First direct observation of isomeric decay in neutron-rich odd-odd ^{186}Ta

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Deexcitation γ rays associated with an isomeric state of ^{186}Ta were investigated. The isomers were produced in multinucleon transfer reactions between a ^{136}Xe beam and a natural W target and were collected and separated by the KEK Isotope Separation System. Two γ transitions with energies of 161.1(2) and 186.8(1) keV associated with an isomeric decay were observed for the first time. The half-life of the isomeric state of the neutral atom ^{186m}Ta was deduced as 17(2) s. Based on the comparison with the previous measurements of the isomeric state using the Experimental Storage Ring at GSI Darmstadt and the coupling of angular momenta of individual particle orbitals in odd-odd nuclei, a decay scheme of ^{186m}Ta was proposed.

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I. INTRODUCTION

Heavy neutron-rich nuclei with mass number (A) between 180 and 200 reveal a variety of properties concerning their complex nuclear structures. Shape transitions from axially symmetric prolate shapes to oblate shapes through triaxial γ softness are known to take place along isotopic chains from the neutron midshell as one approaches the closed shell at $N = 126$ due to intertwining single-particle orbitals in a deformed nuclear potential [1–4]. High-K isomeric states characteristic of this nuclear region arise from the occupation of specific single-particle orbitals with the large angular momentum projection on the symmetry axis of the axial shape [5,6]. Experimental studies of K isomers and nuclear deformations in this nuclear region are essential to enlighten the interplay between single-particle and collective degrees of freedom which formulate midshell nuclear structures. In particular, odd-odd deformed nuclei, which have complex nuclear structures related to proton-neutron interactions, sometimes exhibit long half-lives of isomeric transitions [7]. For example, the isomeric state of $^{180}_{73}\mathrm{Ta}_{107}$ at an excitation energy of 77 keV has a half-life of $> 4.5 \times 10^{16}$ yr which is much longer than the ground-state half-life 8.15 h [8]. Despite such interesting features of odd-odd nuclei in this mass region, experimental data of neutron-rich odd-odd nuclei are scarce in addition to their theoretical difficulties. Because most elements in the region are refractory, those neutron-rich isotopes are difficult to provide as a low-energy beam for their nuclear spectroscopy after their production. Recently, the KEK Isotope Separation System (KISS) [9] provides opportunities to perform spectroscopic studies of those neutron-rich refractory isotopes using multinucleon transfer (MNT) reactions [10] and in-gas laser ionization [11].

The β-decay properties of the ground state in the neutron-rich odd-odd nucleus ^{186}Ta were intensively investigated [12–14] after its discovery in 1955 [15]. However, the information on its excited states was unknown for a long time. Xu *et al.* reported the identification of an isomeric state of ^{186}Ta in 2004, which disintegrates to ^{186}W via a β decay with a half-life of 1.54(5) min [16]. Recently, the excitation energy of an isomeric state of ^{186}Ta was

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measured by the Experimental Storage Ring (ESR) at GSI Darmstadt [17]. They found the isomeric state at an energy of 336(20) keV and observed five γ-decay and three β-decay or internal-conversion (IC) events with a half-life of $3.0^{+1.5}_{-0.8}$ min for the hydrogenlike $^{186m}Ta^{72+}$ ion. Recent theoretical studies concerning the configuration of isomers in ^{186}Ta using a two-quasiparticle rotor model indicates the assignment of $K^{\pi} = 5^{-}\{\pi 7/2^{+}[404] \otimes \nu 3/2^{-}[512]\}$, $2^{-}\{\pi 7/2^{+}[404] \otimes \nu 3/2^{-}[512]\}$, and $8^{-}\{\pi 7/2^{+}[404] \otimes \nu 9/2^{-}[505]\}$ for the ground state and two isomeric states with the half-lives of 1.54 and 3.0 min, respectively [18]. Despite these efforts to understand the isomeric states both in experimental and in theoretical studies, they have not been identified yet in terms of their energies, spins, and parities by direct measurements of isomeric decays. Presently we report a new γ-ray measurement in coincidence with β rays and conversion electrons for ^{186}Ta produced by MNT reactions, which has identified a single isomeric state and its decay scheme.

II. EXPERIMENT

The experiment was performed using the KISS at the RIKEN RIBF facility, Japan. It focused on the isomeric decay of ^{186,187}Ta, and the properties of ^{187}Ta revealed through its isomeric decay were reported in Ref. [19]. ^{136}Xe beams accelerated up to 7.2 MeV/nucleon by the RIKEN Ring Cyclotron accelerator were incident on a natural tungsten target of thickness 5 μm attached to a rotating wheel. The typical beam intensity was 50 pnA on the target. Various nuclei produced by the MNT reactions between ^{136}Xe and ^{nat}W were ejected from the target and passed through a polyimide film of thickness 5 μm into a doughnut-shaped gas cell [20], which was filled with high-purity argon gas of pressure 80 kPa. After thermalization and neutralization in the argon gas, the now neutral atoms were transported to the exit of the gas cell. They were irradiated with two-color lasers for element-selective ionization with the laser resonance ionization technique just before the exit of the gas cell. The ionization scheme of tantalum was investigated before the experiment [21]. The ions ejected from the gas cell were transported by a stack of three multipole radio-frequency ion guides and were accelerated through a voltage of 20 kV. The ions with $A = 186$ were mass-separated by a dipole electromagnet with a resolving power of $A/\Delta A = 900$ and were implanted into an aluminized Mylar tape of thickness 12.5 μm.

The multisegmented proportional gas counter (MSPGC) [22,23] was placed surrounding the tape to detect β rays, x rays, and conversion electrons produced in the decay of the implanted radioactive nuclei. The MSPGC consists of two concentric layers with 16 proportional gas counter tubes in each layer. The geometry is 200 mm in height and 90 mm in outer diameter. Four high-purity germanium (HPGe) clover detectors were placed surrounding the MSPGC to detect γ rays. With the compact configuration of the experimental setup where the distances between the surfaces of the HPGe clover detectors and the tape were around 5 cm, the total absolute detection efficiency for full-energy peaks was 15% for 150-keV γ rays. The beams from KISS were pulsed by an electrostatic deflector after the dipole electromagnet. Two kinds of time cycles were used; one was 1800-s beam-on and 1800-s beam-off periods (long cycle), and the other was 300-s beam-on and 300-s beam-off periods (short cycle). The tape was moved vertically by about 30 cm after each cycle to eliminate radioactivities from both the preceding implantation and the accumulated daughter nuclei. The data were accumulated in three long cycles and 71 short cycles with ^{186}Ta beams of around 5 pps from the KISS.

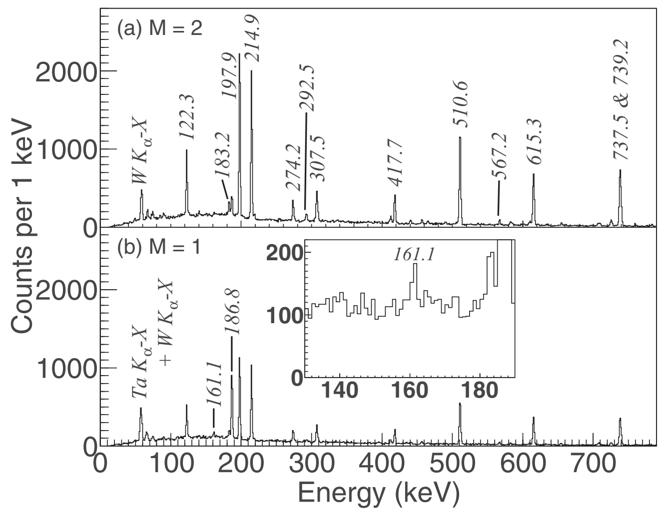

FIG. 1. γ-ray energy spectra in coincidence with the MSPGC hit patterns (a) $M = 2$ and (b) $M = 1$.

III. ANALYSIS AND RESULTS

A hit pattern analysis of the 32 gas counter tubes in the MSPGC makes it possible to separate different kinds of events. The hit pattern "$M = 2$," where one telescope (a pair of both inner and outer countertubes on the same radius vector) fires, is sensitive to energetic β rays. Figure 1(a) indicates the γ-ray energy spectrum in coincidence with the MSPGC hit pattern $M = 2$ summed for long- and short-cycle runs. A peak corresponding to the K_{α} x rays of W indicates that the transitions in the daughter nuclei of the ^{186}Ta β decay contribute when this hit pattern condition is applied. Twelve peaks labeled by energy values correspond to the known β-delayed transitions of ^{186}Ta [24]. Figure 2 shows a γ-ray time spectrum summed for those 12 transitions in coincidence with the MSPGC hit pattern $M = 2$ for the long-cycle run. The half-life was deduced by fitting the time spectrum to a combined function of a decay curve and a constant background for the decay period from 1800 to 3600 s; the fit is shown as a solid line. The half-life obtained in the fitting with a reduced $\chi^2 = 1.09$ is 10.8(5) min, which agrees with the literature value of the ^{186}Ta ground-state half-life, 10.5(3) min [24]. We could find no evidence for a decay component with a half-life around 1.5 min reported in Ref. [16].

Figure 1(b) exhibits the γ-ray energy spectrum in coincidence with the MSPGC hit pattern "$M = 1$," where only one counter tube in the inner layer fires. This detection mode is sensitive to X rays and low-energy conversion electrons

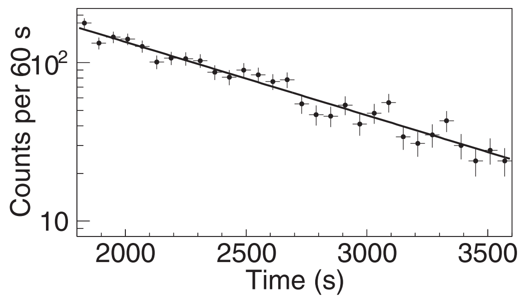

FIG. 2. γ-ray time spectrum summed for 12 transitions labeled in Fig. 1(a) in coincidence with the MSPGC hit pattern $M = 2$. The line through the data is a logarithm-likelihood fit to the decay (beam off) period from 1800 to 3600 s, yielding a half-life of 10.8(5) min.

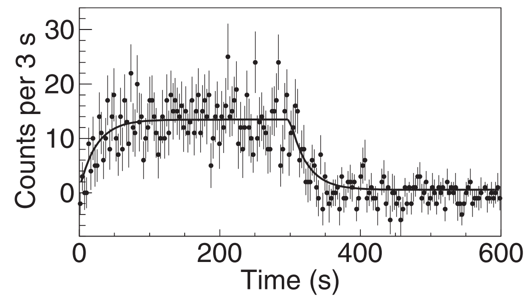

FIG. 4. Background-subtracted γ-ray time spectrum summed for 161- and 187-keV transitions in coincidence with the MSPGC hit pattern $M = 1$. The line through the data is a logarithm-likelihood fit to the growth (beam-on) and decay (beam-off) periods, each of 300-s duration, yielding a half-life of 17(2) s.

by suppressing the events owing to the energetic β rays. It is found that the peak heights corresponding to the β-delayed γ rays decrease. Furthermore, the emergence of a peak corresponding to the K_α x rays of Ta indicates that the conversion electrons fire the MSPGC with this hit pattern condition. Two previously unreported peaks at energies of 161.1(2) and 186.8(1) keV were found. The inset enlarges the spectrum at the energies around 160 keV. Figures 3(a) and 3(b) show background-subtracted γ-ray energy projections measured with gates on the 161- and 187-keV γ rays, respectively, and without MSPGC coincidence. They clearly prove the coincidence between the 187- and 161-keV γ rays, indicating that they are cascading transitions. The counts of the 161- and 187-keV γ peaks in Fig. 1(b) are 147(32) and 1845(50), respectively. Because the 161-keV peak count is more than ten times smaller than the 187-keV peak count, the 161-keV transition is considered to be highly converted implying a large multipolarity and further that the 161-keV transition is the isomeric transition. It is supported by the K_α-x-ray peak of Ta shown in the γ-ray energy spectrum in coincidence with the 187-keV γ rays in Fig. 3(b). Figure 4 shows a background-subtracted γ-ray time spectrum summed for 161- and 187-keV transitions in coincidence with the MSPGC hit pattern $M = 1$ for the short-cycle runs. The half-life was deduced by fitting the time spectrum to a combined function of a growth curve, a decay curve and a constant background; the fit is shown as a solid line. The half-life obtained in the fitting, with a reduced $\chi^2 = 0.94$, is 17(2) s, which is shorter than the half-life of the ground state of ^{186}Ta, 10.5(3) min [24]. Therefore, the newly found 161- and 187-keV γ transitions are considered to be cascading transitions from an isomeric state in ^{186}Ta with a half-life of 17(2) s. To investigate a possible β-decay mode from the isomeric state, a γ-ray energy spectrum with a time gate from 400 to 600 s was subtracted from a spectrum with a time gate from 100 to 300 s in the short-cycle runs with the coincidence of the MSPG hit pattern $M = 1$ and $M = 2$ as shown in Fig. 5. Because there are no γ-ray peaks that follow a decay curve with the half-life of 17 s, except for the 161- and 187-keV lines, the isomeric decay predominantly proceeds either with γ transition or with IC.

The energy of the isomeric state, 347.9(2) keV, was obtained from the sum of the two transition energies. It agrees with the excitation energy measured by the ESR at GSI Darmstadt, 336(20) keV [17].

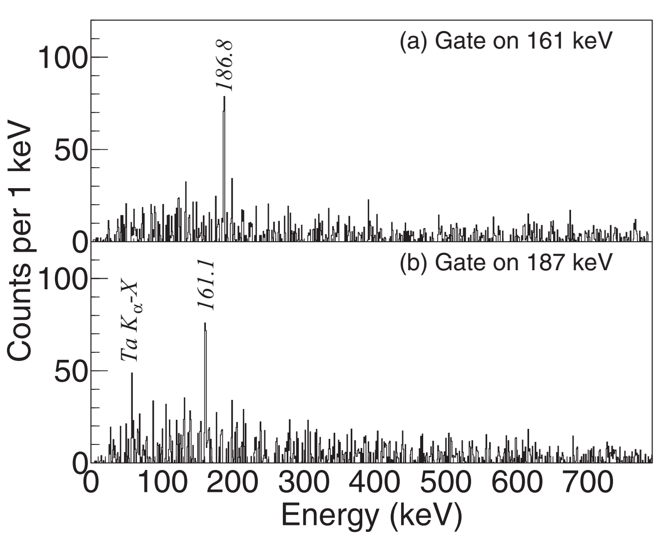

FIG. 3. Backgound-subtracted γ-ray energy projections measured with gates on (a) 161- and (b) 187-keV γ rays, respectively, and without MSPGC coincidence.

IV. DISCUSSION

The obtained half-life of the neutral ^{186m}Ta, 17(2) s, is shorter than the half-life measured in the ESR $3.0^{+1.5}_{-0.8}$ min for

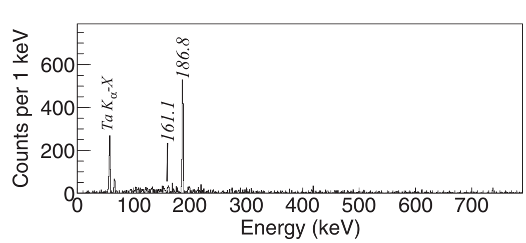

FIG. 5. γ-ray energy spectrum with a time gate from 100 s to 300 s subtracting a spectrum with a time gate from 400 s to 600 s in the short cycle runs with the coincidence of the MSPG hit pattern $M = 1$ and $M = 2$.

TABLE I. Theoretical values of total and K-conversion coefficients in the neutral atom, $\alpha_{\rm tot}$, and α_K [25], respectively, and in the hydrogenlike ion $\alpha_{K(1e)}$ [26] for the 161-keV transition in ^{186}Ta. The last column indicates the ratio of $1+\alpha_{K(1e)}$ to $1+\alpha_{\rm tot}$.

Multipolarity	$\alpha_{\rm tot}$	α_K	$\alpha_{K(1e)}$	$\frac{1+\alpha_{K(1e)}}{1+\alpha_{\rm tot}}$
$E1$	0.11	0.09	0.044	0.94
$M1$	1.1	0.9	0.47	0.70
$E2$	0.6	0.3	0.15	0.72
$M2$	6.9	5.2	2.6	0.46
$E3$	6.3	0.9	0.44	0.20
$M3$	36	20	10	0.30

a hydrogenlike ^{186m}Ta^{72+} [17]. The difference is considered to come from the components of IC, which are suppressed in the hydrogenlike ion. The ratio of the half-lives $0.09^{+0.03}_{-0.05}$ should be compared to $(1+\alpha_{K(1e)})/(1+\alpha_{\rm tot})$ where $\alpha_{\rm tot}$ and $\alpha_{K(1e)}$ are the total conversion coefficient for the neutral atom and the K-conversion coefficient for the hydrogenlike ion, respectively. Table I summarizes those calculated conversion coefficients of the 161-keV transition of ^{186}Ta for various multipolarities. The value of $(1+\alpha_{K(1e)})/(1+\alpha_{\rm tot}) = 0.20$ for the multipolarity $E3$ is the closest to the ratio of the measured half-lives. Furthermore, the ratio of the efficiency-corrected peak count of Ta-K_α x rays to the 161-keV γ rays in Fig. 3(b) is 0.70(18). It should be compared to α_K. The value of $\alpha_K\omega_K/\{1+p(K_\beta)/p(K_\alpha)\} = 0.68$ for the multipolarity $E3$, where $\omega_K = 0.952(4)$ and $p(K_\beta)/p(K_\alpha) = 0.267(4)$ [27] are the K-shell fluorescence yield and the emission probability ratio of Ta, respectively, also agrees with the measured value. Therefore, the transition deexciting the 17-s isomer is interpreted as having a multipolarity of $E3$.

The measurements in the ESR reported five γ-decay and three β-decay or IC events from the isomeric state [17]. It should be noted that the β-decay and IC events cannot be distinguished in the ESR data. When the five γ-decay events are by the $E3$ transition with $\alpha_{K(1e)} = 0.44$, 2.2(10) events are expected by IC on average. Observation of three β-decay or IC events in the ESR support the dominance of γ-decay and IC modes from the isomeric state.

In Fig. 3, the 187-keV (161-keV) γ ray is observed when it is detected by the HPGe detectors in coincidence with a conversion electron from the 161-keV (187-keV) transition detected by the MSPGC. The detected counts of the 161- and 187-keV γ rays N_{161} and N_{187} are written as

$$N_{161} = N_{\rm iso}\frac{1}{1+\alpha_{161}}\varepsilon_{\gamma 161}\frac{\alpha_{187}}{1+\alpha_{187}}\varepsilon_{\rm CE187}, \quad (1)$$

$$N_{187} = N_{\rm iso}\frac{1}{1+\alpha_{187}}\varepsilon_{\gamma 187}\frac{\alpha_{161}}{1+\alpha_{161}}\varepsilon_{\rm CE161}, \quad (2)$$

where $N_{\rm iso}$ is the number of isomeric decays and $\alpha_{161(187)}$, $\varepsilon_{\gamma 161(187)}$, and $\varepsilon_{\rm CE161(187)}$ are the conversion coefficient, the γ-ray full-energy peak efficiency, and the conversion-electron detection efficiency for the 161(187)-keV transition. Therefore, the ratio of conversion coefficients for the 161- and 187-keV transitions is related to the ratio of their detected γ-ray counts through the equation,

$$\frac{\alpha_{161}}{\alpha_{187}} = \frac{N_{187}}{N_{161}}\frac{\varepsilon_{\gamma 161}}{\varepsilon_{\gamma 187}}, \quad (3)$$

where $\varepsilon_{\rm CE187} = \varepsilon_{\rm CE161}$ is assumed. Table II summarizes the calculated conversion coefficients of the 187-keV transition for various electromagnetic multipolarities [25]. The last column indicates the ratio of the conversion coefficients for an $E3$ 161-keV transition compared to a 187-keV transition. The ratio of the efficiency-corrected γ-ray count for the 187-keV transition to the 161-keV transition is 12.8(28), which agrees with ratios for the pure $M1$ and $E2$ transitions in Table II within two standard deviations. The 187-keV transition can most probably be considered as a mixed $M1/E2$ transition. Assuming the theoretical conversion coefficient of the $E3$ 161-keV transition in Table I, $\alpha_{161} = 6.3$, the conversion coefficient of the $(M1/E2)$-mixed 187-keV transition becomes $\alpha(M1/E2) = 0.49(11)$ from the measured γ-ray intensity ratio, 12.8(28). Based on the above considerations, we propose a decay scheme of the isomeric state as shown in Fig. 6. The spin and parity of the ground state for the nearest-neighbor odd-A isotope and isotone of ^{186}Ta, ^{185}Ta, and ^{187}W, is $(7/2^+)$ and $3/2^-$, respectively [28,29]. They are considered to be due to the single-particle orbitals of the

TABLE II. Calculated conversion coefficients for the 187-keV transition of ^{186}Ta, α(187keV) [25], and the ratio compared to the conversion coefficient of the $E3$ 161-keV transition.

Multipolarity	α (187 keV)	$\frac{\alpha(161\ {\rm keV}, E3)}{\alpha(187\ {\rm keV})}$
$E1$	0.072	88
$M1$	0.74	8.5
$E2$	0.37	17
$M2$	4.0	1.6

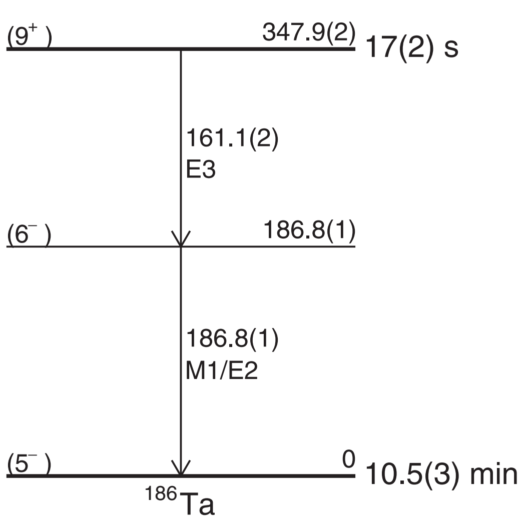

FIG. 6. Proposed decay scheme for the isomeric state in ^{186}Ta. Labels with arrows indicate the γ-ray energies in keV, and the electromagnetic multipolarities are also shown. The ground-state half-life is from Ref. [24].

73rd proton, $\pi 7/2^+[404]$, and the 113th neutron, $\nu 3/2^-[512]$, respectively. The coupling of the angular momenta of those individual particle states in ^{186}Ta gives the lower-energy state with the parallel-spin configuration $K^\pi = 5^-\{\pi 7/2^+[404] \otimes \nu 3/2^-[512]\}$ [18,30], which is considered as the ground state of ^{186}Ta. We assigned 6^- and 9^+ for the 187-keV excited state and the isomeric state, respectively, based on 5^- of the ground state and the multipolarities discussed above. The proposed decay scheme indicates that the $K^\pi = 9^+$ isomeric state decays to the first member of the rotational band of the $K^\pi = 5^-$ ground state. Sood and Gowrishankar suggested $K^\pi = 8^-$ for the isomeric state [18], however, our measurements indicate $K^\pi = 9^+$. They also suggest that a $K^\pi = 9^+$ state is possible with an antiparallel-spin two-quasiparticle configuration $\pi 7/2[404] \otimes \nu 11/2[615]$. The parallel-spin partner of the isomeric state $K^\pi = 2^+\{\pi 7/2^+[404] \otimes \nu 11/2^+[615]\}$ and the antiparallel-spin partner of the ground-state $K^\pi = 2^-\{\pi 7/2^+[404] \otimes \nu 3/2^-[512]\}$ are located at excited energies between the isomeric state and the ground state [18,30]. However, the transition from the isomeric state to those states is strongly suppressed by the large-K change $\Delta K = 7$.

Adopting the theoretical conversion coefficient 6.3 for the $E3$ 161-keV transition in Table I, its partial γ-decay half-life $T^{\gamma}_{1/2}$ becomes 2.1(2) min, which gives a reduced transition strength of $B(E3) = 1.7(2) \times 10^{-3}$ W.u. The hindrance factor $F_W = T^{\gamma}_{1/2}/T^{W}_{1/2}$, where $T^{W}_{1/2}$ is the Weisskopf estimate of the half-life, is obtained as $5.9(7) \times 10^2$. This value is consistent with the experimental range for $E3$ transitions in Refs. [31,32] with K forbiddenness $\nu = \Delta K - \lambda = 1$, where $\Delta K = 4$ and the transition multipolarity is $\lambda = 3$. The direct $M4$ 348-keV transition from the isomeric state to the ground state with $\Delta K = 4$ and $\lambda = 4$ could have a lower hindrance factor $F_W \approx 10^{-1}$ [31]. However, the corresponding partial half-life 20 min, obtained by considering the theoretical conversion coefficient 5.3 [25], is much longer than the measured half-life 17(2) s, indicating that such a direct $M4$ transition is significantly suppressed.

V. SUMMARY

We have measured γ rays associated with the isomeric decay of a 348-keV state in ^{186}Ta and found two transitions with energies of 161.1(2) and 186.8(1) keV for the first time. The fit to the time spectrum for those two transitions indicates an isomeric state with a half-life of 17(2) s. The comparison with the previous measurement obtained with the ESR at GSI Darmstadt, suggests that the 161-keV isomeric transition has an electromagnetic multipolarity of $E3$, which is followed by the 187-keV transition ($M1/E2$). A decay scheme of ^{186m}Ta was proposed by considering the coupling of the angular momenta of individual particle orbitals in odd-odd nuclei, assigning spin parities of 9^+, 6^-, and 5^- to the isomeric, excited, and ground states, respectively. It indicates that the $K^\pi = 9^+$ isomer decays to the first member in the rotational band of the $K^\pi = 5^-$ ground state. The hindrance factor for the isomeric transition is consistent with systematics [31,32] supporting the proposed decay scheme. We could not find any evidence for β decay from the observed isomer or from any other state with a half-life around 1.5 min [16] in our measurements.

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