}Zr isomer is suddenly stabilized.

A possible explanation of the isomerism is that the isomeric state of ^{108}Zr has a tetrahedral shape. The tetrahedral shape is a nonaxial octupole deformation coupled with a vanishingly small quadrupole deformation; therefore, a γ decay to normally quadrupole-deformed states is hindered [8]. The tetrahedral shape will appear only when the competing shell effect for spherical shape is weak [25]. This requirement might be satisfied because the ^{108}Zr results indicate the deformed ground state. Furthermore, the energy barrier against different shapes plays an important role for the stability. The total energies of the tetrahedral- or quadrupole-deformed shape are calculated using the microscopic-macroscopic method with pairing correlations [2]. The energy barrier between the tetrahedral and oblate shapes is predicted for ^{108}Zr, but becomes very small for ^{104,106}Zr. The onset of long-lived isomerism at ^{108}Zr may be caused by the difference of the energy barrier. The excitation energy E_x is predicted to be 1.1 MeV by the Hartree-Fock-Bogoliubov calculation [2]. The spin J of a possible band head is expected to be 3 [2], and the spin of the isomeric state might be larger than that of the band head [25]. These expectations are consistent with the experimental indications, i.e., $E_x > 1$ MeV and $J \geq 4$. While the parity of the tetrahedral shape is predicted to be negative [26], there is no experimental indication.

Another possibility of the isomeric state is a two-quasineutron state with high K value, which is predicted for several even-even nuclei around ^{108}Zr [6]. Considering the gradual change of $E(2_1^+)$ around ^{108}Zr as shown in Fig. 3, this kind of isomeric state is expected to have a similar half-life and be populated not only in ^{108}Zr but also in neighboring even-even nuclei. Thus, we propose the observed isomer is a promising candidate for the tetrahedral shape isomer rather than the high-K isomer.

In summary, decay spectroscopy has been performed to assign the first 2^+ and 4^+ states of ^{106,108}Zr. The systematics of $E(2_1^+)$ indicate that the deformation reaches a maximum at $N = 64$ for Zr isotopes, suggesting a deformed subshell closure at $N = 64$. In addition, the deformed ground state of ^{108}Zr indicates that the spherical $N = 70$ subshell gap is not having a large effect at $N = 68$ for Zr isotopes. For a definite conclusion regarding the $N = 70$ subshell, future measurements of ^{110}Zr are required. A long-lived ($T_{1/2} > 100$ ns) isomer, possibly expected in even-even Zr nuclei, was discovered only in ^{108}Zr. The isomeric state of ^{108}Zr is proposed to be a candidate for a tetrahedral shape isomer because the energy barrier between the tetrahedral and oblate shapes is predicted to be more robust for ^{108}Zr, compared with ^{104,106}Zr. To confirm whether this isomer has the tetrahedral shape or not, the determination of the decay scheme including spins and parities, and the measurement of the band structure above the isomer, are required from future measurements.

The authors acknowledge the accelerator staff and the members of the BigRIPS team for their efforts in delivering the ^{238}U and RI beams. This work was supported by KAKENHI (50126124 and 19340074), RIKEN President Fund 2005, the DFG (EXC 153, KR 2326/2), UK STFC, and AWE plc.

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