Direct determination of the excitation energy of the quasistable isomer ^{180m}Ta

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^{180m}Ta is a naturally abundant quasistable nuclide and the longest-lived nuclear isomer known to date. It is of interest, among others, for the search for dark matter, for the development of a γ laser, and for astrophysics. So far, its excitation energy has not been measured directly but has been based on an evaluation of available nuclear reaction data. We have determined the excitation energy of this isomer with high accuracy using the Penning-trap mass spectrometer JYFLTRAP. The determined mass difference between the ground and isomeric states of ^{180}Ta yields an excitation energy of 76.79(55) keV for ^{180m}Ta. This is the first direct measurement of the excitation energy and provides a better accuracy than that of the previous evaluation value, 75.3(14) keV.

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I. INTRODUCTION

Nuclear isomers were discovered around 100 years ago [1]. Today, around 1938 isomeric ($T_{1/2} \geqslant 100$ ns) states are known [2]. The ^{180m}Ta isomer with spin-parity $J^\pi = 9^-$ and an excitation energy of around 75 keV is a unique nuclear isomer. The half-life of ^{180m}Ta is the longest of all isomeric states, longer than 4.5×10^{16} yr [2]. The isomer is much longer-living than the $J^\pi = 1^+$ ground state of ^{180}Ta, which has a half-life of 8.1 h. As a consequence, ^{180}Ta is present in nature in its isomeric state, making a 0.012 01(8)% fraction of natural tantalum.

Due to the large spin difference between the 1^+ ground state and the 9^- isomer, the internal transition to the ground state is greatly hindered. The ground state decays via β^- decay to ^{180}W and via electron capture (EC) decay to ^{180}Hf (see Fig. 1). For the ^{180m}Ta isomer, the β-decay branches are nonexistent or extremely small as there are no 8^-, 9^-, or 10^- states within the $Q_{\beta^-}/Q_{\rm EC}$ window.

^{180m}Ta has been studied in detail from many perspectives. The synthesis of ^{180m}Ta, the rarest stable isotope in nature, is still not well understood. Thermal excitation and deexcitations are also affected by the nuclear structure of ^{180m}Ta, which is a prolate nucleus with the projection of the spin on the nuclear deformation axis $K = 9$. The bands built on the $K^\pi = 9^-$ isomer and the $K^\pi = 1^+$ ground state have been studied in detail [3–6]. At high stellar temperatures, the isomer can be depopulated via thermal excitation to higher-lying states, which eventually deexcite to the short-lived ground state. The production of ^{180m}Ta via the slow neutron-capture process (s-process) as well as the p- and ν-processes has been studied [7,8]. It has been proposed as a good candidate for the development of a γ laser [3]. ^{180m}Ta also has the potential to be used in experiments devoted to the search for dark matter [9].

So far, a direct decay of ^{180m}Ta has never been observed. Its excitation energy has been evaluated based on various nuclear reactions, resulting in the values 75.3(14) keV [2] and 77.2(12) keV [10]. There are several examples in the literature where an evaluated value has turned out to be in a strong disagreement with the result achieved by direct Penning-trap mass measurements (for example, a 10σ discrepancy was found for ^{102}Pd-^{102}Ru in Ref. [11]). Thus, a direct mass measurement is of uttermost importance. In this work, we have used Penning-trap mass spectrometry to reliably measure the key spectroscopic parameter of the isomer—its excitation energy—as a mass difference between the isomeric state and the ground state of ^{180}Ta.

II. EXPERIMENTAL METHOD AND RESULTS

The measurement was performed with singly charged ^{180}Ta$^+$ and ^{180m}Ta$^+$ ions with the Penning-trap mass spectrometer JYFLTRAP [12] at the Ion Guide Isotope Separator On-Line (IGISOL) facility [13]. Both ^{180}Ta$^+$ and ^{180m}Ta$^+$ ions were simultaneously produced using nuclear reactions with a 40-MeV proton beam impinging into a $^{\rm nat}$Ta target with a thickness of about 5 mg/cm^2. The experimental reaction cross section for a production of ^{180}Ta at this proton energy is a few hundred millibarns [14], and a similar cross section is expected for the production of ^{180m}Ta according to TALYS simulations [15]. The reaction products were thermalized in the IGISOL gas cell filled with helium gas at a pressure of about 150 mbar. A large fraction of ions end up as singly charged. The ions are extracted from the IGISOL gas cell and guided via a sextupole ion guide [16] to a high-vacuum region.

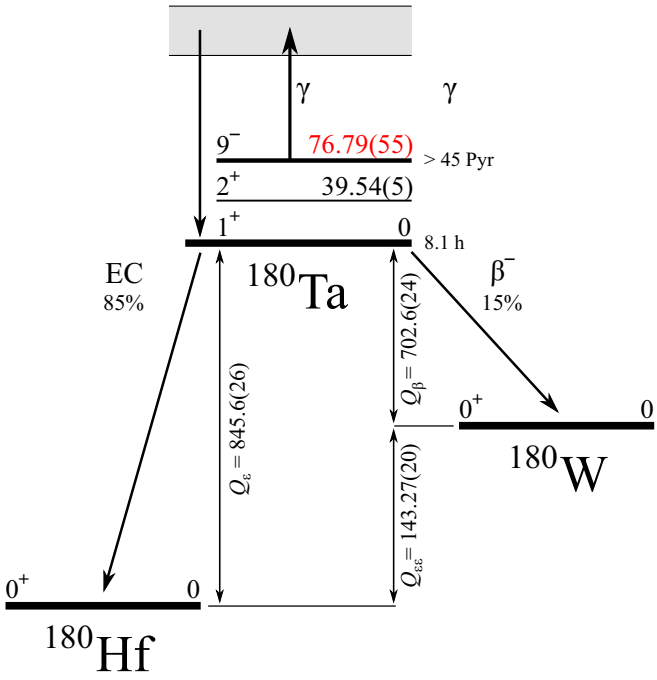

FIG. 1. Decay scheme of ^{180}Ta. Energy levels in ^{180}Ta and Q values [2] are given in keV. Due to the high spin difference, a depopulation of the naturally abundant 76.79-keV state in ^{180m}Ta can only occur by a photoexcitation into resonance intermediate states which have a decay branch into the ground state. The ground state, in turn, decays either by β^- decay to ^{180}W or by electron capture (EC) to ^{180}Hf.

Then they were accelerated to 30-keV energy with subsequent mass separation by a 55° dipole magnet. The continuous ion beam with the selected mass number of $A = 180$ was injected into a gas-filled radio-frequency quadrupole [17], where it was converted into compact ion bunches. Finally, the ion bunches were guided to the JYFLTRAP double-Penning-trap mass spectrometer.

In the mass spectrometer the ions were cooled, centered, and purified in the first preparation Penning trap by means of the mass-selective buffer gas cooling technique [18]. The process requires approximately 300 ms. Since the mass difference between the isobaric contaminant $^{180}W^+$ and the ion of interest $^{180}Ta^+$ is only about 700 keV/c^2, corresponding to about 2.5 Hz in cyclotron frequency difference, the buffer gas cooling technique is unable to resolve them. To get rid of $^{180}W^+$ ions, after cooling in the preparation trap the ions were transported to the second (measurement) trap where a 200-ms dipolar pulse at the corresponding reduced cyclotron frequency ν_+ was applied to excite these ions to large cyclotron radii, larger then the radius of the diaphragm between the traps. Then the ions were sent back to the first trap, and only ions of interest that remained cooled at the trap center passed the diaphragm and were subject to the final cooling that takes 160 ms. Finally, only $^{180}Ta^+$ ions, both the ground and the isomeric state, were transported to the measurement trap for the final determination of their mass. Tantalum is a highly reactive chemical element and easily forms ^{180}Ta $^{16}O^+$ compound in the measurement trap by interactions of the tantalum ions with residual gas particles. To get rid of this constantly forming tantalum oxide contamination, a continuous dipolar excitation at the corresponding ν_+ frequency was constantly applied over the entire duration of the measurement. Since the masses and thus the frequencies of ^{180}Ta $^{16}O^+$ and $^{180}Ta^+$ ions differ significantly, the continuous cleaning excitation pulse does not affect the actual mass measurement of $^{180}Ta^+$ states.

In Penning-trap mass spectrometry, the mass m of an ion is determined by measuring the ion's cyclotron frequency ν_c,

$$\nu_c = \frac{1}{2\pi}\frac{q}{m}B, \tag{1}$$

where q/m is the ion's charge-to-mass ratio and B is the magnetic field strength. The cyclotron frequency is measured with the phase-imaging ion-cyclotron-resonance (PI-ICR) technique that is described in detail in Refs. [19–21]. Here only the main concept and the details specific to the presented measurement are given. The trajectory of an ion in the trap is a superposition of three independent eigenmotions. One of the motions (axial motion) occurs along the magnetic field lines and the other two motions are radial motions perpendicular to the magnetic field [22]. The PI-ICR method is based on the observation of a phase evolution of the magnetron and cyclotron ion radial motions with frequencies ν_- and ν_+, respectively, by projecting the ion's position in the trap onto a position-sensitive detector. The projection of the position of the ion that performs pure magnetron and cyclotron motions are called ν_- and ν_+ phase images (spots), respectively. The positions of the phase images are described with polar angles α_- and α_+, respectively, with respect to the trap center. The cyclotron frequency ν_c calculated as a sum of two radial frequencies [23] is determined from the angle $\alpha_c = \alpha_+ - \alpha_-$ between these two phase images as

$$\nu_c = \nu_- + \nu_+ = \frac{\alpha_c + 2\pi n}{2\pi t_{\mathrm{acc}}}, \tag{2}$$

where n is the full number of revolutions during the phase accumulation time t_{acc}. For the ν_c frequency measurement of a certain ion species the phase spots and the center spot were alternately accumulated with a period of about 3 min. In turn, measurements between the two ion species were alternated after four such rounds, i.e., with a period of about 12 min.

The phase accumulation time t_{acc} of 950 ms has been used to resolve the ground and the isomeric state in ^{180}Ta. As a cross-check, a part of the data was acquired with $t_{\mathrm{acc}} = 1125$ ms (Fig. 2) to ensure that the cyclotron spots on the detector were not contaminated by any possible isobaric impurities. Since both states were simultaneously produced, had similar yields, and could not be efficiently separated prior to the actual PI-ICR measurement, for minimization of the cyclotron frequency shift due to an ion-ion interaction between these different ion species, the measurement was carried out with a low count rate of around 0.5 ions per bunch (corresponding to one detected ion per five bunches). The measurement settings for both the ground and the isomeric state were the same except for the delay and the frequency of the conversion pulse, which were adjusted to minimize the angle α_c between the magnetron and cyclotron phases for

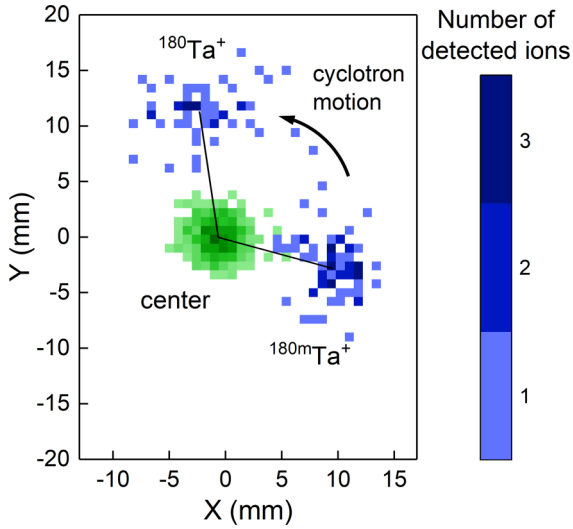

FIG. 2. Projection of the cyclotron phase image (blue bins) and the trap center (green bins) onto the position-sensitive microchannel plate detector for the ^{180}Ta$^+$ and ^{180m}Ta$^+$ ions in a single cyclotron frequency measurement with the PI-ICR method. The phase accumulation time t_{acc} was about 1125 ms.

each state. The average angle α_c in the measurements did not exceed a few degrees to minimize the systematic shifts due to the distortion of the phase projections.

Due to the intentionally low count rate, a couple of hours were required to collect reasonable statistics for an individual ν_c determination. Thus, the total measurement time was divided into 23 approximately 2-h periods. For each period the ν_c frequency for both ion species was determined and a single cyclotron frequency ratio $R = \nu_c(^{180}\text{Ta}^+)/\nu_c(^{180\text{m}}\text{Ta}^+)$ was calculated, as shown in Fig. 3. The final cyclotron-frequency ratio $\bar{R} = 1.000\,000\,458\,1(33)$ was obtained as the weighted mean of these single ratios with the uncertainty taken as the larger of the internal and external statistical uncertainties [24]. The associated Birge ratio was about a unity. The systematic uncertainties specific to the PI-ICR method are discussed in Ref. [25]. The mass-dependent systematic effects for mass doublets are negligible [26], and thus the statistical uncertainty is dominant in the final uncertainty.

The excitation energy E^* of ^{180m}Ta was calculated from the cyclotron frequency ratio as

$$E^* = [M(^{180m}\text{Ta}) - M(^{180}\text{Ta})]c^2 = (\bar{R} - 1)[M(^{180}\text{Ta}) - m_e]c^2, \quad (3)$$

where $M(^{180}\text{Ta})$ and $M(^{180m}\text{Ta})$ are the atomic masses of ^{180}Ta and ^{180m}Ta, respectively, and m_e is the electron mass. The uncertainty of the tantalum mass value $\delta M(^{180}\text{Ta}) =$ 2.1 keV/c^2 [27] does not affect the precision since the first term $(\bar{R} - 1) < 10^{-5}$. For the same reason, the binding energy of the valence electron can be neglected. Ultimately, the determined excitation energy of $^{180\text{m}}$Ta is $E^* = 76.79(55)$ keV.

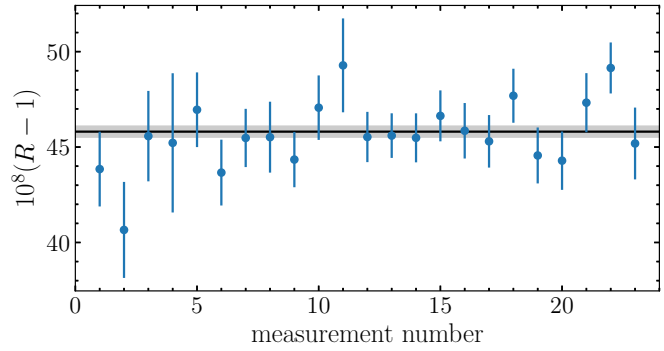

FIG. 3. Cyclotron frequency ratios $R = \nu_c(^{180}\text{Ta}^+)/\nu_c(^{180m}\text{Ta}^+)$ determined for each 2-h measurement period. The gray band represents the total 1σ uncertainty of the weighted mean frequency ratio $\bar{R} = 1.000\,000\,458\,1(33)$.

III. CONCLUSION

^{180m}Ta is an unusual isomeric state. It is the only excited nuclear state present in nature and the longest-lived isomer. It is also a state of potential interest in various fields of physics. However, its main characteristics, namely, the excitation energy, had not been measured directly. In this work, we have measured the cyclotron frequency ratio between the ^{180}Ta$^+$ ground-state and isomeric-state ions in a single experiment at the Penning-trap facility JYFLTRAP. With the obtained cyclotron frequency ratio, $R = 1.000\,000\,458\,1(33)$, we calculated the excitation energy of the longest-lived isomer ^{180m}Ta to be $E^* = 76.79(55)$ keV. Our new value is a factor of 2 more precise and in a good agreement with the previously known literature values [2,10] evaluated on the basis of various nuclear reactions with ^{180}Ta. The new accurate value improves the precision of the excitation energy and related reaction Q values relevant for stellar nucleosynthesis [3,8], for driven γ emission in ^{180}Ta and γ laser development [28], and to a dark matter search [29].

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