High resolution isotope shifts and hyperfine structure measurements of tungsten by laser-induced fluorescence spectroscopy

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Received 23 December 2012, in final form 19 February 2013
Published 22 March 2013
Online at stacks.iop.org/JPhysB/46/075003

Abstract
Isotope shifts and hyperfine structure of tungsten were studied in the near UV range. We have used laser-induced fluorescence spectroscopy on a pulsed supersonic beam to probe the $^{5}D_{0} \rightarrow ^{5}F_{1}$ transition at 384.9 nm, $^{7}S_{3} \rightarrow ^{7}P_{4}$ transition at 400.9 nm and $^{7}S_{3} \rightarrow ^{7}P_{3}$ transition at 407.4 nm. Three new magnetic hyperfine constants are reported for $^{7}P_{3}$, $^{7}P_{4}$ and $^{5}F_{1}$ states. The isotope shifts of the 384.9 nm transition are presented for the first time, and the isotope shifts of 400.9 and 407.4 nm transitions are measured with an order of magnitude higher precision compared to the previous measurements. As a result, the nuclear parameters $\lambda$ and $\lambda_{\text{rel}}$ are extracted from the isotope shifts with an improved precision.

1. Introduction
The high-resolution spectroscopic study of tungsten (W) has been of interest in various fields of physics, ranging from astrophysics [1] to nuclear physics [2–4]. Accurate identification of atomic transition lines of high-Z elements has been known to provide useful information on the nucleosynthesis process in metal-poor stars [1]. On the other hand, being one of the 5D shell atoms, its isotope shifts and hyperfine structure allow enhanced understanding of nuclear structure in the deformed region on the nuclear chart. Böttgenbach [2] analysed the hyperfine structure of 4D- and 5D-shell atoms to study the contact interaction terms. Aufmuth [3, 4] used the isotope shifts of tungsten to extract the nuclear parameter $\lambda$.

More recently, tungsten was proposed as a plasma-facing material in controlled fusion [5]. Particularly, the International Thermonuclear Experimental Reactor (ITER) plans to use W tiles on the divertor plasma [6], along with the spectroscopic diagnostic system [7] to estimate the tungsten influx rate for plasma-edge modelling. According to Skinner [8], the $^{7}S_{3} \rightarrow ^{7}P_{4}$ transition of neutral tungsten at 400.9 nm is considered promising due to its high transition probability. However, there is a complication in spectroscopic diagnostics due to singly ionized tungsten lines (W II) being coincidentally at nearly the same wavelength as the neutral lines (W I), causing line blending issues [8]. Another problem is the incomplete information on the $^{7}S_{3} \rightarrow ^{7}P_{4}$ transition, as the latest study did not reveal the hyperfine structure due to limited resolution [4]. This would lead to further difficulties in accurate modelling of the tungsten influx rate at ITER.

In this paper, we present isotope shifts and hyperfine structure measurements of three neutral tungsten transitions in the near UV range. Laser-induced fluorescence (LIF) spectroscopy on an atomic beam were performed on the $^{5}D_{0} \rightarrow ^{5}F_{1}$ transition at 384.9 nm, the $^{7}S_{3} \rightarrow ^{7}P_{4}$ transition at 400.9 nm and the $^{7}S_{3} \rightarrow ^{7}P_{3}$ transition at 407.4 nm. A magnetic dipole hyperfine constant of the $^{7}S_{3}$ level is compared with a prior report [9], and three new measurements are made for the constants of the $^{7}P_{3}$, $^{7}P_{4}$ and $^{5}F_{1}$ levels.

The isotope shifts of the 384.9 nm transition is presented for the first time and the isotope shifts of 400.9 and 407.4 nm
were measured with approximately ten times higher precision compared with the previous results. A high-resolution isotope shift study of these three transitions enabled us to extract the nuclear parameters $\lambda$ and $\lambda_{\text{rel}}$, which were compared with the previous results [4]. Also, the completely resolved tungsten spectrum at 400.9 nm is directly related to the application at ITER [8]. Experimental details are shown in sections 2 and 3, followed by analysis in sections 4 and 5 and the conclusion in section 6.

2. Experimental methods

The beam production of refractory metals has been limited due to its high melting point. Having the highest melting point of all metals, a tungsten atomic beam cannot be generated efficiently through the conventional oven method [10]. In the past, we have used a resistively heated tungsten wire to generate a high flux beam [11]. However, the blackbody radiation coming from the heated wire was limiting our ability to detect fluorescing light from the atomic transitions. Here, we use the Smalley-type pulse supersonic beam technique [12] to overcome these issues. Only a brief review of the technique is given here and the details can be found in [12].

Tungsten atoms are ablated from a rod (American Elements, 99.9% purity) by the third harmonic of the Nd:YAG pulse laser (Quantel), while the solenoid gas valve (Parker, general valve series 999) entrains the atoms with Argon buffer gas. The atoms get cooled down through buffer gas collisions resulting in a narrow Doppler linewidth. The turbo pump with 1500 L s$^{-1}$ of pumping speed maintained the operating pressures to be, $5 \times 10^{-6}$ Torr inside the vacuum chamber. A diagram of the experimental apparatus is shown in figure 1.

A tunable cw Ti-SAPL laser (Coherent MBR110) generated the IR light, which was frequency doubled with LBO crystal inside a cavity (Coherent MBD200) to produce the light in the 380–410 nm range. This wide range of tunability enabled us to probe all of the transitions presented in this paper using the same laser system. The probe laser was focused with an intensity of approximately 1 mW cm$^{-2}$ at the intersection point where the laser beam is crossing the atomic beam perpendicularly. The laser-induced fluorescing light was collected by a spherical lens into a water-cooled photo multiplying tube (Hamamatsu) connected to the photon counter. The photon counts were recorded simultaneously as the wavelength meter (High Finesse WSU series) measures the frequency of the probe laser with an accuracy level of a few MHz. From the methods that are shown in the appendix, we have estimated the total uncertainty of frequency measurements of 1.7 MHz for the 384.9 nm transition, 2.1 MHz for the 400.9 nm transition and 2.9 MHz for the 407.4 nm transition.

3. Experimental results

Here, we have measured three W optical transitions at the wavelengths of 384.9, 400.9 and 407.4 nm. The 384.9 nm transition is from the $5D_0$ ground electronic state with the $5d^46s^2$ configuration to the $5F_1$ excited state with the $5d^36s^26p$ configuration [13]. Two other transitions of 400.9 and 407.4 nm share the same low lying metastable state of $7S_3$ with the $5d^66s$ configuration, and excited states of $7P_4$ and $7P_3$ both have the $5d^66s6p$ configuration [13].

Tungsten has four even isotopes of $^{180\text{W}}, ^{182\text{W}}, ^{184\text{W}}$ and $^{186\text{W}}$, with nuclear spin of 0, and one odd isotope of $^{183\text{W}}$, with nuclear spin of 1/2 giving rise to hyperfine structure. As shown in figures 2–4, the 400.9, 407.4 and 384.9 nm transitions have three, four and two allowed electric dipole hyperfine transitions, respectively.

Previous studies of the 400.9 and 407.4 nm transitions reported 20–30 MHz precision on isotope shift measurements with the hyperfine structures being unresolved [4]. In this paper, we have achieved ten times higher precision on the isotope shift measurements of both transitions and the hyperfine structures are clearly resolved as well. These are shown in figures 2 and 3. The ratio of the peak amplitudes agreed within the 10% level of the natural abundance ratio.
combined with the hyperfine line strengths, which were calculated from the Clebsch–Gordan coefficients. The small disagreement comes from the fluctuation of atomic beam intensity. The linewidth of the measurements were mainly limited by Doppler broadening, with a Gaussian linewidth of 5 MHz. We extract the line positions of each isotope from individual Gaussian fits, except for the merged peaks of $^{183}$W(b) and $^{183}$W(c) in figure 3, which we fit to two Gaussians simultaneously. The fit gave a frequency uncertainty of 200 kHz in line positions, which is negligible compared with the measurement uncertainty described in the previous section.

As seen in figure 4, the isotope shifts of the 384.9 nm transition were partly unresolved due to limited resolution, and the $^{180}$W with 0.12% natural abundance is believed to be masked underneath the hyperfine state (b) of $^{183}$W. As some of the peaks were not resolved, we had to take a different approach to analyse the isotope shifts. The first step was to take an average of the transition’s multiple scans, in order to eliminate the beam fluctuation effect and to obtain an exact line profile. After this, we used a multi-parameter Gaussian fit to extract the isotope and hyperfine shifts. Based on the natural abundance ratio and the intensity rule for the hyperfine transitions, we identified two peaks on the wing side in figure 4 as the hyperfine states of $^{183}$W. Also, we noticed the centre of gravity of $^{183}$W lying closer to the right side of the unresolved centre peak, which indicates negative isotope shifts for the 384.9 nm transition.

With the above constraints for peak assignments, the least-squares fit was performed to match the experimental data. The least-squares fitting curve is shown as the red dashed line in figure 4 with a Gaussian linewidth of 5 MHz, which overlaps reasonably well with the experimental data shown by black hollow circles.
hollow circles. There is a small amount of disagreement for the fit of the hyperfine state (a) of 182W, where the fit seems to be underestimating the experimental data. This could have come from either the residual intensity fluctuation of the atomic beam or the probe laser power, which was not averaged out. Nevertheless, the current fitting method was the only possible way to fit the experimental data with a given natural abundance of W and the hyperfine intensity rules. The impact of this deviation in terms of the hyperfine constant measurement will be discussed in the following section. The blue solid line of figure 4 is the result of the simulation with a linewidth of 0.5 MHz to show the line positions, and the yellow dotted line is the centre of gravity position of 183W. The 180W isotope was excluded from the fit, as the experimental data does not have a high enough resolution to constrain its position. We note that regardless of the peak assignments, the upper limits of 20 MHz amu−1 for the isotope shifts and 80 MHz for the hyperfine splitting of the 5F1 state, can be deduced from figure 4. No previous study of isotope shift exists for the 384.9 nm transition, however, Gluck [14] has reported 0 isotope shifts for this transition with a frequency resolution of 30 MHz, which is consistent with our result within their uncertainty of measurement.

4. W hyperfine structure analysis

The hyperfine structure of the 183W isotope is only caused by magnetic dipole interaction as it has a nuclear spin of 1/2 [15]. In this case, the hyperfine energy levels are given by,

$$E_F = E_J + h\alpha \frac{F(F+1)-J(J+1)-I(I+1)}{2},$$

where \(h\) is Planck’s constant and \(\alpha\) is the magnetic hyperfine constant. The shifted energy levels due to this hyperfine interaction are shown in figures 2–4. The hyperfine frequency splitting within the electronic state becomes,

$$\delta\nu_{\text{hyperfine}} = \alpha \left(I + \frac{1}{2}\right).$$

It is straightforward to extract the magnetic hyperfine constant \(\alpha\) from measured frequency splittings using equation (2). As some of the hyperfine splittings are measured in multiple transitions, such as the splitting of the 7S3 state, we take the weighted average of the measurements from different transitions to get \(\alpha\), where we take the uncertainty of each independent measurements as the weight. The results are shown in table 1. We report three new hyperfine constants for 7P3, 7P4, 5F1 states, and the constant for the 7S3 state is compared with the previous result [9]. Good agreement is found within the error of the measurement for the 7S3 state.

For the 7S3, 7P3, and 7P4 states, the dominant error of the measurement came from the wavelength meter calibration error deduced in the appendix, since the fitting error was a magnitude of order smaller. However, for the hyperfine constant of the 5F1 state, we had to take both the fitting error and the wavelength meter calibration error into account as the fitting error became much more pronounced. We take the 1σ fitting error of the hyperfine splitting measurement in figure 4, which was given by 2.3 MHz, and combined with the wavelength meter calibration error to get the final uncertainty.

Wyart suggested 5d6s6p as the dominant configuration for 7P3, 7P4 states, and the 5d6s6p configuration for the 5F1 state [13]. Our measurements of larger constants \(A\) in the cases of the 7P3 and 7P4 states compared with the case of the 5F1 state, support Wyart’s configurations. This is due to the open shell 6s electron from the 5d6s6p configuration having bigger contributions to \(A\) than the 5d6s6p configuration.

5. W isotope shift analysis

The theory of atomic isotope shifts are well developed in many publications [16–19]. Due to the way our experiment is designed, we examine the total isotope shift of the electronic transition rather than the isotope shifts of individual electronic states. The total isotope shift of the electronic transition is equivalent to the difference in isotope shifts of upper and lower electronic states that are involved. Analysing the isotope shift of individual electronic states will be discussed at the end of this section.

The isotope shift between two isotopes with mass numbers \(A\) and \(A'\) of an optical transition \(i\) can be written as,

$$\delta\nu_{i,\text{MS}} = \delta\nu_{i,\text{MS}}' + \delta\nu_{i,\text{FS}},$$

where MS stands for the mass shift and FS is for field shift. The mass shift is further separated into normal mass shift (NMS) and specific mass shift (SMS), both coming from the change in nuclear mass.

$$\delta\nu_{i,\text{MS}} = \delta\nu_{i,\text{NMS}} + \delta\nu_{i,\text{SMS}}$$

$$= (M_{i,\text{NMS}} + M_{i,\text{SMS}}) \frac{A' - A}{AA'}. \quad (5)$$

The FS originates from the change in volume and shape of the nucleus, therefore, being directly related to the changes in mean square nuclear charge radii \(\langle r^2 \rangle\). This relation is shown below,

$$\delta\nu_{i,\text{FS}} = F_i \lambda_{AA'} \quad (6)$$

$$\lambda_{AA'} = \delta\langle r^2 \rangle_{AA'} + \frac{C_2}{C_1} \delta\langle r^4 \rangle_{AA'} + \frac{C_3}{C_1} \delta\langle r^6 \rangle_{AA'} + \cdots \quad (7)$$

$$\approx \delta\langle r^2 \rangle_{AA'} \quad (8)$$

$$F_i = E_i f(Z), \quad (9)$$

where \(E_i\) and \(f(Z)\) are the electronic and relativistic correction factors defined in [19] and \(C_n/C_1\) ratios are tabulated by Seltzer [16].
These King plots are shown in figure 5 where the axes are the modified isotope shifts of each transition relative to the reference transition. A factor of 2 was multiplied to both axes for display purposes, which was to obtain the units in MHz. The 180W isotope was not measured for the 384.9 nm transition.

The 180W isotope was not measured for the 384.9 nm transition. 1σ errors are shown inside the parentheses.

From the linear fit, we obtain the relations,

\[ \text{Slope} = \frac{E_i}{E_{543.5 \text{ nm}}} \],

\[ \text{Intercept} = M_{i,NMS} - M_{543.5 \text{ nm},NMS} \times \frac{E_i}{E_{543.5 \text{ nm}}} \]  \hspace{1cm} (13)

The SMS of the 543.5 nm transition is estimated by a semi-empirical relation,

\[ \delta \nu_{543.5 \text{ nm}, \text{SMS}} = (0 \pm 0.5) \delta \nu_{543.5 \text{ nm}, NMS} \]  \hspace{1cm} (14)

which is only valid for ms²–nsp transitions [17]. Combining equations (12) to (14), we can calculate the SMS of our measured transitions. Knowing NMS and SMS, the FS is readily calculated from equations (3) and (5). The extracted values of NMS, SMS and the FS for 384.9, 400.9 and 407.4 nm transitions are given in table 2. As discussed previously, isotope shifts of the 384.9 nm transition are given by the least-squares Gaussian fitting method.

From equations (6) and (9), the FS is linked to the nuclear parameter \( \lambda_{AA'} \) through \( E_i \) and \( f(Z) \). The electronic factors \( E_{384.9 \text{ nm}} \), \( E_{400.9 \text{ nm}} \) and \( E_{407.4 \text{ nm}} \) are derived from equation (12), with an electronic factor \( E_{543.5 \text{ nm}} = 0.40882 \) calculated by Aufmuth [3]. The relativistic correction factor \( f(Z) \) was calculated by [19] using the isotope shift constant of Blundell [21]. The extracted \( \lambda_{AA'} \) from three different transitions are shown in the last column of table 2. All of the results show good agreement within the errors.
We analyse different sources of uncertainties in equation (15) for all three transitions, which are shown in table 3 for the case of the isotope shift between $^{184}\text{W}$ and $^{186}\text{W}$. Note that the second term of the right-hand side of equation (15), is a well-defined value from equation (10), therefore has no error. Rows 1–3 of table 3 show uncertainties that are linked to the experimental uncertainty of our frequency measurements. However the uncertainty described in the last row, which contains the SMS of the reference line, relies only on the semi-empirical relation of equation (14). This becomes the dominating error contribution in the case of the 400.9 and 407.4 nm transitions. Therefore, improving the experimental errors alone, which only affects the first three rows of table 3, would not further decrease the uncertainty of the FS of the 400.9 and 407.4 nm transitions. Better estimation of the SMS is required for further improvement.

Since the extracted $\lambda^{AA'}$ in table 2 showed good agreement among different transitions, we take the weighted mean of these to report the final values of $\lambda^{AA'}$ and compare with the previous results of [19] in table 4. The errors are presented in the same manner as before, showing the experimental uncertainty inside the first parenthesis and the theoretical uncertainty inside the second parenthesis. We notice experimental uncertainties being much smaller than the theoretical uncertainties for all isotope pairs.

From the above discussion, it is clear that the limiting factors for high-precision $\lambda^{AA'}$ measurement mainly comes from the uncertainty in estimation of SMS through the semi-empirical relation, and the theoretical uncertainties in the FS. First we discuss the possibility of SMS calculation. Instead of using the King plot approach, there has been several attempts of multi-configuration calculation of SMS [23–25], however, as pointed out by Aufmuth [19], a reliable prediction was never made. Following the approach of [25], the specific mass effect perturbs the energy of the electronic state $\psi$ between mass $A$ and $A'$ by an amount of,

$$\Delta E_{\text{SMS}}(\psi) = \frac{\lambda^{AA'}}{\lambda^{AA'}} \left( \psi \left| \sum_{i>j} \bf{p}_i \cdot \bf{p}_j \right| \psi \right),$$

(16)

where $\bf{p}_i$ is the momentum of the $i$th electron. Thus, the frequency difference in the upper state of $\psi_u$ and the lower state of $\psi_l$ can be written as,

$$\delta \nu_{u \to l,\text{SMS}} = \frac{\Delta E_{\text{SMS}}(\psi_u) - \Delta E_{\text{SMS}}(\psi_l)}{\hbar}.$$  

(17)

This would be a more general way of estimating the SMS as it does not rely on the semi-empirical relation shown in equation (14), however, knowledge of tungsten electron wavefunctions is required.

Similarly, we can write down an expression for the field effect perturbing the energy of $\psi$ between mass $A$ and $A'$ by,

$$\Delta E_{\text{FS}}(\psi) = \frac{\pi \alpha^2_s |\psi(0)|^2}{Z} f(Z) \lambda^{AA'},$$

(18)

where $|\psi(0)|^2$ is the non-relativistic electron charge density at the nucleus and $f(Z)$ is the same relativistic correction factor which was mentioned above. The frequency difference between the upper state of $\psi_u$ and the lower state of $\psi_l$ becomes,

$$\delta \nu_{u \to l,\text{FS}} = \frac{\Delta E_{\text{FS}}(\psi_u) - \Delta E_{\text{FS}}(\psi_l)}{\hbar}.$$  

(19)

As indicated from equation (18), the FS analysis could be used as a study of $|\psi(0)|^2$. This term could be used for calculation of electric fields inside the atoms, which are important in electron electric dipole moment experiments [26].

The tungsten electronic wavefunctions have not been studied in detail so far, which makes the SMS and FS calculations described in equations (16)–(19) unavailable. At the current state, the best way to reduce the fractional error of the nuclear parameter is by defining a relative $\lambda^{AA'}_{\text{rel}}$ as,

$$\lambda^{AA'}_{\text{rel}} = \frac{\lambda^{AA'}}{\lambda^{184,186}}.$$  

(20)

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### Table 3. The sources of uncertainties in $\delta \nu_{u \to l,\text{FS}}$ from different transitions of tungsten.

| Source of uncertainty | Transition (nm) | | | |
|-----------------------|----------------|----------------|----------------|
|                       | 384.9 | 400.9 | 407.4 |
| Statistical Uncertainty of $\nu_{184,186}$ (MHz) | 1.6 | 1.6 | 1.6 |
| Systematic Uncertainty of $\nu_{184,186}$ (MHz) | 0.06 | 1.2 | 2.4 |
| Uncertainty of $\lambda^{186}_{184}$ × Intercept of King plot (MHz) | 0.9 | 1.1 | 1.4 |
| Uncertainty of $\lambda^{184,186}_{\text{nuc},\text{SMS}}$ × Slope of King Plot (MHz) | 0.2 | 3.6 | 3.9 |

### Table 4. $\lambda^{AA'}$ and $\lambda^{AA'}_{\text{rel}}$ compared with the previous results from [19]. 1σ errors are shown inside the parentheses. For our results, the first parenthesis shows the experimental uncertainty from the FS measurement and the second parenthesis showing theoretical uncertainty of $E_i$ and $f(Z)$ from equation (9).

| Isotopes (A, A') | Weighted $\lambda^{AA'}$ (fm$^2$) | Weighted $\lambda^{AA'}_{\text{rel}}$ |
|------------------|-----------------------------------|-----------------------------------|
|                   | This work | Reference [19] | This work | Reference [19] |
| 184, 186          | 0.0807(5) (33) | 0.084(7) | 1 | 1 |
| 182, 186          | 0.0931(5) (38) | 0.097(8) | 1.1537(11) | 1.154(4) |
| 182, 183          | 0.0488(4) (20) | 0.051(5) | 0.6047(22) | 0.607(5) |
| 180, 182          | 0.0652(5) (26) | 0.068(8) | 0.8079(14) | 0.808(23) |
The fractional error of $\lambda_{\text{rel}}^{\text{rel}}$ is much smaller than $\lambda_{\text{rel}}^{\text{abs}}$, which can be seen in the last two columns of table 4. The results are in very good agreement with [19], with 3–16 times better precision.

6. Conclusion

We performed laser-induced fluorescence spectroscopy on a tungsten atomic beam to study isotope shifts and hyperfine structure of three different optical transitions. Three new magnetic hyperfine constants were shown, which agreed with Wyart's configuration of $5d^46s^26p$ for $^7P_3$, $^7P_4$ states, and $5d^36s^26p$ for the $^5F_1$ state. The isotope shifts of 384.9, 400.9 and 407.4 nm transitions were analysed to give nuclear parameters $\lambda$ and $\lambda_{\text{rel}}$. We have shown the dominating error contributions in the field shift process and the $\lambda$ determination. The $\lambda_{\text{rel}}$, which is an alternative way of representing the nuclear parameter, had a smaller fractional error than the absolute $\lambda$. Both cases showed good agreement with the previous studies as well as improved precision. The 400.9 nm transition is related to the spectroscopic diagnostics of the tungsten influx rate at ITER.

Appendix. Wavelength meter calibration

The uncertainty in relative frequency measurement could be divided into statistical uncertainty and systematic uncertainty. First we describe the way we characterize the statistical uncertainty. We have measured one of the hyperfine splittings of the tungsten transition multiple times and compare the uncertainty. We have measured one of the hyperfine splittings divided into statistical uncertainty and systematic uncertainty. The uncertainty in relative frequency measurement could be shown as $\sigma_{\text{rel}}$, which is an alternative way of representing the nuclear parameter $\lambda$. We have shown the dominating error contributions in the field shift process and the $\lambda$ determination. The $\lambda_{\text{rel}}$, which is an alternative way of representing the nuclear parameter, had a smaller fractional error than the absolute $\lambda$. Both cases showed good agreement with the previous studies as well as improved precision. The 400.9 nm transition is related to the spectroscopic diagnostics of the tungsten influx rate at ITER.

In order to test for a possible systematic uncertainty in relative frequency measurements from the wavelength meter, we measured the known ytterbium (Yb) isotope shifts. Ytterbium was chosen due to the notable high-precision isotope shift measurements from Natarajan’s group [27].

Having a relatively low melting point, a Yb atomic beam was produced using the conventional method of a resistively heated oven, rather than the technique we described in section 2. Multiple apertures along the beam line minimized the Doppler linewidth. A tunable frequency doubled diode laser system (Toptica DL pro) generated 555.6 nm light for the LIF spectroscopy on the $^1S_0 \rightarrow ^1P_1$ transition of Yb. Yb has seven isotopes, from $^{168}$Yb to $^{176}$Yb, with two of them, $^{171}$Yb and $^{173}$Yb, having hyperfine structure. Detailed analysis on the Yb spectrum is omitted, as it goes outside the subject of this section.

We compare the centre line positions of all of our Yb isotopes and hyperfine structure measurements with [27], which are shown in table A1. Figure A2 shows the plot of table A1 by black squares, as well as the red solid line showing the least-squares linear fit. Ideally, this line should have slope of 1, and intercept at 0. Our fit gave a slope of 0.9990(6), and an intercept at 2.0(1.7) MHz with 1σ errors inside the parenthesis. The error of the slope, which is $6 \times 10^{-4}$, represents the fractional uncertainty over the measured frequency range. Accordingly, we can assign a systematic uncertainty of 3.6 MHz over the range of 6 GHz in the case of Yb isotope shift measurements at the 555.6 nm transition. By assuming that the wavelength meter would perform in a similar way at the wavelength range of 385–410 nm, we can apply these results for the estimation of systematic uncertainty of tungsten transitions being studied in this work.

In conclusion, we report 1.6 MHz of statistical uncertainty and $6 \times 10^{-4}$ of fractional systematic uncertainty for the frequency measurements from our wavelength meter. The fractional systematic uncertainty converts to 60 kHz for the
Our Yb relative isotope shifts from ref. [12]

Figure A2. Our relative frequency measurements of Yb isotopes and hyperfine structure are shown by black squares. The uncertainty of both ours and the reference measurements are within the symbols. The red solid line shows the least-squares linear fit of the data points.

384.9 nm transition, 1.2 MHz for the 400.9 nm transition, and 2.4 MHz for the 407.4 nm transition. Adding statistical and systematic uncertainties in quadrature, the total uncertainty of frequency measurements are 1.7 MHz for the 384.9 nm transition, 2.1 MHz for the 00.9 nm transition and 2.9 MHz for the 407.4 nm transition.

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