Model independent determination of the spin of the ^{180}Ta naturally occurring isomer

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The hyperfine structures of the 33715.27 cm^{-1} and 33706.47 cm^{-1} transitions from the ground state of singly ionized Ta have been measured by collinear laser spectroscopy. The structures were found to contain a large second order contribution. From fitting the observed hyperfine components for both ^{181}Ta and the ^{180}Ta naturally occurring isomer it was possible to determine the first and second order hyperfine structure coefficients. As no model independent determination of the nuclear spin of the ^{180}Ta isomer has been performed, fitting was attempted for a range of spins. A clear chi-squared minimum is obtained for a nuclear spin of 9, in agreement with model dependent measurements.

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Naturally occurring tantalum is unique in that the 0.012% abundant ^{180}Ta isotope is found not to be in the nuclear ground state but in a high spin isomeric state. The significance of this naturally occurring isomer led Wakasugi *et al*. [1] to recommend further work in order to determine the nuclear spin by model independent means. In this work a nuclear model independent determination of the spin has been performed using high resolution collinear laser spectroscopy at the University of Jyväskylä [2].

Hyperfine structure in atomic levels can give important information on nuclear properties such as the magnetic dipole and electric quadrupole moments [3]. However, when the separation of fine structure levels is small and the hyperfine structure within a level is large, second order hyperfine contributions may be detectable. In the Ta II transitions studied here, large second order pertubations are found. The sensitivity of these pertubations to the nuclear spin and the separation of atomic fine structure levels is used to confidently determine both parameters.

A comprehensive discussion of the collinear laser spectroscopy technique and setup has previously been reported [4]. In this work ^{180}Ta^m and ^{181}Ta ions were produced by an electrical discharge between the natural Ta cathode and a second electrode in the presence of low pressure He gas. Spectroscopy on a mass separated ion beam was performed using 1mW of UV generated by intracavity frequency doubling of light in a ring dye laser.

All nine of the ^{181}Ta hyperfine structure components and eight of the nine ^{180}Ta^m components were detected in the $5d^36s$ ($^5F_1$) ground state to $5d^26s6p$ ($^5G_2^o$), 33715.27 cm^{-1} transition. However, for the weaker $5d^36s$ ($^5F_1$) to $5d^26s6p$ ($^5F_1^o$), 33706.47 cm^{-1} transition it was only possible to detect three of the seven ^{180}Ta^m components along with all seven ^{181}Ta components. Due to the large size of the hyperfine structure splitting in both transitions data was collected over a series of scan regions. The separation of hyperfine structure components was extracted from the full data set by taking the weighted mean of each component separation for all scans covering the appropriate region of the spectrum.

A selection of scans demonstrating the features of the observed transitions have been compiled in Fig. 1. The scans presented for the 33715.27 cm^{-1} transition were acquired using the bunched collinear laser spectroscopy technique [5]. Due to space charge constraints it was necessary to significantly attenuate the beam current. This resulted in the apparent reduction in count rate for this transition, despite its larger transition probability.

The shift in energy $W^{(1)}_{F,J}$ of a hyperfine structure component F due to the first order hyperfine interaction between a nucleus with spin I and an atomic level with total angular momentum J is given by

$$W^{(1)}_{F,J} = \sum_k (-1)^{I+J+F} \frac{\begin{Bmatrix} F & J & I \\ k & I & J \end{Bmatrix}}{\begin{pmatrix} I & k & I \\ -I & 0 & I \end{pmatrix}\begin{pmatrix} J & k & J \\ -J & 0 & J \end{pmatrix}} \times \langle I\,I|T^{(k)}_n|I\,I\rangle\langle J\,J|T^{(k)}_e|J\,J\rangle, \quad (1)$$

where $\langle I\,I|T^{(k)}_n|I\,I\rangle$ is the nuclear electromagnetic moment of order k and $T^{(k)}_e$ operates on the atomic system. Following the formalism adopted by Schwartz in his reformulation of the theory of hyperfine interactions [6], one may express the product of the two matrix elements in Eq. (1) as A_k. Due to symmetry constraints only odd magnetic and even electric interactions are possible and in most cases it is only possible to detect interactions for $k \leqslant 2$. Consequently the first order interaction for a given atomic level may be characterized by two coefficients, the magnetic dipole interaction strength A_1 and the electrostatic quadrupole interaction strength A_2. The A_k coefficients may be related to the conventional hyperfine A and B coefficients using

$$A_1 = IJA, \quad A_2 = \tfrac{1}{4}B. \quad (2)$$

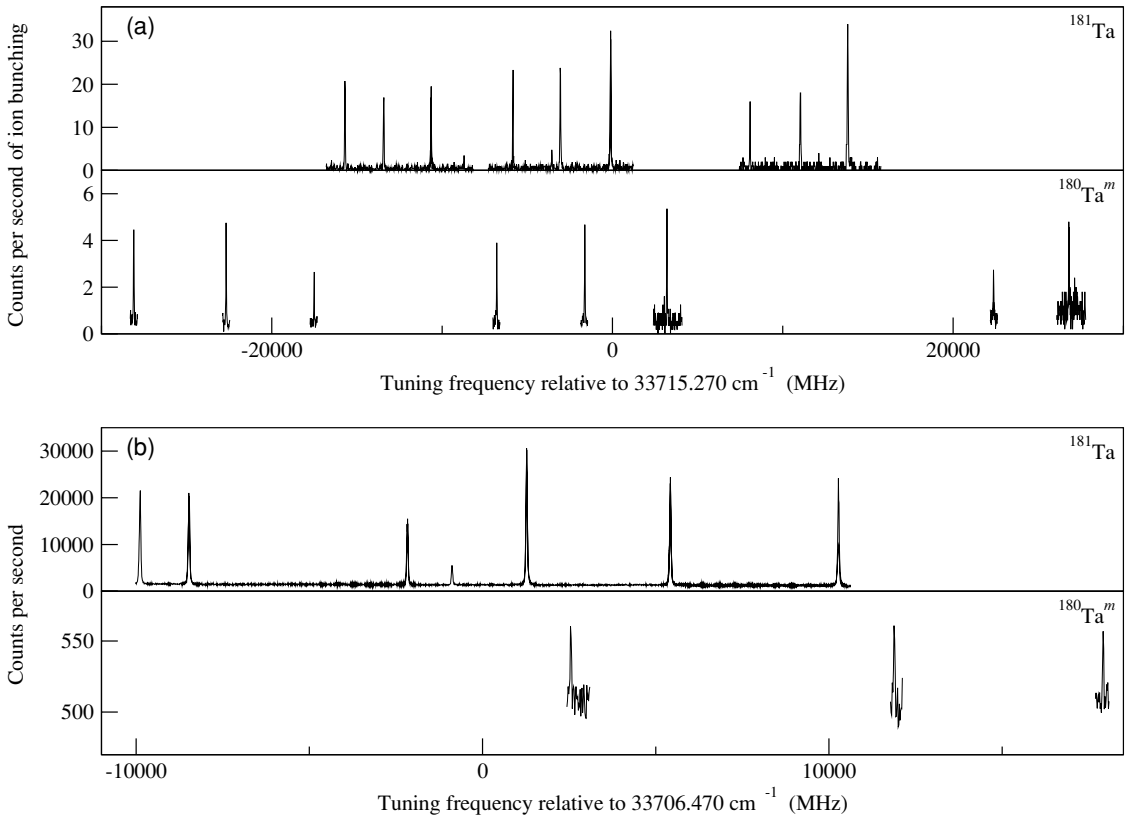

FIG. 1. A selection of scans demonstrating the observed hyperfine structures of ^{180}Ta^{m} and ^{181}Ta in (a) the 33715.270 cm^{-1} transition and (b) the 33706.47 cm^{-1} transition.

The isotope dependence of the A_k coefficients may be expressed in terms of the following relations:

$$\frac{A_1^{180m}}{A_1^{181}} = \frac{\mu^{180m}}{\mu^{181}}(1+\Delta),$$
$$\frac{A_2^{180m}}{A_2^{181}} = \frac{Q^{180m}}{Q^{181}}, \tag{3}$$

where μ^X is the magnetic dipole moment and Q^X is the spectroscopic quadrupole moment of the nucleus X. Scaling of A_1 coefficients between isotopes may be subject to a small anomaly, typically of the order of less than 1%. This is due to the finite size of the nucleus and relativistic effects [7], and may be accounted for by Δ in Eq. (3).

The shift in energy $W^{(2)}_{F,JJ'}$ of a hyperfine structure component due to the second order hyperfine interaction with a neighbouring level may be expressed as

$$W^{(2)}_{F,JJ'} = \sum_{k_1}\sum_{k_2}(-1)^{2I+J+J'+2F}\frac{\begin{Bmatrix} F & J & I \\ k_1 & I & J' \end{Bmatrix}\begin{Bmatrix} F & J' & I \\ k_2 & I & J \end{Bmatrix}}{\begin{pmatrix} I & k_1 & I \\ -I & 0 & I \end{pmatrix}\begin{pmatrix} I & k_2 & I \\ -I & 0 & I \end{pmatrix}}$$
$$\times \langle II|T_n^{(k_1)}|II\rangle\langle II|T_n^{(k_2)}|II\rangle$$
$$\times \frac{\langle J\|T_e^{(k_1)}\|J'\rangle\langle J'\|T_e^{(k_2)}\|J\rangle}{\triangle E_F}, \tag{4}$$

where J' is the total angular momentum of the neighboring level. The denominator $\triangle E_F$ may be related to the separation of the two fine structure levels, $E_J - E_{J'}$ via

$$\triangle E_F = E_J - E_{J'} + E_{X,J} - E_{X,J'} + W^{(1)}_{F,J} - W^{(1)}_{F,J'}, \tag{5}$$

where $E_{X,J}$ is the difference between the nominal energy of the fine structure level, E_J and the centroid of the hyperfine structure for the isotope X.

When the first order interaction may be described in terms of a dipole contribution and a quadrupole contribution, the second order interaction may be limited to $k_1 \leqslant 2$ and $k_2 \leqslant 2$. This results in a set of three second order contributions, a dipole-dipole term for $k_1 = 1$ and $k_2 = 1$, a quadrupole-quadrupole term given by $k_1 = 2$ and $k_2 = 2$ and a dipole-quadrupole contribution given by the sum of the two remaining terms in the double summation.

If the ratio of nuclear moments for two isotopes is known from the first order hyperfine structure, the second order contributions may be scaled between the two isotopes. This is due to the separation of nuclear and atomic factors obtained in Eq. (4).

In many cases in which the second order hyperfine interaction has been detected the separation of fine structure levels is large in comparison to the hyperfine splitting. For these cases it is often possible to assume only dipole-dipole type interactions and to approximate the separation of hyperfine structure components to the separation of their respective fine structure levels [8,9], thus neglecting the $W^{(1)}$

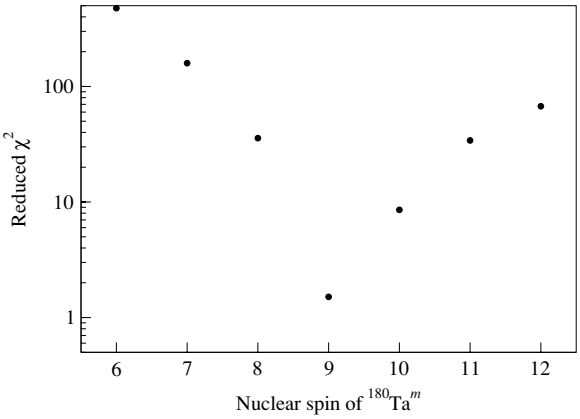

FIG. 2. The reduced χ^2 achieved on fitting the observed spectra over a range of nuclear spins.

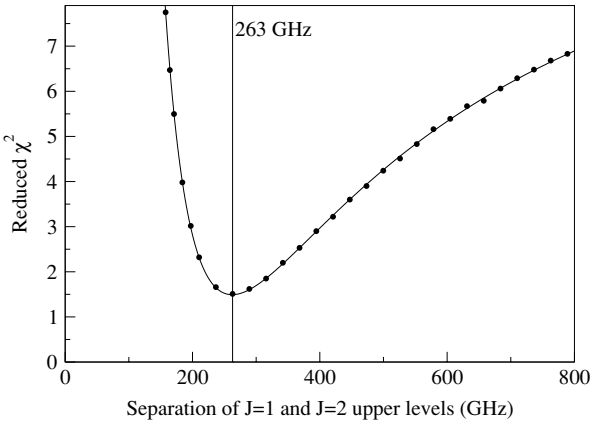

FIG. 3. Confirmation of the separation dependence of the second order interactions used in the fitting.

contributions in Eq. (5). In this work no such approximations are made as interactions of the dipole-dipole, dipole-quadrupole and quadrupole-quadrupole type are considered and the F dependence of $\triangle E_F$ is included.

Weighted least-squares fitting of the centroids of the 27 detected hyperfine structure components for the two isotopes in the two transitions was undertaken. The second order contributions to the upper states were scaled between the two isotopes using the ratios of nuclear moments obtained from the lower state hyperfine structure. In addition, the first order upper state electric quadrupole type contributions were scaled between the two isotopes using the ratio of quadrupole moments from the lower state. Adopting this approach results in a set of 17 fit parameters and allows for the possibility of hyperfine anomaly between the two isotopes in the three atomic states. By carrying out this fitting for a range of values of nuclear spin of $^{180}Ta^m$ it was possible to generate a plot of chi-squared per degree of freedom against spin as shown in Fig. 2. A clear minimum can be seen in this plot for a nuclear spin of 9.

It is also important to confirm the assumption that the observed deviations from first order splitting are due to the second order hyperfine interaction between the two levels and not other effects. Figure 3 shows the reduced χ^2 plotted against the separation of fine structure levels used in the fitting, $E_J - E_{J'}$, a clear minimum is detected at 263 GHz, the expected separation of the two fine structure levels [10]. In previous work on the second order hyperfine interaction it has often been possible to approximate $\triangle E_F$ to the fine structure separation thus removing the hyperfine structure component dependence on the separation of levels. It is clear that this approximation would not be valid in this work as the reduced χ^2 tends towards 13 for large separations which are equivalent to a removal of this component dependence.

As the spin of $^{180}Ta^m$ is confirmed by this approach it is possible to use Wakasugi's high resolution laser rf double resonance determination of the magnetic dipole moment and electric quadrupole moment of $^{180}Ta^m$ [1] as the scaling factor for the second order coefficients. This is preferable to scaling the second order coefficients from the lower state first order coefficients as it is thought that Wakasugi's measurement is only subject to a small hyperfine anomaly. The resulting first order fit parameters are reported in Table I where the first order coefficients are presented in the A_k form adopted by Schwartz.

Using this approach the hyperfine anomaly has been determined for the three states. Assuming that the hyperfine A coefficients reported by Wakasugi were indeed free from hyperfine anomaly our A_1 values would suggest no detectable anomaly in the $J = 2$ upper state and a reatively large anomaly of 1% for both the ground state and the $J = 1$ upper state.

The products of the four matrix elements in Eq. (4) give the second order parameters for the interactions between the two upper states. The contributions of dipole-dipole, dipole-quadrupole, and quadrupole-quadrupole type for $^{180}Ta^m$ were $-1258(14)$ GHz^2, 127(2) GHz^2 and $-0.6(11)$ GHz^2, respectively. The small size of the quadrupole-quadrupole interaction compared to the experimental uncertainty serves as justification for neglecting higher order interactions.

TABLE I. The first order hyperfine interaction coefficients.

Configuration		$^{180}Ta^m$ A_1(MHz)		$^{180}Ta^m$ A_2(MHz)		^{181}Ta A_1(MHz)		^{181}Ta A_2(MHz)	
$5d^36s$	($^5F_1$)	−16655	(3)	−205.7	(9)	−8265.3	(3)	−135.6	(1)
$5d^26s6p$	($^5F_1^o$)	−13573	(3)	−153.6	(7)	−6735.4	(1)	−101.4	(5)[a]
$5d^26s6p$	($^5G_2^o$)	9324	(4)	−427	(1)	4582.0	(8)	−281.9	(8)[a]

[a]The A_2 of ^{181}Ta are included for completeness. As these upper state coefficients have been scaled between the isotopes in the fitting they do not constitue additional fit parameters.

The isotope shift for the transition with upper level J is given by $E_{181,J} - E_{180m,J}$. For the 33706.47 cm^{-1} and 33715.27 cm^{-1} transitions the isotope shifts obtained were −49(7) MHz and 68(5) MHz, respectively. With the present information it is not possible to separate the mass shift and field shift components of these isotope shifts. This is due to the heavily mixed nature of the configurations and the possibility of large specific mass shifts which are often found when d electrons contribute to the transition.

Norquist and Beck performed multiconfigurational Dirac-Fock calculations for the even parity lower levels and the $J = 1$ odd parity levels in Ta II [11]. The wavefunctions determined from these calculations have been used to predict the hyperfine structure A coefficients. It is instructive to compare the theoretical A coefficient for the $J = 1$ upper state with that determined by this work. Adjusting the theoretical A coefficient of the upper state with the more precise value of the magnetic moment of ^{181}Ta used in this work ($\mu^{181} = +2.3705(7)\mu_N$ [12]) produces a value[1] of −1945 MHz. Using the relation $A_1 = IJA$, our A_1 value is equivalent to an A coefficient of −1924 MHz. Clearly good agreement is obtained, well within the 100 MHz uncertainty quoted by Norquist and Beck [11].

It has been shown that inclusion of the full separation dependence of the second order hyperfine interaction introduces a degree of sensitivity to the separation of fine structure levels. Furthermore, inclusion of this dependence is vital in the case studied in order to quantitatively describe the observed structures.

The substantial hyperfine anomalies detected in the ground state and $J = 1$ upper state are some of the largest measured. At present a full interpretation is not possible due to the limited nuclear and atomic data for this system, however such cases may permit the systematic study of the distribution of nuclear magnetization in the Ta isotopes and isomers.

The model independent determination of the nuclear spin of the ^{180}Ta isomer has provided conclusive evidence for a nuclear spin assignment of 9. In addition, the magnetic moment of ^{180}Ta^m has been shown to be in good agreement with that determined by Wakasugi. Consequently the $\pi[514]9/2^-$ $\nu[624]9/2^+$ configuration is confirmed by this work.

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[1] Comparison of Norquist and Beck's A values with those measured by Zilio and Pickering [13] would suggest that the nuclear spin used by Norquist and Beck for the odd parity upper states was not 3/2 as reported, but was indeed 7/2, as one would expect. However, for the lower states where no spin is indicated, a value of 3/2 produces excellent agreement with both the A values of Zilio and Pickering and the ground state A value of ^{181}Ta measured in this work.

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