

## Isotope shift in the tantalum atomic spectrum

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Optical isotope shifts have been measured in 11 lines of the neutral-tantalum spectrum. The tantalum atoms were introduced into a hollow cathode discharge, whose cathode wall was covered with 4 mg of a Ta<sub>2</sub>O<sub>5</sub> sample containing <sup>180</sup>Ta enriched to 4.1%. Using laser excitation with either optogalvanic or laser-induced fluorescence detection, the hyperfine constants of the levels involved and the shifts of the centers of gravity between <sup>180</sup>Ta and <sup>181</sup>Ta could be determined. The measurements show differences in the level isotope shifts within the terms  $a^6D$ ,  $z^6D^o$ ,  $y^4D^o$ , and  $z^4P^o$ :  $\Delta T\{5d^4(a^5D)6s a^6D_{3/2} - 5d^4(a^5D)6s a^6D_{1/2}\} = 527(35)$  MHz,  $\Delta T\{5d^36s(a^5F)6p z^6D_{3/2}^o - 5d^36s(a^5F)6p z^6D_{1/2}^o\} = 355(35)$  MHz,  $\Delta T\{5d^36s(a^3F)6p y^4D_{3/2}^o - 5d^36s(a^3F)6p y^4D_{1/2}^o\} = -591(30)$  MHz, and  $\Delta T\{5d^26s^2(a^3P)6p z^4P_{3/2}^o - 5d^26s^2(a^3P)6p z^4P_{1/2}^o\} = 58(30)$  MHz, which are caused most probably by strong heretofore unknown perturbations.

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### I. INTRODUCTION

<sup>180</sup>Ta<sub>107</sub> is a very rare isotope, whose abundance in the natural isotopic mixture amounts to only 0.0123% [1]. The ground state of this (odd-proton and -neutron number) nucleus is unstable (lifetime  $\tau=8.152$  h); the still nonzero natural abundance originates from the existence of a metastable state 75.3 keV above the ground state with  $\tau > 1.2 \times 10^{15}$  years [2]. The nuclear-spin quantum number  $I=9$  listed in [2] agrees with an earlier determination from optical hyperfine splittings [3,4]. The isotope shifts of two lines measured by means of laser-atomic-beam spectroscopy using natural tantalum are given by Harzer [4] even though they have not yet been published elsewhere.

It should be pointed out that for a relatively large number of the tantalum energy levels, the hyperfine-structure constants are unknown, and that there exist some differences in the notation of the energy levels, given in the tables of Moore [5] and the papers of Klinkenberg, van den Berg, and van den Bosch [6] and van den Berg, Klinkenberg, and van den Bosch [7]. Isotope-shift measurements are a very useful tool in finding out the electron configurations, and we could find some irregularities looking at the shifts of the investigated levels. If not otherwise specified, we use the notation of Moore [5] for naming the levels.

### II. EXPERIMENT

The experimental method used is well established in hyperfine-structure investigations and has been described in more detail, e.g., in Refs. [8] and [9]. A hollow cathode discharge (the carrier gas was argon) generated free tantalum atoms by the well-known sputtering and

dissociation mechanisms at and near the inner wall of the cathode, made of pure aluminum and covered with Ta<sub>2</sub>O<sub>5</sub>. The discharge was also populating the lower levels of the interesting lines sufficiently. Generally, the laser resonance with the Ta lines should affect the impedance of the discharge. By chopping the tunable, exciting laser beam, we were able to use the optogalvanic method to record the spectra with a lock-in amplifier. Of course, the fluorescence light stemming from the upper level of the laser resonant transitions bears the modulation the laser is prepared to, and in this way laser-induced fluorescence spectra could be obtained by the lock-in technique using a 1-m Czerny-Turner monochromator tuned to a proper fluorescence line.

The investigated transitions and the detection method had to be chosen so that they were adequate to the experimental circumstances and to the small amount of the enriched Ta<sub>2</sub>O<sub>5</sub> sample. In a pilot test the cathode wall was covered with a small amount (5 mg) of natural Ta<sub>2</sub>O<sub>5</sub> to ensure the best experimental conditions with regard to signal-to-noise ratio, burning time, etc. Finally, the same preparation was done with 4-mg Ta<sub>2</sub>O<sub>5</sub> electromagnetically enriched to 4.1% <sup>180</sup>Ta (purchased from Oak Ridge National Laboratory, Oak Ridge, TN). Nevertheless, the isotope ratio was 96:4, and we were expecting only small peaks caused by <sup>180</sup>Ta. As known to us from former measurements [10], the hyperfine splittings in the <sup>181</sup>Ta lines are relatively large, and for small values of  $J$  the components are in most cases separated much wider than their Doppler width (overall splittings up to 20 GHz). Assuming isotope shifts that are small as compared with the hyperfine splitting, we expected the components of <sup>180</sup>Ta to be overlaid by the much stronger components of <sup>181</sup>Ta, and therefore we selected transitions between levels with small values of  $J$  ( $\leq \frac{3}{2}$ ) in order (i) to get a small number of components and (ii) to have a good chance to find components of <sup>180</sup>Ta separated completely from the stronger components of <sup>181</sup>Ta. In order to reach intensities that are as high as possible, we have done all experi-

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ments by Doppler-limited laser spectroscopy. The wavelength range of the actively stabilized dye laser was between 630 and 691 nm using DCM as laser dye and between 580 and 605 nm using rhodamine 6G, respectively. The investigated transitions are listed in Table I. For each transition the relative frequency positions of the hyperfine components of both isotopes were determined from several scans (typically ten) and the evaluation of the hyperfine-structure constants and the isotope shifts was performed using the average of these positions.

It was especially important in our investigations to obtain a good signal-to-noise ratio detecting the weak components of  $^{180}\text{Ta}$ . Considering the optogalvanic detection, the laser-generated signal is proportional to the number density of the lower level of the resonant transition, multiplied by a specific sensitivity factor which takes into account the change of the impedance of the discharge. The sensitivity factor depends in a microscopically complicated manner on the collision processes between all particles embedded in the plasma, because the laser-induced population changes between the resonant levels not only vary the respective ionization rates but also lead to a new balance point of the local thermodynamic equilibrium whose evolution in time is very complex. Moreover, the sensitivity factor depends not only on the transition under study but also on the carrier gas used.

By modulating the intensity of the laser light with a suitable frequency, the impedance of the discharge is also modulated. In every case the variation of the impedance is very small as compared with the impedance itself. Therefore, it is extremely important to have the discharge at a very low noise level.

In view of a  $\Lambda$ -type laser-induced fluorescence detection, the modulated part of the fluorescence light is proportional to the laser-induced change in the number density of the upper level of the resonant transition, multiplied by the branching ratio into the selected fluorescence transition. The dc part stems from the population of the upper level via the gaseous discharge. The best lines for fluorescence detection do not have a very strong intensity produced by the discharge and show an advantageous branching ratio compared with the laser transition. In this case the change in the fluorescence intensity produced by resonant laser radiation is large, and the signal-to-noise ratio may be larger than is achievable by detecting optogalvanically.

Lacking a simple and universal estimation procedure, the choice between the two aforementioned detection methods, i.e., laser-induced fluorescence or optogalvanic detection, was decided experimentally by looking at the signal-to-noise ratio.

### III. EVALUATION OF THE SPECTRA

The evaluation of all spectra in this work is based on using  $I^{180}=9$ . Spectral lines which are transitions from  $J=\frac{1}{2}$  to  $J'=\frac{1}{2}$  show a simple hyperfine pattern consisting only of four hyperfine components. The quantum numbers  $F$  of the two hyperfine levels belonging to each fine-structure level are therefore  $I+\frac{1}{2}$  and  $I-\frac{1}{2}$  and the sepa-

TABLE I. Investigated lines and detection method (LIF, laser-induced fluorescence; OG, optogalvanic). The configurations are taken from the tables of Moore [5].

Line $\lambda$ (nm)	Lower level			Upper level			Detection				
	Configuration	Designation	$\sigma$ ( $\text{cm}^{-1}$ )	Configuration	Designation	$\sigma$ ( $\text{cm}^{-1}$ )	Method	Line $\lambda$ (nm)	Configuration	Designation	$\sigma$ ( $\text{cm}^{-1}$ )
584.394	$5d^4(a^5D)6s$	$a^6D_{1/2}$	9758.97	$5d^26s^2(a^3P)6p$	$z^4P_{1/2}^o$	26866.05	LIF	591.895	$5d^4(a^5D)6s$	$a^6D_{3/2}$	9975.81
591.895	$5d^4(a^5D)6s$	$a^6D_{3/2}$	9975.81	$5d^26s^2(a^3P)6p$	$z^4P_{1/2}^o$	26866.05	LIF	584.394	$5d^4(a^5D)6s$	$a^6D_{1/2}$	9758.97
593.976	$5d^4(a^5D)6s$	$a^6D_{1/2}$	9758.97	$5d^26s^2(a^3P)6p$	$z^4P_{3/2}^o$	26590.03	LIF	406.724	$5d^36s^2$	$a^4F_{5/2}$	2010.10
602.072	$5d^4(a^5D)6s$	$a^6D_{1/2}$	9758.97	$5d^36s(a^5F)6p$	$y^4D_{3/2}^o$	26363.69	LIF	492.600	$5d^36s^2$	$a^4P_{3/2}$	6068.91
634.597	$5d^4(a^5D)6s$	$a^6D_{1/2}$	9758.97	$5d^36s(a^5F)6p$	$y^4D_{1/2}^o$	25512.63	LIF	513.647	$5d^36s^2$	$a^4P_{1/2}$	6049.42
643.454	$5d^4(a^5D)6s$	$a^6D_{3/2}$	9975.81	$5d^36s(a^5F)6p$	$y^4D_{1/2}^o$	25512.63	LIF	513.647	$5d^36s^2$	$a^4P_{1/2}$	6049.42
667.369	$5d^4(a^5D)6s$	$a^6D_{1/2}$	9758.97	$5d^36s(a^5F)6p$	$z^6D_{3/2}^o$	24739.03	OG				
677.172	$5d^4(a^5D)6s$	$a^6D_{3/2}$	9975.81	$5d^36s(a^5F)6p$	$z^6D_{3/2}^o$	24739.03	OG				
677.424	$5d^4(a^5D)6s$	$a^6D_{1/2}$	9758.97	$5d^36s(a^5F)6p$	$z^6D_{1/2}^o$	24516.69	LIF	541.913	$5d^36s^2$	$a^4P_{3/2}$	6068.91
687.526	$5d^4(a^5D)6s$	$a^6D_{3/2}$	9975.81	$5d^36s(a^5F)6p$	$z^6D_{1/2}^o$	24516.69	LIF	541.913	$5d^36s^2$	$a^4P_{3/2}$	6068.91
690.205	$5d^4(a^5D)6s$	$a^6D_{1/2}$	9758.97	$5d^36s(a^5F)6p$	$z^6F_{3/2}^o$	24243.42	OG				
577.675	$5d^36s^2$	$a^4P_{1/2}$	6049.42	$5d^36s(a^5F)6p$	Harzer [4] $z^6F_{1/2}^o$	23355.41	LIF	735.286	$5d^4(a^5D)6s$	$a^6D_{1/2}$	9758.97
566.490	$5d^36s^2$	$a^4F_{5/2}$	2010.10	$5d^26s^2(a^3F)6p$	$z^4D_{3/2}^o$	19657.78	LIF	734.641	$5d^36s^2$	$a^4P_{1/2}$	6049.42

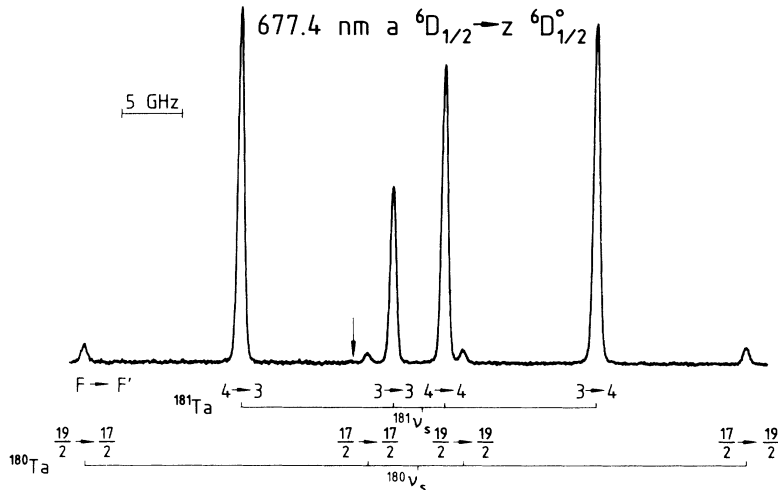


FIG. 1. Typical recording of the line  $\lambda=677.4$  nm (single scan; detection method: laser-induced fluorescence at  $\lambda=541.9$  nm). All hyperfine components are well separated. The intensity ratio of the components of  $^{181}\text{Ta}$  to those of  $^{180}\text{Ta}$  is due to the abundance ratio of 96:4 in our enriched sample. Because of the large total splitting of the  $^{180}\text{Ta}$  isotope (ca. 50 GHz), the complete pattern had to be recorded in two subsequent laser scans. The arrow indicates the frequency at which the two scans are connected in the figure (overlap is not shown).

ration between these hyperfine levels is  $(I + \frac{1}{2})|A|$  ( $A$  is the hyperfine-structure constant of the level). The total hyperfine splitting of the line is then

$$\Delta\nu_{\text{tot}} = (I + \frac{1}{2})(|A| + |A'|).$$

By measuring  $\Delta\nu_{\text{tot}}$ —for instance, in the line  $\lambda=677.4$  nm ( $a^6D_{1/2} - z^6D_{1/2}$ )—for both isotopes we found the ratio

$$\Delta\nu_{\text{tot}}^{180} / \Delta\nu_{\text{tot}}^{181} = 1.864(1).$$

This is in very good agreement with the corresponding value 1.86(2) published by Burghardt *et al.* [3] taken from the hyperfine structure of the line  $\lambda=577.7$  nm.

Figure 1 shows a typical recording of the hyperfine structure of the line  $\lambda=677.4$  nm. The four hyperfine components of  $^{180}\text{Ta}$  are well separated from the much stronger hyperfine components of  $^{181}\text{Ta}$ . With this line it was possible to determine the positions of all components of the hyperfine pattern and to calculate  $A$ ,  $A'$ , and  $\nu_s$  (the center-of-gravity frequency) for both isotopes. The isotope shift (IS) of the line is defined as

$$\Delta\nu_{\text{IS}}(181-180) = \nu_s^{181} - \nu_s^{180}.$$

During the data evaluation, we have used a very precise value of

$$A^{181}(a^6D_{1/2}) = 3100.3(3) \text{ MHz}$$

taken from Doppler-free investigations in the line  $\lambda=602.1$  nm ( $a^6D_{1/2} - y^6D_{3/2}$ ) [10]. (This  $A$  value was also introduced in the evaluation of the lines  $\lambda=634.6$  and  $667.4$  nm.)

The line  $\lambda=687.5$  nm ( $a^6D_{3/2} - z^6D_{1/2}$ ) splits into six hyperfine components. The center frequencies of only three components of the  $^{180}\text{Ta}$  pattern could be fitted; the remaining components are overlaid by  $^{181}\text{Ta}$  components. In order to increase the accuracy, the  $A$  values of  $z^6D_{1/2}$  for both isotopes were taken from the line  $\lambda=677.4$  nm. With this assumption, it was possible to determine the  $A$  and  $B$  values of  $a^6D_{3/2}$  of  $^{181}\text{Ta}$  with higher accuracy and to evaluate these hyperfine constants and the center-of-gravity frequency using only the three resolved components of  $^{180}\text{Ta}$ . The  $A$  and  $B$  values of  $a^6D_{3/2}$  of both

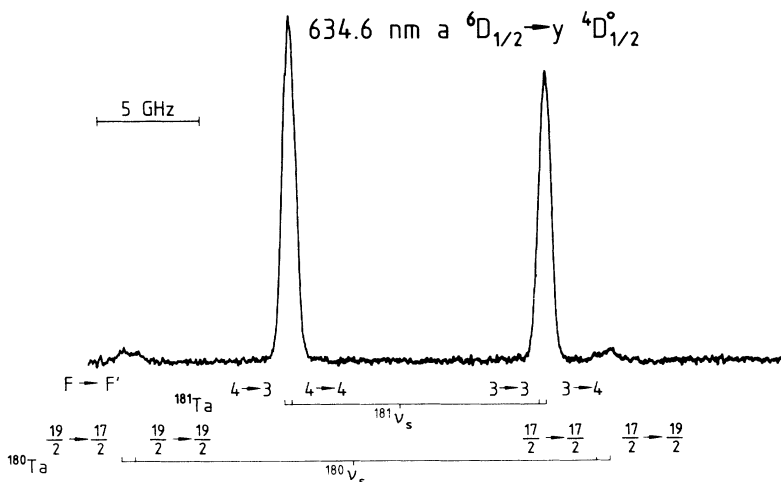


FIG. 2. Typical recording of the line  $\lambda=634.6$  nm (single scan; detection method: laser-induced fluorescence at  $\lambda=513.6$  nm). The  $A$  factor of  $^{181}\text{Ta}$   $y^4D_{1/2}$  amounts to only 89.5 MHz; therefore, the Doppler broadening is larger than the separation of the doublets. In the case of  $^{180}\text{Ta}$ , all hyperfine components are well separated, and the  $A$  factor of  $^{180}\text{Ta}$   $y^4D_{1/2}$  could be determined directly.

TABLE II. Hyperfine-structure constants  $A$  and  $B$ , line isotope shifts and differences of level isotope shifts (LIS<sup>1</sup>:  $a^6D_{3/2}-a^6D_{1/2}$ ; LIS<sup>2</sup>:  $z^6D_{3/2}-z^6D_{1/2}$ ; LIS<sup>3</sup>:  $z^4P_{3/2}-z^4P_{1/2}$ ; LIS<sup>4</sup>:  $y^4D_{3/2}-y^4D_{1/2}$ ).

Line $\lambda$ (nm)	Isotope	Isotope shift (MHz)	Lower level			Upper level						
			Designation	$A$ (MHz)	$B$ (MHz)	LIS <sup>1</sup> (MHz)	Designation	$A$ (MHz)	$B$ (MHz)	LIS <sup>2</sup> (MHz)	LIS <sup>3</sup> (MHz)	LIS <sup>4</sup> (MHz)
584.394	181		$a^6D_{1/2}$	3100.3 <sup>a</sup>	0	0	$z^4P_{1/2}$	2445.14 <sup>b</sup>	0			
	180	1207(20)		2437.1 <sup>b</sup>	0	0		1924.1 <sup>b</sup>	0			
591.895	181		$a^6D_{3/2}$	1214.6 <sup>b</sup>	-11 <sup>b</sup>	0	$z^4P_{1/2}$	2445.14 <sup>b</sup>	0			
	180	664(40)		958.4 <sup>b</sup>	-19 <sup>b</sup>	543(35)		1924.1 <sup>b</sup>	0			
593.976	181		$a^6D_{1/2}$	3100.3 <sup>a</sup>	0	0	$z^4P_{3/2}$	1445.7 <sup>b</sup>	230.7 <sup>b</sup>			
	180	1265(20)		2437.1 <sup>b</sup>	0	0		1137.6 <sup>b</sup>	387.0 <sup>b</sup>		58(30)	
602.072	181		$a^6D_{1/2}$	3100.3 <sup>a</sup>	0	0	$y^4D_{3/2}$	635.4 <sup>b</sup>	390.2 <sup>b</sup>			
	180	1682(20)		2437.1 <sup>b</sup>	0	0		500.0 <sup>b</sup>	587.0 <sup>b</sup>			-591(30)
634.597	181		$a^6D_{1/2}$	3100.3 <sup>a</sup>	0	0	$y^4D_{1/2}$	89.5 <sup>b</sup>	0			
	180	2273(20)		2437.1 <sup>b</sup>	0	0		70.3 <sup>b</sup>	0			
643.454	181		$a^6D_{3/2}$	1214.6 <sup>b</sup>	-11 <sup>b</sup>	0	$y^4D_{1/2}$	89.5 <sup>b</sup>	0			
	180	1710(75)		958.4 <sup>b</sup>	-19 <sup>b</sup>	563(95)		70.3 <sup>b</sup>	0			
667.369	181		$a^6D_{1/2}$	3100.3 <sup>a</sup>	0	0	$z^6D_{3/2}$	1414.4	636.6			
	180	687(20)		2437.1 <sup>b</sup>	0	0		1117.1	966.6		350(30)	
667.172	181		$a^6D_{3/2}$	1214.6 <sup>b</sup>	-11 <sup>b</sup>	0	$z^6D_{3/2}$	1413.4	631.4			
	180	172(20)		958.4 <sup>b</sup>	-19 <sup>b</sup>	515(40)		1113.1	977.1		361(40)	
677.424	181		$a^6D_{1/2}$	3100.3 <sup>a</sup>	0	0	$z^6D_{1/2}$	4155.0	0			
	180	337(10)		2437.1 <sup>b</sup>	0	0		3257.1	0			
687.526	181		$a^6D_{3/2}$	1214.6 <sup>b</sup>	-11	0	$z^6D_{1/2}$	4155.0 <sup>b</sup>	0			
	180	-189(20)		958.4 <sup>b</sup>	-19	526(30)		3257.1 <sup>b</sup>	0			
690.205	181		$a^6D_{1/2}$	3100.3 <sup>a</sup>	0	0	$z^6F_{3/2}$	594.0	-278.0			
	180	827(30)		2437.1 <sup>b</sup>	0	0		466.9 <sup>b</sup>	-426.4 <sup>b</sup>			
577.675	181		$a^4P_{1/2}$	884(2)	0	Harzer [4]	$z^6F_{1/2}$	-1784(2)	0			
	180	921(10)		693(4)	0			-1396(4)	0			
566.490	181		$a^4F_{5/2}$	313(1)	-830(14)	0	$z^4D_{3/2}$	643.5(4)	-279(5)			
	180	894(8)		248.2(4)	-1247(17)	0		507.2(5)	-415(11)			

<sup>a</sup>Introduced from  $\lambda = 602.1$  nm [10].

<sup>b</sup>Introduced from other lines, respectively, calculated using  $A$ - or  $B$ -value ratios (see the text for details).

$^{181}\text{Ta}$  and  $^{180}\text{Ta}$  were then introduced in the evaluation of the lines  $\lambda=677.2$  and  $643.5$  nm.

The upper level of the lines  $\lambda=634.6$  and  $643.5$  nm,  $y^4D_{1/2}^o$ , has a very small  $A$  value. Therefore, the four hyperfine-structure (hf) components of  $^{181}\text{Ta}$  are arranged in two groups of doublets which show only a broadened linewidth. Fortunately, the hyperfine pattern of  $^{180}\text{Ta}$  gives insight into the structure of the corresponding doublets in the line  $\lambda=634.6$  nm (see Fig. 2). Thus we could determine the  $A$  value of  $^{180}\text{Ta}$  ( $y^4D_{1/2}^o$ ) directly. For  $^{181}\text{Ta}$ , we calculated the  $A$  factor, neglecting the hyperfine anomaly, by taking the  $A$ -value ratio  $A^{180}$  ( $a^6D_{1/2}$ )/ $A^{181}$  ( $a^6D_{1/2}$ ) of the line  $\lambda=677.4$  nm and used these values for the determination of the IS's of both lines.

The lines  $\lambda=667.4$  and  $677.2$  nm have a common upper level,  $z^6D_{3/2}^o$ . Using for the lower levels of both

isotopes the formerly determined  $A$  and  $B$  values of  $a^6D_{1/2}$  and  $a^6D_{3/2}$ , we could determine the  $A$  and  $B$  values of the upper levels of both isotopes on both lines independently from each other, having a good test of consistency in this way, as can be seen by comparing the results in Table II.

In the line  $\lambda=690.2$  nm we were not able to determine  $A^{180}$  and  $B^{180}$  values because the number of observable components of this isotope was too small. Nevertheless, calculating the hyperfine constants by use of the  $A$ -value ratio and the  $B$ -value ratio observed at the lines  $\lambda=667.4$  and  $677.2$  nm, we could determine the IS of this line, too.

The situation in the case of the four lines lying in the yellow spectral region ( $\lambda=584.4, 591.9, 594.0,$  and  $602.1$  nm) was somewhat different. In each of these patterns, we could localize only one or two hyperfine components of the  $^{180}\text{Ta}$  isotope. The other components were either

TABLE III. Hyperfine-structure constants  $A$  and  $B$  of  $^{181}\text{Ta}$  in comparison with results of other authors.

Designation	$\sigma$ (cm $^{-1}$ )	$A^{181}$ (MHz)	$B^{181}$ (MHz)	Source
Even-parity levels				
$a^4F_{5/2}$	2010.10	313(1)	-830(14)	[4]
		313.469(1)	-834.797(12)	[11]
		312	-1008	[12]
		324	-252	[13]
$a^4P_{1/2}$	6049.42	884(2)	0	[4]
		884.115(3)	0	[11]
		873	0	[14]
$a^6D_{1/2}$	9758.97	3100(3)	0	This work
		3109	0	[11]
		3111	0	[14]
		3100.3(6)	0	[10]
		3102(2)	0	[15]
$a^6D_{3/2}$	9975.81	1214.6(12)	-11(6)	This work
		1214	-18	[11]
		1200	-67	[14]
		1214.017(62)	-13.85(46)	[15]
Odd-parity levels				
$z^4D_{3/2}^o$	19 657.78	643.5	-279(5)	[4]
$z^6F_{1/2}^o$	23 355.41	-1784(2)	0	[4]
		-1822.5	0	[12]
$z^6F_{3/2}^o$	24 243.42	594(6)	-278(5)	This work
$z^6D_{1/2}^o$	24 516.69	4155(4)	0	This work
		4238(16)	0	[16]
		4147.5	0	[12]
		4132.5	0	[13]
$z^6D_{3/2}^o$	24 739.03	1413.7(14) <sup>a</sup>	634(13) <sup>a</sup>	This work
		1421.7(27)	652(22)	[16]
$y^4D_{1/2}^o$	25 512.63	89.5(50)	0	This work
		64.6(56)	0	[16]
		95.3(15)	0	[17]
$y^4D_{3/2}^o$	26 363.69	635.4(8)	390.2(50)	[10]
$z^4P_{1/2}^o$	26 866.05	2445.2(15)	0	[10]
$z^4P_{3/2}^o$	26 590.03	1445.7(8)	231(4)	[10]

<sup>a</sup>Averaged values from the lines  $\lambda=667.4$  and  $677.2$  nm.

overlaid by the much stronger components of the  $^{181}\text{Ta}$  isotope or too weak to observe. So we could not determine the hyperfine-structure constants  $A$  and  $B$  of  $^{180}\text{Ta}$  directly from the spectra. Instead, we used the ratios of the  $A$  and  $B$  constants (mean values determined from the red lines,  $A^{180}/A^{181}=0.7869$  and  $B^{180}/B^{181}=1.504$ ) together with the respective hyperfine constants of the  $^{181}\text{Ta}$  isotope, fortunately known from former experiments [10], to calculate the hyperfine constants of the  $^{180}\text{Ta}$  isotope. Using these constants, we calculated the  $^{180}\text{Ta}$  hyperfine pattern for each of the four yellow lines, and in this way we were able to identify the observed spectral structures. Determining the frequency differences between only one or two  $^{180}\text{Ta}$  components and the  $^{181}\text{Ta}$  components, we could evaluate the isotope shifts of these lines, too.

The results, concerning the hyperfine constants and the isotope shifts of all investigated lines, are collected in Table II.

In Table III, the values of the hyperfine constants of  $^{181}\text{Ta}$  are compared with those of other authors. In Table IV, the corresponding values of  $^{180}\text{Ta}$  are listed, together with the  $A$ - and  $B$ -value ratios  $A^{180}/A^{181}$  and  $B^{180}/B^{181}$  taken from the observed  $A$  and  $B$  values. The results of Harzer [4] are included in all of the tables.

#### IV. DISCUSSION

Figure 3 shows part of the level scheme of Ta with the investigated lines, including the two lines from the work of Harzer [4]. Some strange behavior of the observed IS has to be stated.

The two lines  $\lambda=677.4$  nm ( $a^6D_{1/2}-z^6D_{1/2}^o$ ) and  $\lambda=687.5$  nm ( $a^6D_{3/2}-z^6D_{1/2}^o$ ) have a common upper level. The lower levels are assigned to the

$5d^4(a^5D)6s a^6D$  term. The difference of the isotope shifts of the two lines gives therefore the difference in the level isotope shift between  $a^6D_{1/2}$  and  $a^6D_{3/2}$  and should be close to zero because the levels belong to the same term. Nevertheless, as shown in Table II, we find a difference in the isotope shifts of 526 MHz. The  $J$  dependency of the isotope shift should be much smaller (as caused by crossed second-order effects). In Table II, three other pairs of lines can be found which have, again, a common upper level and the same lower levels:  $\lambda=667.4-677.2$  nm,  $\lambda=634.6-643.5$  nm, and  $\lambda=584.4-591.9$  nm. In all four cases we found nearly the same difference of the isotope shifts of the lines.

Testing the very unlikely assumption that the value of  $I^{180}$  could be wrong, we tried to evaluate the hyperfine structure of these four line pairs under the assumption of other values of  $I^{180}$ . It was not possible in this way to reduce the difference of the isotope shifts to about zero simultaneously for all pairs of lines. So, the fact that we get for all four pairs the same difference establishes  $I^{180}=9$ .

From these four pairs of lines, we determined the difference in the level isotope shift

$$\Delta T\{5d^4(a^5D)6s a^6D_{3/2}-5d^4(a^5D)6s a^6D_{1/2}\}$$

to be 527(35) MHz. Therefore, we must assume that the levels of the  $a^6D$  term are not purely built of the configuration  $5d^4(a^5D)6s$  but are mixed with other even configurations. Indeed, the intervals between neighboring  $^6D$  levels are 1116, 991, 1268, and 217  $\text{cm}^{-1}$  (ordered descending in  $J$ ). In pure configurations with pure  $LS$  coupling, the ratio of the intervals is 9:7:5:3.

The levels  $z^6D_{3/2}^o$  and  $z^6D_{1/2}^o$  belong to the same term. The level isotope shift should therefore be very similar.

TABLE IV. Hyperfine-structure constants  $A$  and  $B$  of  $^{180}\text{Ta}$  in comparison with results of other authors and ratios of the determined  $A$  and  $B$  factors.

Designation	$\sigma$ ( $\text{cm}^{-1}$ )	$A^{180}$ (MHz)	$B^{180}$ (MHz)	Source	$A^{180}/A^{181}$	$B^{180}/B^{181}$
Even-parity levels						
$a^4F_{5/2}$	2010.10	284.2(4)	-1247(17)	[4]	0.7930(36)	1.502(50)
$a^4P_{1/2}$	6049.42	693(4)	0	[4]	0.7839(28)	
$a^6D_{1/2}$	9758.97	2437(12)	0	This work	0.7861(40)	
$a^6D_{3/2}$	9975.81	958(5)	-19(10)	This work	0.7891(40)	1.73(85) <sup>a</sup>
Odd-parity levels						
$z^4D_{3/2}^o$	19657.78	507.2(5)	-415(11)	[4]	0.7882(13)	1.487(68)
$z^6F_{1/2}^o$	23355.41	-1396(4)	0	[4]	0.7825(33)	
$z^6F_{3/2}^o$	24243.42	467(2)	-426(20)	This work <sup>b</sup>		
$z^6D_{1/2}^o$	24516.69	3257(16)	0	This work	0.7839(40)	
$z^6D_{3/2}^o$	24739.03	1115(5)	972(50)	This work <sup>c</sup>	0.7888(40)	1.533(85)
$y^4D_{1/2}^o$	25512.63	70(4)	0	This work <sup>b</sup>		
$y^4D_{3/2}^o$	26363.69	500(2)	587(30)	This work <sup>b</sup>		
$z^4P_{1/2}^o$	26866.05	1924(8)	0	This work <sup>b</sup>		
$z^4P_{3/2}^o$	26590.03	1138(5)	347(20)	This work <sup>b</sup>		
Averaged ratios					0.7869(30)	1.504(70)

<sup>a</sup>Not used for averaging.

<sup>b</sup>Calculated using averaged ratios of  $A$  and  $B$  constants.

<sup>c</sup>Averaged values for the lines  $\lambda=667.4$  and  $677.2$  nm.



For comparison, the isotope shifts of investigated lines which have  $a^6D_{1/2}$  as the lower level are summarized in Table V. The differences of the isotope shifts given in column 5 are direct differences of level isotope shifts. In columns 3 and 4 the configurations given by Moore [5] and van den Berg, Klinkenberg, and van den Bosch [7] are listed.

In Table V, we find three groups of lines: The lines  $\lambda=667.4$ ,  $677.4$ , and  $690.2$  nm, which have the upper levels  $z^6D_{1/2}^o$ ,  $z^6D_{3/2}^o$ , and  $z^6F_{3/2}^o$ , belonging to the configuration  $5d^36s(a^5F)6p$ , show positive isotope shifts of medium magnitude. The lines  $\lambda=602.1$  and  $634.6$  nm with the upper levels  $y^4D_{1/2}^o$  and  $y^4D_{3/2}^o$  show a very large positive isotope shift of about 2 GHz. For this reason, we argue that these two levels belong to a configuration different from that of the three levels mentioned before. Van den Berg, Klinkenberg, and van den Bosch [7] suggest another configuration,  $5d^26s^2(^3P)6p$ , for these two levels, which is favored by our results. The isotope shifts of the two remaining lines are between those of the two groups mentioned above and the values lie very close together. As mentioned above, we believe therefore that both upper levels belong to the same term,  $z^4P^o$ , as designated by Moore [5]. On the other side, the absolute value of the isotope shift is significantly smaller than for the  $y^4D^o$  levels, and therefore we assume that the electron configuration may be  $5d^36s(^3P)6p$  as suggested by van den Berg, Klinkenberg, and van den Bosch [7] or a mixture between  $5d^36s(^3P)6p$ , and  $5d^26s^2(a^3P)6p$ .

Looking at the isotope-shift results of Harzer [4] in the two lines  $\lambda=577.7$  and  $566.5$  nm, it is interesting to note that both values coincide nearly to within the limits of error. This fact is surprising if one considers that the two upper levels involved belong to quite different electron configurations. Comparing now the IS's of the lines

$\lambda=690.2$  and  $577.7$  nm, we find closely lying values (827 and 921 MHz). The upper levels of the two transitions belong to the same term ( $z^6F^o$ ), but the lower levels [ $5d^4(a^5D)6s a^6D_{1/2}$  and  $5d^36s^2 a^4P_{1/2}$ , respectively] belong to different electron configurations. Therefore, a large change in the isotope shift is expected, in contradiction to the experimental result. These circumstances indicate a need for deeper knowledge of the tantalum atomic level classification and wave functions.

## V. CONCLUSION

Our investigations led to an extension of the available IS data of  $^{181}\text{Ta}$  and  $^{180}\text{Ta}$ . Moreover, it was possible to determine previously unmeasured differences of level isotope shifts between these two isotopes. As is well known, IS can be very helpful in checking the electron configuration given in the existing literature. We could show that it is not possible to explain the observed isotope shifts by using the electron configurations proposed in tantalum fine-structure identification. We therefore see an urgent need for a revision of the interpretation of the tantalum level scheme. A theoretical analysis of the atomic fine structure as well as an experimental determination of hyperfine-structure constants of further levels are in progress.

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