Spectroscopy of the $^2S_{1/2} \rightarrow ^2P_{3/2}$ transition in Yb II: Isotope shifts, hyperfine splitting and branching ratios

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We report on spectroscopic results on the $^2S_{1/2} \rightarrow ^2P_{3/2}$ transition in single trapped Yb$^+$ ions. We measure the isotope shifts for all stable Yb$^+$ isotopes except $^{173}$Yb$^+$, as well as the hyperfine splitting of the $^2P_{3/2}$ state in $^{174}$Yb$^+$. Our results are in agreement with previous measurements but are a factor of 5-9 more precise. For the hyperfine constant $A(^2P_{3/2}) = 875.4(10)$ MHz our results also agree with previous measurements but deviate significantly from theoretical predictions. We present experimental results on the branching ratios for the decay of the $^2P_{3/2}$ state. We find branching fractions for the decay to the $^2D_{3/2}$ state and $^2D_{5/2}$ state of 0.17(1)% and 1.08(5)%, respectively, in rough agreement with theoretical predictions. Furthermore, we measured the isotope shifts of the $^2F_{7/2} \rightarrow ^1D[5/2]_{5/2}$ transition and determine the hyperfine structure constant for the $^1D[5/2]_{5/2}$ state in $^{171}$Yb$^+$ to be $A(^1D[5/2]_{5/2}) = -107(6)$ MHz.

I. INTRODUCTION

Laser-cooled ions in Paul traps form one of the most mature laboratory systems for performing optical metrology, precision measurements as well as quantum-computation and quantum-simulation [1-5]. The ion species Yb$^+$ is a particularly versatile system for many of these applications owing to its rich electronic structure with multiple meta-stable states [6,7]. Furthermore, the hyperfine structure of $^{171}$Yb$^+$ provides a first-order magnetic field-insensitive qubit in the electronic ground state [8-10] that may be used in quantum information applications [11,12].

While many transitions between low-lying electronic states in Yb$^+$ have been studied with great precision [10,13-16], there has been only one measurement of the isotope shifts in the $^2S_{1/2} \rightarrow ^2P_{3/2}$ (D2) transition as well as of the hyperfine splitting of the $^2P_{3/2}$ state [17] so far, which was performed in a hollow-cathode discharge lamp. Remarkably, the experimental result for the hyperfine splitting disagrees with theoretical predictions [18-22]. Although there has been a lot of theoretical work on transition amplitudes for the decay of the $^2P_{3/2}$ state [19-21,23,24], there seems to be no experimental data available for the branching ratios of the decay of the $^2P_{3/2}$ state up until now.

Here, we present experimental results on the isotope shifts in the D2-transition, the hyperfine splitting of the $^2P_{3/2}$ state as well as on the branching ratios of its decay obtained from a single trapped and laser-cooled ion. Furthermore, we present measurements of the isotope shift in the $^2F_{7/2} \rightarrow ^1D[5/2]_{5/2}$ transition, as well as the hyperfine splitting in the $^1D[5/2]_{5/2}$ state in $^{171}$Yb$^+$. Single trapped ions are very well suited to perform such precision measurements, as both state preparation and detection can be performed with great accuracy while at the same time errors due to back-ground gas collisions are negligible. Using isotope-selective photo-ionization to load the Paul trap, we are able to conduct the experiments even with the rare isotope $^{168}$Yb$^+$ (0.13% abundance, [25]) for which no previous data seems to exist.

II. EXPERIMENTAL SETUP

The experiments have been performed in a linear Paul trap described in Ref. [26]. We load a single Yb$^+$ ion into the trap by two-step photo-ionization with lasers at 399 nm wavelength for the resonant excitation of the $^1S_0 \rightarrow ^1P_1$ transition in neutral Yb and 369 nm wavelength for the excitation into the ionization continuum. Tuning the wavelength of the first step to the resonance of a specific isotope allows for isotope selective loading of Yb$^+$ ions. Due to overlapping resonances [27], $^{170}$Yb$^+$ and $^{172}$Yb$^+$ cannot be loaded deterministically, but only in combination with $^{171}$Yb$^+$ and $^{173}$Yb$^+$, respectively. However, by temporally lowering the trap drive amplitude we can expel the heavier isotopes from the trap and keep only the isotope $^{170}$Yb$^+$ or $^{172}$Yb$^+$, respectively.

Lasers near wavelengths of 369 nm and 935 nm are used to Doppler cool the ion on the $^2S_{1/2} \rightarrow ^2P_{1/2}$ transition and pump population trapped in the metastable $^2D_{3/2}$ state back into the cooling cycle via excitation to the $^3D[3/2]_{1/2}$ state, as shown in Fig. 1a. We image the ion’s fluorescence at 369 nm wavelength to a photo-multiplier-tube (PMT) for detection.

For cooling of the isotope $^{171}$Yb$^+$, which has a nuclear spin of $I = 1/2$ and accordingly hyperfine splittings of the electronic states, we use the closed transition $^2S_{1/2}, F = 1 \rightarrow ^2P_{1/2}, F = 0$. However, due to off-resonant excitation of the $^2P_{1/2}, F = 1$ state, the ion occasionally decays to the $^2S_{1/2}, F = 0$ state. Microwave radiation at 12.6 GHz couples the $F = 0$ and $F = 1$ ground states to ensure continuous cooling. In order to prepare the ion in the $^2S_{1/2}, F = 0$ state, we...
We perform spectroscopic measurements on the $^2S_{1/2} \rightarrow ^2P_{1/2}$ transition near 329 nm wavelength. The $^2P_{3/2}$ state decays in $\tau = 6.15(9)$ ns [28] either back into the ground state or into one of the metastable states $^2D_{5/2}$ or $^2D_{3/2}$. While an ion which is initially in the states $^2S_{1/2}$ or $^2D_{3/2}$ scatters light during Doppler cooling (thin gray arrows), it will not scatter light when it is in the $^2D_{5/2}$ state. This allows for detection of a successful excitation of the $^2S_{1/2} \rightarrow ^2P_{3/2}$ transition as well as for measurement of the branching fractions of the $^2P_{3/2}$ decay. From the $^2D_{5/2}$ state the ion decays in $\tau = 7.2(3)$ ns [13] to either the ground state or the very long lived $^2F_{7/2}$ state ($\tau \approx 10$ yr [29]). We use the $^2F_{7/2} \rightarrow ^1D_{5/2}$ transition near 638 nm wavelength to depopulate the $^2F_{7/2}$ state.

Excite the $\mid ^2S_{1/2}, F = 1 \rangle \rightarrow \mid ^2P_{1/2}, F = 1 \rangle$ transition resonantly while the microwave radiation is switched off.

In addition to the lasers required for cooling and detection of the ion, we use light near the wavelengths of 329 nm and 638 nm to drive the transitions $^2S_{1/2} \rightarrow ^2P_{3/2}$ and $^2F_{7/2} \rightarrow ^1D_{5/2}$, respectively. We generate light at 329 nm wavelength with a frequency quadrupled, amplified diode laser. After the first doubling cavity is coupled to a commercial waveguide. The laser is stabilized to the wavelength meter to compensate for frequency drifts and has a short-term frequency accuracy of 60 MHz according to specification.

We generate light at 638 nm wavelength with a home-built ECDL. This laser is stabilized to the wavelength meter to compensate for frequency drifts and has a short-term frequency stability of better than 10 MHz. We switch the light with a mechanical shutter. The light is coupled to a fiber EOM which allows for modulating sidebands in order to drive transitions between multiple hyperfine states in the case of $^{171}$Yb$^+$. The laser at 638 nm wavelength is a home-made external cavity diode laser (ECDL) with a grating in Littrow configuration. It is frequency stabilized to the wavelength meter and switched by a mechanical shutter. Using a fiber EOM, sidebands can be modulated in order to drive transitions between multiple hyperfine states.

A mechanical shutter prevents any light from reaching the ion if switched off. Light from the first doubling cavity is coupled to a commercial wavelength meter, allowing for a course absolute frequency determination of the frequency-quadrupled light with an accuracy of 60 MHz according to specification.

We generate light at 638 nm wavelength with a home-built ECDL. This laser is stabilized to the wavelength meter to compensate for frequency drifts and has a short-term frequency stability of better than 10 MHz. We switch the light with a mechanical shutter. The light is coupled to a fiber EOM which allows for modulating sidebands in order to drive transitions between multiple hyperfine states in the case of $^{171}$Yb$^+$, and guided to the experiment. The part of the setup relevant for the spectroscopy is shown in Fig. 2.

We use an acousto-optical modulator (AOM) in double-pass configuration with a center frequency of 200 MHz and a bandwidth of 100 MHz. The signals for the AOM and the fiber EOM are generated by a two channel microwave generator, which is stabilized to a 10 MHz reference signal from a signal generator. A mechanical shutter prevents any light from reaching the ion if switched off. Light from the first doubling cavity is coupled to a commercial wavelength meter, allowing for a course absolute frequency determination of the frequency-quadrupled light with an accuracy of 60 MHz according to specification.

We generate light at 638 nm wavelength with a home-built ECDL. This laser is stabilized to the wavelength meter to compensate for frequency drifts and has a short-term frequency stability of better than 10 MHz. We switch the light with a mechanical shutter. The light is coupled to a fiber EOM which allows for modulating sidebands in order to drive transitions between multiple hyperfine states in the case of $^{171}$Yb$^+$, and guided to the experiment. The part of the setup relevant for the spectroscopy is shown in Fig. 2.

**III. RESULTS**

### A. Isotope shifts and hyperfine splitting

In case of the isotopes without nuclear spin, we measure the resonance frequency of the D2-transition by applying laser pulses with a width of 5 µs and a saturation parameter $s \approx 1$ to the ion. From the $^2P_{3/2}$ state,
TABLE I. Isotope shifts of the \( ^2P_{3/2} \) state in Yb\(^+\) as measured by our group compared to values from Ref. [17]. The number in brackets denotes the error in the last digit. The isotope shift of the \( ^2F_{7/2} \rightarrow 1D_{5/2} \) transition as measured with the wavelength meter is given in the last column.

| Isotope Mass Number | \( ^2P_{3/2} - \text{this work} \) | \( ^2P_{3/2} - \text{Ref. [17]} \) | \( ^2F_{7/2} \rightarrow 1D_{5/2} \) |
|---------------------|----------------------------------|----------------------------------|----------------------------------|
| 168                 | 3007.8(30) MHz                   | -                               | 6.04(2) GHz                     |
| 170                 | 1457.9(30) MHz                   | 1459(21) MHz                     | 2.93(2) GHz                     |
| 171                 | 922.5(25) MHz                    | 920(15) MHz                      | 1.87(2) GHz                     |
| 172                 | 0                               | 0                               | 0                               |
| 174                 | -1152.3(15) MHz                  | -1154(11) MHz                    | -2.26(2) GHz                    |
| 176                 | -2254.8(15) MHz                  | -2259(13) MHz                    | -4.41(2) GHz                    |

There is a probability of about 1\% for the ion to decay to the metastable state \( ^2D_{5/2} (\tau = 7.2 \text{ ms}) \) from where it decays with 83\% probability [13] to the long-lived \( ^2F_{7/2} \) state. An ion in either of these states does not scatter light during Doppler cooling. On resonance, about 50\% of the population decays to the dark \( ^2D_{5/2} \) state in 5 \( \mu \text{s} \). To detect whether the ion is in one of the dark states, we image the ion’s fluorescence to a PMT for 4 \( \text{ms} \) during Doppler cooling, allowing for almost perfect state detection. We scan the laser over the atomic resonance in steps of 2 \( \text{MHz} \) by tuning the drive frequency of the AOM (\( \nu_{\text{aom}} \)). We compensate for the frequency-dependence of the diffraction efficiency in the AOM by supplying appropriate radio-frequency power at each frequency. After the detection, we pump the ion back into the cooling cycle by exciting the \( ^2F_{7/2} \rightarrow 1D_{5/2} \) transition. A post-selection measurement is performed before each spectroscopy pulse in order to check if the ion was successfully pumped out of the \( ^2F_{7/2} \) state. The measurement data for a single scan of the transition in \( ^{174}\text{Yb}^+ \) is plotted in Fig. 3a.

We repeat the experiment for the isotopes \( ^{168}\text{Yb}^+, \^{170}\text{Yb}^+, \^{172}\text{Yb}^+, \^{174}\text{Yb}^+ \) and \( ^{176}\text{Yb}^+ \). For each measurement, we frequency-stabilize the laser to the same cavity mode, but with different offset frequencies \( \nu_{\text{aom}} \) given by the modulation frequency of the fiber EOM. The relative frequency of the spectroscopy light compared to the fixed cavity resonance is determined by \( \nu_{\text{rel}} = 2\nu_{\text{aom}} - 2\nu_{\text{OM}} \). We estimate the drift of the cavity by measuring the same resonance at different times as shown in Fig. 3c.

The uncertainties in the energy shifts are given by the statistical error of the fitted resonance and the uncertainty in the cavity drift during the measurement time. In principle the error should not depend on the abundance of the isotope. However, the measurements with the rare isotopes \( ^{168}\text{Yb}^+ (0.13\% \text{ abundance}) \) and \( ^{170}\text{Yb}^+ (3.05\% \text{ abundance}) \), can only be loaded in combination with \( ^{172}\text{Yb}^+ \) [25] take significantly longer due to low ion loading rates. This leads to larger uncertainties for the cavity drift. We use \( \pi \)-polarized light and a small magnetic field of \( B = 0.05 \text{ mT} \) to avoid errors due to Zeeman-shifts of the D2-transition. The results are shown in Fig. 3d and Tab. I.

In case of \( \^{171}\text{Yb}^+ \) we make use of the hyperfine structure (see Fig. 1b) in order to detect the excitation to the \( ^2P_{3/2} \) state. We prepare the ion in the \( |F = 0 \rangle \) ground state before transferring it to the \( |F = 1, m_F = 0 \rangle \) state via rapid adiabatic passage (RAP) using microwave radiation. We apply a laser pulse with a width of 200 ns to excite the \( ^2P_{3/2}, F = 1 \) or \( ^2P_{3/2}, F = 2 \) states, followed by a second RAP pulse on the \( |F = 0 \rangle \rightarrow |F = 1, m_F = 0 \rangle \) transition. At the end of this sequence, we perform state-selective fluorescence detection, based
TABLE II. Hyperfine structure constants $A$ in $^{171}$Yb$^+$ in MHz. The number in brackets denotes the error in the last digit. The magnetic-dipole hyperfine structure constant $A(2P_{3/2})$ measured by our group is consistent with previous measurements (Exp.) but deviates significantly from theory (Th.) predictions. In Ref. [20] results are given for a single-electron approach ($a$) and a many-electron approach ($b$). The many-electron results give the best agreement with our experimental results by far.

|         | $^{18}$Th. | $^{19}$Th. | $^{20}$aTh. | $^{20}$bTh. | $^{21}$Th. | $^{22}$Th. | $^{17}$Exp. | This work  |
|---------|-----------|-----------|------------|------------|-----------|-----------|------------|------------|
| $A(2P_{3/2})$ | 391       | 311.5     | 330        | 765        | 322       | 388       | 877(16)    | 875.4(10)  |
| $A(1D[5/2]_{5/2})$ | -         | -         | -          | -          | -         | -         | -          | -107(6)    |

We calculate the isotope shift of the $^{2}P_{3/2}$ state given in Tab. I.

During the spectroscopy of the $^{2}S_{1/2} \rightarrow ^{2}P_{3/2}$ transition, the $^{2}F_{7/2}$ state with a lifetime of $\tau > 10$ yr [29] is populated via decay of the $^{2}D_{5/2}$ state. After the state-detection, we pump population out of the $^{2}F_{7/2}$ state by excitation of the $^{2}F_{7/2} \rightarrow ^{1}D[5/2]_{5/2}$ transition. Wavelength meter readings during the measurement give us the isotope shifts of this transition with an accuracy of 20 MHz. To the best of our knowledge, this is the most complete (in terms of measured isotopes) and precise measurement of the isotope shifts of the $^{2}F_{7/2} \rightarrow ^{1}D[5/2]_{5/2}$ transition. In $^{171}$Yb$^+$ we drive the transitions $|F = 4 \rangle \rightarrow |F = 3 \rangle$ and $|F = 3 \rangle \rightarrow |F = 2 \rangle$ (see Fig. 1b). We use sidebands at 3940 MHz to excite both transitions efficiently. With a hyperfine splitting of the $^{2}F_{7/2}$ state of 3620(2) MHz [14], we determine an energy splitting of the upper $^{1}D[5/2]_{5/2}$ state of 320(20) MHz and a hyperfine constant $A(1D[5/2]_{5/2}) = -107(6)$ MHz. There seems to be no previous experimental data available for the $^{1}D[5/2]_{5/2}$ hyperfine splitting, only a theoretical estimation of $A = 199$ MHz [31], which deviates significantly from the value we find in the experiment.

Experiments with $^{171}$Yb$^+$ are conducted in a magnetic field of 0.18-0.23 mT in order to allow for efficient Doppler cooling on the $^{2}S_{1/2}, F = 1 \rightarrow ^{2}P_{1/2}, F = 0$ transition [10]. We use linear polarized light ($\sigma^+ + \sigma^-$) at 329 nm wavelength to avoid the dipole forbidden $^{2}S_{1/2}, F = 1, m_F = 0 \rightarrow ^{2}P_{3/2}, F = 1, m_F = 0$ transition. Due to the symmetric excitation of the transition (see Fig. 1b) the magnetic field should not lead to a frequency shift of the transition. Measurements at different fields corroborate this assumption.

We repeat the experiment for both hyperfine states and determine the hyperfine energy splitting given in Tab. II. Together with the well-known energy splitting of the ground state of $12642.812$ MHz [9] we calculate the isotope shift of the $^{2}P_{3/2}$ state given in Tab. I.

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B. Branching fractions

To measure the branching fractions for decay out of the $^{2}P_{3/2}$ state, we excite the D2-transition with a short pulse of resonant light, followed by fluorescence detection of 100 $\mu$s duration. During the detection, an ion initially in the $^{2}S_{1/2}$ or $^{2}D_{3/2}$ state scatters light, while an ion in the $^{2}D_{5/2}$ state appears dark. Scanning the pulse width, we obtain the time constant $\tau_{\text{dark}} = 1.04(4)$ $\mu$s for the exponential decay of the PMT counts versus pulse width plotted in Fig. 4. From this time constant, the lifetime of the $^{2}P_{3/2}$ state of $\tau_{p32} = 6.15(9)$ ns [28] and the probability to be in the excited state during the laser-pulse $p_{p32}$, we determine the combined decay probability to both $D$ states as follows:

$$p(D\text{-state}) = \frac{\tau_{p32}}{p_{p32} \times \tau_{\text{dark}}} \quad (1)$$

The saturation parameter $s$ and thus the excited state...
population probability is determined by a frequency scan over the resonance. We normalize the power in the 329 nm beam during the scan by appropriate power settings of the AOM as described above. We choose a short pulse width of $t_{329} = 500$ ns to avoid saturation of the decay to the $^2D_{5/2}$ state. We measure a saturation parameter of $s = 11$, the data is shown in the inset of Fig. 4. For the branching ratio experiment we tune the laser to resonance and operate the AOM at the frequency of maximum diffraction efficiency which allows us to increase the power in the laser pulse by a factor of 2.2 compared to the scan. Accordingly we calculate a saturation parameter of $s = 24(13)$ for the branching ratio experiment, corresponding to a probability of being in the excited state of $p_{329} = 0.48(1)$. This scan is repeated frequently during the measurement to monitor intensity and frequency drifts.

The ratio between the decay to the $^2D_{5/2}$ state and $^2D_{3/2}$ state is given by the ratio between the fluorescence for a pulse width $t_{329} = 0$, all population in the $^2S_{1/2}$ state ($n(t_{329} = 0) = 0.741$), compared to the fluorescence when all population is pumped to the $D$ manifold ($n(t_{329} = \infty) = 0.104$), as indicated in Fig. 4.

We determine branching fractions to the $^2D_{5/2}$ state and $^2D_{3/2}$ state of 1.08(5)% and 0.17(1)% respectively which is in agreement with theoretical predictions from Ref. [23]. The errors include the statistical uncertainties in $\tau_{\text{dark}}$, $n(t_{329} = 0)$, $n(t_{329} = \infty)$ as well as uncertainties in the excited state population and the lifetime of the $^2P_{3/2}$ state.

The cumulative duration of the excitation pulse ($t_{329} < 12 \mu$s) and the detection bin ($t_{\text{det}} = 100 \mu$s) is small compared to the lifetimes of the $^2D_{3/2}$ ($\tau = 52.7$ ms [15]) and $^2D_{5/2}$ ($\tau = 7.2$ ms [13]) states. The probability for a decay back to the ground state during that time is less than 0.3% and thus negligible compared to other experimental errors.

**IV. CONCLUSIONS**

The isotope shifts in the $^2S_{1/2} \rightarrow ^2P_{3/2}$ transition in Yb⁺ and hyperfine splitting (in $^{171}$Yb⁺) of the $^2P_{3/2}$ state have been determined. Our results on both agree with previous results from Ref. [17] but are more precise by a factor of 5-9. Both experiments contradict theoretical predictions obtained from single-electron approaches for the hyperfine splitting by a factor 2-3. The results from a many-electron approach [20] agree much better with the experimental results. This corroborates their hypothesis that mixing with the energetically close state $4f^{13}5d6s^3 [3/2]_{5/2}$ has a strong influence on the $^2P_{3/2}$ state, which can not be described by a single-electron approach. We find the branching fractions for decay of the $^2P_{3/2}$ state to the $^2D_{5/2}$ state and $^2D_{3/2}$ states to be 1.08% and 0.17%, respectively, in rough agreement with theoretical predictions.

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**TABLE III. Branching fractions of the decay from the $^2P_{3/2}$ state:** Our work and the theoretical (Th.) predictions. In Ref. [20] results from a single-electron approach (a) and a many-electron approach (b) are given. Our values agree roughly with theoretical predictions.

| Branching from $^2P_{3/2}$ to: | $^{23}\text{Th.}$ | $^{20}\text{Th.}$ | $^{20}b\text{Th.}$ | $^{21}\text{Th.}$ | $^{24}\text{Th.}$ | This work |
|--------------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|----------|
| $^2S_{1/2}$                     | 98.77%          | 98.86%          | 99.09%          | 98.86%          | 98.83%          | 98.75(6)% |
| $^2D_{5/2}$                     | 0.21%           | 0.18%           | 0.15%           | 0.18%           | 0.19%           | 0.17(1)%  |
| $^2D_{3/2}$                     | 1.02%           | 0.96%           | 0.76%           | 0.96%           | 0.98%           | 1.08(5)%  |
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