Precision measurement of the hyperfine structure and nuclear moments of ^{180m}Ta by laser-rf double resonance

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Hyperfine structure of the $5d^36s^2\,{}^4F_{3/2}$ ground state in ^{180m}Ta I was precisely measured by means of atomic-beam laser-rf double-resonance spectroscopy. Hyperfine-interaction constants A and B for the ground state $^4F_{3/2}$ were determined with accuracies of 3 and 10 ppm, respectively. The nuclear electric quadrupole moment was derived, as well as a precise value of the magnetic moment for ^{180m}Ta ($I^\pi=9^-$): the magnetic dipole moment is $\mu_I^{180m}=+4.825(11)\mu_N$ and the spectroscopic quadrupole moment is $Q_s^{180m}=+4.946(20)b$.

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I. INTRODUCTION

Since the discovery of the extremely long-lived and rare isotope ^{180}Ta [natural abundance 0.0123(3)%] in 1955 [1], this exotic nuclide has attracted much interest in astrophysics as well as in nuclear physics [2–10]. Wapstra and Bos first suggested that the long-lived ^{180}Ta is an isomer [5]. Later, based on the ^{181}Ta(p,d) reaction [6], it was strongly suggested that the spin value should be $I^\pi=9^-$, which results from parallel coupling of a proton orbital $9/2^-[514]_p$ and a neutron orbital $9/2^+[624]_n$. This was immediately confirmed by optical hyperfine structure measurement [7]. Data on such nuclear reactions as ^{179}Hf(^{3}He,d) and (α,t) also supported the spin assignment of $I^\pi=9^-$ [9]. Today, it is considered to be a naturally occurring spin isomer ^{180m}Ta with $T_{1/2}>1.2\times10^{15}$ yr [11] at 75.3 keV [12], i.e., the low-lying highest-spin isomer that has ever been known.

It should be pointed out, however, that determination of the spin value has not been made in an absolutely model-independent way. It is vitally important to determine model independently the nuclear spin of ^{180m}Ta in view of the nuclear and astrophysical significance; alternatively, model-independent nuclear data such as the spectroscopic electric-quadrupole moment, which as yet is not known, are of great value. Very recently, Guthöhrlein, Helmrich, and Windholz reported optical spectroscopy of Ta I using a hollow cathode discharge, whose wall was covered with 4 mg of a Ta_2O_5 sample containing ^{180m}Ta enriched to 4.1% [13]; nevertheless, hyperfine-interaction constants, from which nuclear moments are extracted, still need much improvement.

We have made a precise measurement of the hyperfine structure (HFS) of the atomic ground state $5d^36s^2\,{}^4F_{3/2}$ in ^{180m}Ta I by means of laser-rf double resonance (LRDR) spectroscopy. This paper presents an application of the LRDR method to ^{180m}Ta. Hyperfine-interaction constants A and B thus obtained yield the nuclear magnetic dipole moment μ_I and the spectroscopic quadrupole moment Q_s. Here we shall report a determination of the previously unmeasured quadrupole moment of ^{180m}Ta as well as a more precise value of its magnetic moment.

II. EXPERIMENT

The LRDR technique has been introduced to our atomic-beam laser-spectroscopy system [14], which has an argon-ion sputtering atomic-beam source to produce intense and stable atomic beams of refractory elements. Details of our experimental setup are described in Ref. [15]. First, we shall briefly describe our setup.

A metallic target of natural Ta was sputtered by the 10-keV argon ions. The laser beam from a ring dye laser

(Coherent 699-29) pumped with an argon-ion laser (Spectra Physics 171-19) was used to excite Ta atoms from the ground state $^4F_{3/2}$ to the $5d^26s^26p\ ^4D_{1/2}$ state ($\lambda=540.3$ nm); the laser output power was about 400 mW. It was split into a pump laser and a probe laser which crossed the atomic beam perpendicularly. The fluorescence induced by the probe laser was detected with a single-photon-counting photomultiplier (Hamamatsu 1333). A rf field was produced with a wire (rf loop) parallel to the atomic beam between the two lasers; the length of the rf loop was changed (3 cm to 2.5 cm) according to applied rf frequency as described in Sec. III. A synthesizer (Hewlett Packard 8341B) coupled to a power amplifier (Hewlett Packard 8348A) was used as a rf source, and the applied rf power was typically 100 mW at the terminal connected to the rf loop. To cancel the stray magnetic field in the rf region, Helmholtz coils were used in three dimensions.

III. HYPERFINE STRUCTURE OF THE GROUND STATE IN ^{180m}Ta

Figure 1 shows the laser-induced fluorescence spectrum of the 540.3-nm transition. Six strong peaks are hyperfine transitions in ^{181}Ta I; the identification of each peak is shown in Fig. 2(a). Two small peaks indicated by α and β are those in ^{180m}Ta I. We checked probable impurities (C, Cu, Fe, Mo, Nb, Ni, Si, Ti, and W) in the Ta target, and no traces of these elements were found in and out of this wavelength region. The relative intensities of the small and strong peaks were in agreement with the natural abundances of ^{180m}Ta and ^{181}Ta. The hyperfine transitions α and β were identified as shown in Fig. 2(b). The order of hyperfine multiplets of both the ground state $^4F_{3/2}$ and the excited state $^4D_{1/2}$ in ^{180m}Ta I should be similar to those in ^{181}Ta I because the sign of the magnetic dipole interaction in HFS in ^{180m}Ta I was found to be the same with that in ^{181}Ta I from relative intensities of α and β, and this interaction dominates in determination of the order of hyperfine multiplets.

Unfortunately, the other hyperfine transitions in ^{180m}Ta I were masked by strong peaks of ^{181}Ta I. It would be impossible to observe them unless enriched ^{180m}Ta is used. However, two hyperfine splittings of $\Delta\nu(F\text{-}F'=21/2\text{-}19/2)$ and $\Delta\nu(17/2\text{-}15/2)$ of the ground

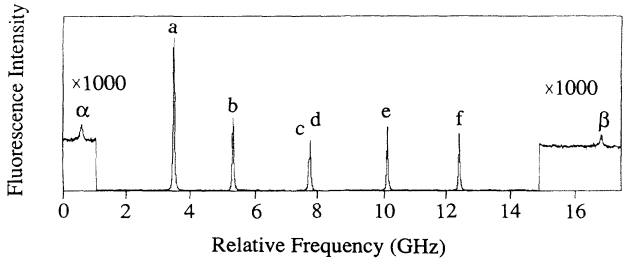

FIG. 1. Laser-induced fluorescence spectrum of the 540.3-nm ($^4F_{3/2}$-$^4D_{1/2}$) transition in Ta I.

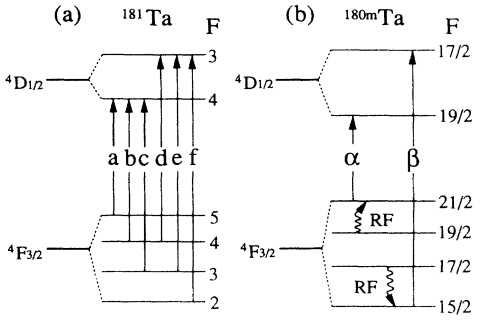

FIG. 2. Optical and rf transition schemes (a) for ^{181}Ta and (b) for ^{180m}Ta. The alphabetical indication for each optical transition corresponds to that in Fig. 1.

state $^4F_{3/2}$ in ^{180m}Ta I have been determined by observing LRDR of the α and β transitions, respectively [see Fig. 2(b)].

We had a fluorescence yield of 1500 cps without the pump laser for the peak α above a constant background of about 2500 cps. When the pump laser was on, the yield was reduced by 70% due to the optical pumping of the $F=21/2$ state. Figure 3(a) shows the LRDR spectrum for the peak α measured as a function of rf frequency; that for β is also shown in Fig. 3(b).

We used a 3-cm-long rf loop to measure the rf resonance for α, and a 2.5-cm one for β because transmission of rf signals with frequencies over 4 GHz was more efficient with the latter rf loop than with the former. Experimental linewidths were about 130 kHz for α and about 170 kHz for β, and were about 30% larger than calculated linewidths, which are 105 and 126 kHz for α

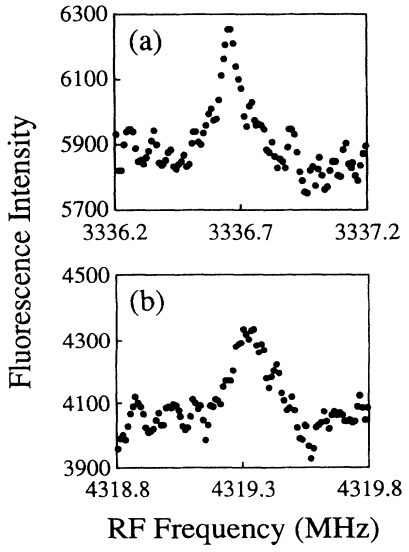

FIG. 3. rf resonance spectrum (a) for $F\text{-}F'=21/2\text{-}19/2$ and (b) for $F\text{-}F'=17/2\text{-}15/2$ in ^{180m}Ta I. Spectrum (a) was measured for the optical transition α in Fig. 1, and (b) for β.

and β, respectively; the linewidth calculation was made by taking into account the velocity distribution of sputtered atoms, length of rf loops, remnant magnetic field ($\sim$50 mG), and geometrical effect [15]. This discrepancy was caused by rf-power broadening; here we chose a rf power of 100 mW to obtain rf-resonance saturation because rf-resonance signals were so small compared with the background. According to Ref. [16], the rf-power shift of the resonance line in this case was estimated to be less than 1 kHz, which is negligibly small compared with the experimental linewidth. In fact, there were no significant shifts of the rf-resonance peaks observed when the rf power was changed in the present measurement as well as in the previous one [15].

The hyperfine splittings were thus determined to be $\Delta\nu(21/2\text{-}19/2)=3336.657(10)$ MHz and $\Delta\nu(17/2\text{-}15/2)=4319.329(10)$ MHz by making a least-squares fit to the rf-resonance line shape with the Lorentzian function. According to the well-known formalism of HFS [17], the hyperfine interaction constants A and B for the ground state $^4F_{3/2}$ were thus derived:

$$A=402.9466(10)\ \text{MHz}\ ,$$
$$B=-1533.056(15)\ \text{MHz}\ .$$

IV. NUCLEAR MOMENTS OF ^{180m}Ta

The relation between the ratio of A constants and magnetic moments is written as

$$\frac{A^{180m}}{A^{181}}=\frac{(\mu_I^{180m}/I^{180m})}{(\mu_I^{181}/I^{181})}(1+{}^{180m}\Delta^{181})\ , \tag{1}$$

where ${}^{180m}\Delta^{181}$ is defined by ${}^{180m}\Delta^{181}=\varepsilon^{180m}-\varepsilon^{181}$; ε is the Bohr-Weisskopf correction (hyperfine structure anomaly) [18]. The ratio $A^{180m}/A^{181}=0.791\,52(2)$ was obtained from our value of A^{180m} and the known A^{181} [19]. The factor ${}^{180m}\Delta^{181}\ll 1$ is expected in the present case because the s-electron shell is closed in the ground-state configuration $5d^36s^2$. The values $|{}^A\Delta^{A'}|$ measured for the $5d^N6s^2$ configuration in the neighboring elements are smaller than 0.0024 [20–23]. Therefore, for safety estimation we assume ${}^{180m}\Delta^{181}=0.0\pm0.0024$; then we have the magnetic moment of ^{180m}Ta:

$$\mu_I^{180m}=+4.825(11)\mu_N\ , \tag{2}$$

using $\mu_I^{181}=+2.3705(7)\mu_N$ [24]. This is in agreement with that given by Burghardt *et al.* [7] within their experimental error; the accuracy has been improved by a factor of 5.

According to the additivity theorem [25], a magnetic moment is expressed as

$$\mu_I=\frac{I}{(I+1)}(g_R+g_{K_p}K_p+g_{K_n}K_n) \tag{3}$$

for a two-particle state such as ^{180m}Ta, where K_p and K_n are K quantum numbers for an unpaired proton and neutron, respectively. We have a calculated magnetic moment $\mu_I^{\text{calc}}=+4.82(9)\mu_N$ for ^{180m}Ta with the configuration $\{9/2^-[514]_p,9/2^+[624]_n\}9^-$ using g_R, g_{K_p}, and g_{K_n} measured for the neighboring isotopes [21,26,27]. The calculated value is in good agreement with the experimental one. This strongly supports the assignments of the nuclear spin and the Nilsson orbital configuration in previous works [6,7,9,13].

The ratio of B constants for ^{180m}Ta and ^{181}Ta should be equal to the ratio of spectroscopic quadrupole moments of these nuclides, because the Sternheimer factor R does not change between isotopes:

$$\frac{B^{180m}}{B^{181}}=\frac{Q_s^{180m}}{Q_s^{181}}\ , \tag{4}$$

where $B^{180m}/B^{181}=+1.514\,50(3)$ is obtained from our value of B^{180m} and B^{181} given previously [19]. The quadrupole moment Q_s^{181} has been measured by several groups, and those are compiled in Ref. [24]. Using a weighted average, $Q_s^{181}=+3.266(13)b$, we have the spectroscopic quadrupole moment of ^{180m}Ta:

$$Q_s^{180m}=+4.946(20)b\ . \tag{5}$$

To estimate the intrinsic quadrupole moment Q_0, we employ the well-known formula based on the strong-coupling limit [28,29]

$$Q_s=\frac{3K^2-I(I+1)}{(I+1)(2I+3)}Q_0\ , \tag{6}$$

where $K=I=9$ for ^{180m}Ta; thus we have $Q_0^{180m}=+6.79(3)b$ and $\beta_2=0.268(1)$. The Q_0^{180m} value is close to $Q_0^{180m}=+6.80(13)b$ [30] for ^{181m}Ta, which has a $9/2^-[514]_p$ proton. It should be noted that the latter was determined from the Mössbauer effect, i.e., model-independent Q_s of ^{181m}Ta. Because of this, the present result almost provides evidence for the choice of the $9/2^-[514]_p$ orbital in ^{180m}Ta.

V. SUMMARY

We have measured a laser-rf double resonance in ^{180m}Ta I and determined the nuclear quadrupole moment; a more precise value of the magnetic moment has also been obtained. In the framework of the deformed nuclear model that has been well established in this mass region, the only choice of the orbital configuration is $\{9/2^-[514]_p,9/2^+[624]_n\}9^-$ in order to describe the observed magnetic dipole and electric quadrupole moments; otherwise we have to assume a sudden change in particle orbitals or nuclear deformation in ^{180m}Ta. If we observed, for instance, one of the masked hyperfine transitions in the present measurement, we could determine the spin of ^{180m}Ta independently of the nuclear model. In view of the significance of this exotic nuclide in astrophysics and nuclear physics, it would be very desirable to determine the spin value of ^{180m}Ta independently of the nuclear model.

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