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Absolute frequency measurements of the ${}^1S_0 \rightarrow {}^1P_1$ transition in ytterbium

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Abstract: The ytterbium atom is widely used in the fields of atomic physics, cavity quantum electrodynamics, quantum information processing and optical frequency standards. There is however a strong dispersion among the reported values of the $^1S_0 \rightarrow ^1P_1$ transition frequency. In this article, we present two independent measurements of the absolute frequency of this transition performed with two different wavemeters using atomic fluorescence spectroscopy. The cancellation of Doppler shifts is obtained by fine tuning the angle between the probe laser and the atomic beam. The resulting 174 Yb isotope transition frequency is estimated to be $751526537 \pm 27\,\text{MHz}$.

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1. Introduction

The ytterbium atom has several advantages for modern applications in atomic physics. Cold ytterbium experiments have therefore spread worldwide with applications in metrology, quantum information and quantum gases. Neutral ytterbium offers a narrow optical clock transition used for state-of-the-art frequency metrology [1, 2] and can be cooled down to ultra-low temperatures to study the physical properties of ultra-cold quantum gases [3]. In addition, the Yb⁺ ion offers two optical clock transitions at 436 nm and 467 nm [4, 5], and its microwave clock transition at 12.6 GHz can be used for q-bit manipulation in quantum information processing experiments [6].

The ${}^{1}S_{0} \rightarrow {}^{1}P_{1}$ transition is the first cooling transition of neutral ytterbium and the first isotopic-selective stage in 2-photon ionization of ytterbium. However, there is a large dispersion in the measurements of the ${}^{1}S_{0} \rightarrow {}^{1}P_{1}$ transition frequency [7–11]. The first measurement was performed in 1977 by acquiring spectrograms [7] and listed the energy of the transition as 25 068.227 cm⁻¹ (751.526 54 THz), which was used as the reference for the NIST database [12]. More recent measurements started in 2005, when Das et al. used a ring-cavity resonator stabilized on a Rb transition to measure the absolute frequency in ¹⁷⁴Yb with high accuracy [8]. However, the value they report, 751.525 987 761(60) THz, differs from the one measured in [7] by more than 550 MHz. Nizamani et al. measured the transition frequency in 2010 [9] using a wavelength-meter specified with a 60 MHz accuracy. Their measurement, yielding the value 751.526 65(6) THz, is closer to [7] (110 MHz difference) but not compatible with [8]. In 2016, Enomoto et al. [10] measured the $F = 1/2 \rightarrow F' = 3/2$ hyperfine transition frequency in ¹⁷¹Yb to be 751.527 48(10) THz by accurately calibrating resonance frequencies of an ultra-stable Fabry-Perot cavity with lasers referenced to transitions in rubidium and calcium. They did not measure the transition frequency in the ¹⁷⁴Yb isotope, but their result is in good agreement with [7] and with [9], given their uncertainty and the existing isotopic shift measurements from literature [8, 9, 11, 13–16]. Using an optical frequency comb referenced to the D_2 line of Rb, Kleinert et al. [11] made a very accurate measurement of the transition frequency in 2016, measuring 751.526 533 49(33) THz, disagreeing with the values from [8] and [9].

Given the non-negligible dispersion of the measurements published so far, this work aims

to establish and report on additional independent measurements of the absolute frequency of the $^1\mathrm{S}_0 \to {}^1\mathrm{P}_1$ transition in Yb. Two independent measurements of this transition for different isotopes of ytterbium are obtained by recording the fluorescence spectrum of an atomic beam. Care was taken in the experimental setup to mitigate Doppler shifts and power broadening effects, reducing systematic effects below 5 MHz. The weighted mean of our measurements results in the value 751526537 ± 27 MHz for the $^{174}\mathrm{Yb}$ isotope transition frequency.

2. Experimental setup



Fig. 1. Sketch of the experimental setup. In an ultra-high vacuum chamber, a 399 nm laser beam crosses an atomic beam of neutral ytterbium atoms to excite the $^1S_0 \rightarrow {}^1P_1$ transition. The resulting fluorescence is detected by a low-noise photodiode. An angle θ can be introduced with respect to the perfect orthogonal configuration and a mirror allows for back reflection of the laser beam for alignment purposes (see text for details).

The experimental set-up is sketched out in Fig. 1. A commercial dispenser of ytterbium with natural isotopic abundance [17] is driven at 5.5 A and thus heated close to 600°C in order to produce an atomic beam inside an ultra-high vacuum chamber (background pressure $< 10^{-10}$ mbars). The ${}^{1}\text{S}_{0} \rightarrow {}^{1}\text{P}_{1}$ transition in ytterbium is excited by a 399 nm laser beam crossing the atomic beam nearly orthogonally. The laser light, produced by a commercial external-cavity diode laser (ECDL) [18] with a linewidth of a few MHz, is brought to the experiment chamber via a single-mode fiber. The fiber output coupler is mounted on a rotatable breadboard in order to adjust the angle θ between the probe laser and the direction perpendicular to the atomic beam. The average intensity of the probe beam is $\sim 8 \text{ mW.cm}^{-2}$ corresponding to a saturation parameter of 0.14. The laser frequency is slowly scanned over a ~ 3 GHz range using the piezoelectric actuator driving its grating (PZT) during approximately 100 s. High-apperture optics collect photons scattered by atoms excited to the upper state and decaying back to the ground state. The fluorescence signal is detected with a photodiode [19]. While the fluorescence spectrum of the transition is recorded, the frequency of the laser is sampled every ~ 25 ms on two independent commercial wavemeters [20]. The first wavemeter is a WS/7 Super Precision specified by the manufacturer with an absolute accuracy of ±60 MHz when calibrated with its built-in neon lamp. Its frequency drift in absence of thermal isolation has been measured to be below 30 MHz/day [21]. The second wavemeter is a WS8-2 calibrated to the $F = 3 \rightarrow F' = 4$ hyperfine transition of the D₁ line of caesium at 894 nm, which results in an absolute accuracy of ±30 MHz at 399 nm as specified by the manufacturer.

A mirror placed at the output of the chamber can be used to reflect the laser beam back onto itself. In that case, the two counter-propagating beams have perfectly opposite Doppler shifts with respect to the atomic beam for non-zero angles θ , allowing to tune θ to zero with mrad precision (see section 4).

3. Results

The fluorescence spectrum was recorded at $\theta=0^\circ$ as a function of the frequency for each wavemeter. A Lorentzian fit is applied to the data to extract the frequency of the $^1\mathrm{S}_0 \to {}^1\mathrm{P}_1$ transition for each of the ytterbium isotopes. Fig. 2 shows the spectrum obtained with the WS8-2. The common estimated full width at half-maximum (FWHM) of 37 MHz is larger than the 28 MHz natural linewidth. This minor broadening is mainly attributed to laser linewidth (~ 4 MHz), power broadening (~ 2 MHz) and Doppler broadening due to the transverse velocity distribution in the atomic beam.

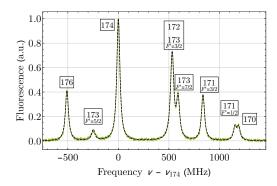


Fig. 2. Fluorescence spectrum of ytterbium atoms, where the laser frequency is measured with the WS8-2. Green points: experimental data. Black dashed curve: Lorentzian fit with common FWHM. Frequencies are plotted relatively to the ¹⁷⁴Yb isotope line.

3.1. Transition frequency of the ¹⁷⁴Yb isotope

Our measurement, including all sources of uncertainty (see discussion section 4), returns the value 751 526 528 \pm 60 MHz for the frequency of the $^1S_0 \rightarrow ^1P_1$ transition in the $^{174}\mathrm{Yb}$ isotope when measured with the WS/7, and 751 526 540 \pm 30 MHz when measured with the WS8-2. These results are compared with previously reported values in Fig. 3. No uncertainty is provided for the measurement from [7], but Kleinert *et al.* gave a $\pm150\,\mathrm{MHz}$ estimate for this datapoint from discussions with the NIST team [11]. In [10], the authors do not provide a frequency value for the $^{174}\mathrm{Yb}$ isotope. For comparison purposes, we use the value of 751 526.65 \pm 0.11 GHz calculated from their measurement for the $^{171}\mathrm{Yb}$ isotope translated by an isotopic shift of 832.6 MHz, the weighted mean [22] of the shifts measurements from [8,9,11,13–16]. An uncertainty of 10 MHz was added to account for the uncertainty on the isotopic shifts measurements.

The reported values are spread out over 600 MHz. When restraining the analysis to values from [7,9–11] as well as ours, we find a reasonable agreement, with error bars still covering nearly 400 MHz. Within the error bars, our results are compatible with [7, 10, 11], and our WS8-2 measurement differs from [9] by a few MHz only. By doing a simple averaging of all 6 values, we find $\bar{v}_{174} = 751\,526\,574 \pm 35\,\text{MHz}$, while a weighted mean [22] yields $\tilde{v}_{174} = 751\,526\,533.5 \pm 0.3\,\text{MHz}$.

3.2. Other isotopes

Table 1 displays our measurements of the ytterbium $^1S_0 \rightarrow ^1P_1$ transition absolute frequencies for the different isotopes and hyperfine transitions. The uncertainty of the wavemeters when measuring a frequency difference over the \sim GHz range is specified by the manufacturer to be equal to the absolute accuracy. For this reason, we do not find necessary to present a thorough comparison of our results with the isotopic shifts measurements found in literature, which have

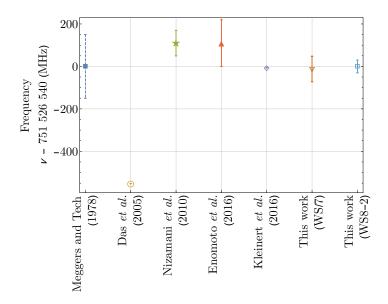


Fig. 3. Absolute measurements of the ^{174}Yb $^{1}\text{S}_{0} \rightarrow ^{1}\text{P}_{1}$ transition frequency. Blue square: [7]; yellow circle: [8]; green star: [9]; red upward pointing triangle: calculated from [10]; purple diamond: [11]; brown downward pointing triangle: this work with WS/7; light blue square: this work with WS8-2. Frequencies are plotted relatively to the atomic transition in the ^{174}Yb isotope as measured in [7].

a much smaller dispersion than this uncertainty. Our results are nevertheless in very good agreement with all the isotopic shifts measurements found in [8,9,11,13–16].

It should be noted that even though the wavemeters manufacturer recommends using the absolute uncertainty for frequency difference measurements, this seems to be a too conservative estimate. To verify this, we successively locked an 870 nm ECDL to adjacent teeth of a frequency comb. The teeth had a fixed spacing of about 250 MHz and stable well below the MHz level. By reading the frequency-doubled output of this laser with the wavemeters after each 250 MHz step, we checked that the uncertainty on frequency difference measurements was below 6 MHz for both the WS/7 and WS8-2 wavemeters across a \sim 7 GHz range around 435 nm. We are unfortunately unable to perform this test around 399 nm to verify that our measurement could be used to estimate the isotope shifts at the few MHz level.

4. Sources of uncertainty

4.1. Measurement method and frequency scale

We record fluorescence spectra by acquiring a time trace of the fluorescence signal with an oscilloscope while simultaneously monitoring the laser frequency with the wavemeters. During a measurement the 100 s-long frequency scan is made over a range of 3 GHz, which induces uncalibrated non-linearity when using the laser PZT. In practice, we sample the laser frequency every ~ 25 ms using the wavemeters and interpolate between the measurements to convert the oscilloscope time trace to a frequency scale. The error on the frequency scale origin from this conversion results in an uncertainty corresponding to one time step at maximum, and is thus evaluated to be below 0.8 MHz.

	Transition frequency		
Isotope	WS/7 (MHz)	WS8-2 (MHz)	Weighted Mean (MHz)
¹⁷⁶ Yb	751 526 021(60)	751 526 031(30)	751 526 029(27)
¹⁷³ Yb	751 526 280(60)	751 526 292(30)	751 526 289(27)
¹⁷² Yb & ¹⁷³ Yb (F'=3/2)	751 527 063(60)	751 527 072(30)	751 527 070(27)
¹⁷³ Yb (F'=7/2)	751 527 122(60)	751 527 131(30)	751 527 129(27)
¹⁷¹ Yb (F'=3/2)	751 527 370(60)	751 527 378(30)	751 527 376(27)
¹⁷¹ Yb (F'=1/2)	751 527 689(60)	751 527 694(30)	751 527 693(27)
¹⁷⁰ Yb	751 527 725(60)	751 527 730(30)	751 527 729(27)

Table 1. Absolute measurement of the frequency of the $^1S_0 \rightarrow ^1P_1$ transition depending on the isotope and the hyperfine transition. A single measurement is provided for 172 Yb and $F = 5/2 \rightarrow F' = 3/2$ in 173 Yb as the two resonances are not distinguishable.

4.2. Systematic errors

In addition to the uncertainty inherent to the measurement method, different sources of error of physical origin can be identified, including frequency shifts and broadenings due to the laser intensity and linewidth, the residual magnetic fields and the laser polarization, and the Doppler effect.

First, the resonances in the recorded spectrum have no detectable amount of asymmetry. The position of their maxima stays within the error bar returned by the fit, which is found to be sub-MHz for all of them. It is therefore very conservative to assume an error of ± 1 MHz from the combined effects leading to a broadening of the resonances, including non-linearity of the frequency measurement over the linewidth range.

The experiment, not protected by a magnetic shield, is subject to the earth field and environmental background fields. The residual magnetic field around the chamber was measured to have an amplitude below 1 G leading to a splitting of the transitions of less than 1.4 MHz for both odd and even isotopes. Moreover, the laser polarization is random. In these conditions, Zeeman splitting leads to a minor asymmetric broadening of the recorded resonances (natural linewidth of 28 MHz) and sub-MHz shifts of the maxima. For odd isotopes, quantum interferences and polarization effects [11,23] can also lead to sub-MHz shifts of the resonances. In the particular case of the $F = 5/2 \rightarrow F' = 3/2$ hyperfine transition in 173 Yb, very specific polarizations of the laser with respect to the residual magnetic field would result in an optical pumping effect [24] and forbid detection of fluorescence for this transition. We do not claim in this work to measure separately transition frequencies for 172 Yb and 173 Yb $_{F=5/2\rightarrow F'=3/2}$, and the presence or the absence of the 173 Yb $_{F=5/2\rightarrow F'=3/2}$ peak in the fluorescence spectrum can only induce sub-MHz level uncertainty on the common measurement largely dominated by the 172 Yb transition frequency value. In conclusion, residual magnetic fields and polarization effects are conservatively considered to induce an overall error of ± 3 MHz on the transition frequency.

Doppler shifts can be caused by the non-perpendicularity of the laser beam relatively to the atomic beam direction, which is not perfectly known inside the vacuum chamber. The longitudinal velocity distribution of the atomic beam is centered around a non-zero value, which leads to a Doppler shift depending on the angle θ . From the temperature of the dispenser $T \sim 875$ K, we can roughly estimate this shift [25] to $\frac{\mathbf{v}_{at}\sin\theta}{\lambda} \sim \sin\theta \times 725$ MHz, with \mathbf{v}_{at} the most probable speed and λ the laser wavelength. This amounts to ~ 13 MHz/ $^{\circ}$ at small angles. This sensitivity

can thus induce a significant contribution to the systematic error budget. In order to reduce the uncertainty due to this effect, we recorded the fluorescence spectrum at different angles θ using the previously described retro-reflection mirror (section 2 and Fig. 1).

Figure 4 shows recorded fluorescence spectra for different values of the angle θ . For each θ value, each resonance peak is split into a doublet of separate resonances corresponding to opposite Doppler shifts. For clarity, spectra have been normalized to the spectrum obtained with $\theta = 1^{\circ}$. A horizontal average has also been applied in order to smooth out the curves for better readability. For analysis, the data have been fitted with the sum of two translated spectra which include all isotopes lines (one for each way of the laser propagation). In the fitting function, each line is considered a Voigt profile, which reduces processing to an acceptable time, and the symmetrical Doppler shift of each of the two spectra is left as a common free parameter. Error bars returned by the fits are all sub-MHz regarding the Doppler shift results. These shifts are then plotted as a function of the angle θ and finally linearly fitted as a function of $\sin \theta$ in order to find the position for which the laser beam is perpendicular to the most probable velocity, ie $\theta = 0^{\circ}$. These results are presented in the inset of Fig. 4. The 1° data point has been discarded in this last analysis even though the fitting process of the recorded spectrum seems to return a reliable result. The fit indicates a $\sin \theta \times (787 \pm 5)$ MHz dependence of the Doppler shift. Given the error on the perpendicular position from the analysis $(\pm 0.3^{\circ})$ and the angular resolution of the rotatable breadboard (5' $\sim 0.083^{\circ}$), we estimate the uncertainty on frequency measurements caused by this parameter to be ±4.3 MHz. Note that an asymmetrical Doppler broadening due to the same effect is to be expected on the final recorded spectrum, but this has also been minimized by the above procedure.

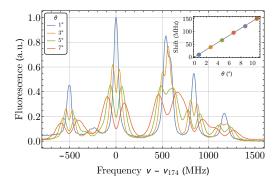


Fig. 4. Fluorescence spectra of ytterbium atoms as a function of the laser frequency while retro-reflecting the probing beam at different angles: $\theta \sim 1^\circ$ (blue), $\theta \sim 3^\circ$ (yellow), $\theta \sim 5^\circ$ (green), $\theta \sim 7^\circ$ (red). Frequencies are plotted relatively to the atomic transition in the ¹⁷⁴Yb isotope. Inset: Doppler shift as a function of the angle θ and associated linear fit indicating the zero-shift angle for perpendicular configuration.

4.3. Overall uncertainty

All pre-mentionned uncertainties have been summarized in Table 2 as well as the accuracy for each wavemeter. From these results, we deduce an overall uncertainty of $\pm 60\,\text{MHz}$ for the WS/7 measurement and $\pm 30\,\text{MHz}$ for the WS8-2. With all other sources of uncertainties having a very small contribution, the overall uncertainty is completely determined by the wavemeters.

Using these uncertainties and deriving the weighted mean of our two results, the absolute frequency of the $^1S_0 \rightarrow {}^1P_1$ transition of ^{174}Yb is found to be 751 526 537 \pm 27 MHz. For other isotopes, the weighted averages have been indicated in Table 1.

Source of error	Uncertainty (±MHz)
Frequency scale conversion	0.8
Line broadening effects	1
Residual magnetic field and laser polarization	3
Laser/atomic beams non orthogonality	4.3
Wavemeter WS/7	60
Wavemeter WS8-2	30

Table 2. Summary of the conservative estimates on the uncertainties affecting the $^1S_0\to\,^1P_1$ transition measurement.

5. Conclusion

In this article, we have reported on two independent measurements of the $^1S_0 \rightarrow ^1P_1$ transition frequency for different isotopes of neutral ytterbium. The measurements were done by recording fluorescence spectra using two independent commercial wavemeters. We implemented a method based on counterpropagating beams that minimizes the uncertainty due to the Doppler shift, by identifying the angle for which the laser beam is perpendicular to the atomic beam.

By presenting these results, we hope to reduce the existing uncertainty on this transition frequency, suffering from a non-negligible dispersion in the literature. Our results confirm in particular the most recent accurate measurement by Kleinert *et al.* [11] from 2016. This might be useful to future starting experiments based on neutral and ionized ytterbium where knowing accurately this transition frequency is crucial.

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Disclosures

The authors declare that there are no conflicts of interest related to this article.

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