Comparison of Two Single-Ion Optical Frequency Standards at the Hertz Level

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Abstract—We describe experimental investigations of an optical frequency standard based on a single laser-cooled $^{171}$Yb$^+$ ion confined in a radiofrequency Paul trap. The electric quadrupole transition from the $^2S_{1/2}(F=0)$ ground state to the $^2D_{3/2}(F=2)$ state at 435.5 nm is used as the reference transition. The transition was resolved with a Fourier-limited width of approximately 30 Hz. In order to investigate the instability and accuracy of a future frequency standard based on the reference transition, a first experiment was done comparing two laser frequencies stabilized individually to two separate ions in different traps. As a result we found a relative frequency instability $\sigma_f/(640\text{s})=1.5\times10^{-15}$ for the difference of the two laser frequencies. We also derived a first estimate $\Theta(2D_{3/2})=(3.9\pm1.9)e\sigma_0^2$ for the electric quadrupole moment of the $^2D_{3/2}$ state.

Keywords--single ion, optical frequency standard, frequency comparison, laser spectroscopy, quadrupole shift, Ytterbium

I. INTRODUCTION

Laser-cooled ions in radiofrequency traps offer unique possibilities for precision measurements on isolated atomic systems and for frequency metrology since long interaction times with external radiation fields are possible in an environment with small perturbations [1]. The electric-quadrupole transition from the $^2S_{1/2}$ ground state to the metastable $^2D_{3/2}$ level in $^{171}$Yb$^+$ is a promising candidate for an optical frequency standard. The natural linewidth of this clock transition is only 3.1 Hz at a wavelength of 435.5 nm (688 THz), allowing for good stability of a future frequency standard even for a single particle. The transition also has a $m_I=0$ to $m_I=0$ Zeeman component that is not subject to the linear Zeeman effect in low magnetic fields. Moreover, all the necessary wavelengths for laser cooling of the ion and spectroscopy can be generated from diode lasers, either directly or via frequency doubling. The absolute optical frequency of the $(F=0,m_I=0)-(F=2,m_I=0)$ hyperfine component of the clock transition was recently measured with a 1σ relative uncertainty of $1\times10^{-14}$ by comparison with a cesium fountain microwave frequency standard, so that it is now one of the most accurately known atomic transition frequencies in the optical wavelength range [2]. The uncertainty was dominated by a 3 Hz estimate for the frequency shift caused by the interaction of the $^2D_{3/2}$ level with electric field gradients due to stray potentials on the trap electrodes. This so-called quadrupole shift is expected to cause the largest contribution to the uncertainty of a future $^{171}$Yb$^+$ single-ion frequency standard. All other relevant systematic frequency shifts for our system, namely the quadratic Zeeman shift, blackbody ac Stark shift, quadratic Doppler and quadratic Stark shift are expected to be one to three orders of magnitude smaller than the quadrupole shift.

In order to investigate all these systematic effects we operate two trap systems simultaneously and compare two optical frequencies locked individually to two separate ions for different experimental parameters.

II. EXPERIMENT

A detailed description of the trap setup, the laser system and the measurement scheme used for spectroscopy of the clock transition is given in [3]. Briefly, we store ions in two identical radiofrequency Paul traps consisting of a 1.3 mm diameter conical ring electrode and two conical endcap electrodes located 0.5 mm from the ring center. The ions in the traps are laser cooled to the Doppler limit on the $^2S_{1/2}(F=1)$ to $^2P_{1/2}(F=0)$ transition at 369 nm. In addition, two repumping lasers at 935 nm and 638 nm are needed. For spectroscopy, the cooling and repumping lasers are blocked by mechanical shutters and the clock transition is interrogated with a pulse of the probe laser. The success of the excitation attempt is determined via the electron shelving method [4] by observing the fluorescence level on the cooling transition after opening the mechanical shutters again. While the probe laser frequency is incremented, this cycle of cooling, excitation and detection is repeated several times for each frequency. By recording the number of successful excitations divided by the total number of excitation attempts for each probe laser frequency, one obtains an absorption spectrum of the clock transition. The laser frequency is stabilized to the atomic transition by comparing the excitation probability for the laser being detuned by half the resolved linewidth to the blue and the red side of the line center respectively. In the experiment the mean difference in excitation probability of typically 30 excitation attempts on both sides of the resonance is used as the error signal for an integrating servo loop with a time constant of about 30s.

A simplified schematic of the experimental setup for the
frequency comparison is shown in Fig. 1. The light of the probe laser is split into two beams that pass through acousto-optic modulators (AOM) into trap #1 and trap #2 respectively. The AOMs allow independent control of the frequencies of the two probe light beams. Two independent servo systems are used to stabilize the two laser frequencies to the ion resonances. For the comparison experiment, we used a Fourier-limited resolution of the atomic resonance of 30 Hz for the trap #1 system and 100 Hz for the trap #2 system due to larger frequency fluctuations in the probe laser light transferred to the second trap. The frequencies applied to the AOMs by the servo loops are recorded for each system. For consecutive 10 s intervals the mean frequencies are calculated and their differences can be evaluated.

III. FREQUENCY COMPARISON

Fig. 2 shows about one hour of data from our comparison measurement. The mean frequency difference for the whole data set is \(-1.7\) Hz with a standard deviation of \(4.1\) Hz, corresponding to a relative frequency offset of about \(2.5 \times 10^{-15}\). With respect to the ability to resolve small frequency offsets, this comparison yields an improvement by nearly an order of magnitude over previous comparisons between single-ion frequency standards [5] and is comparable to the results of comparisons between caesium fountain standards. The possible influence of servo errors on the mean frequency difference has not yet been evaluated. Fig. 3 shows the instability analysis of the measurement. For an averaging time of 640 seconds the Allan standard deviation is \(\sigma_y(640s) = 1.5 \times 10^{-15}\). This value can be compared to a simple theoretical model for the quantum projection-noise-limited instability of a single-particle atomic frequency standard:

\[
\sigma^y_\nu(\tau) = C \frac{\Delta \nu}{\nu} \sqrt{\frac{T_c}{\tau}}
\]  

(1)

\(\Delta \nu/\nu\) is the fractional resolution of the atomic transition. \(T_c\) is the time for one measurement cycle including the time for interrogation of the ion with the probe laser as well as the time needed for laser cooling and detection. In a Monte Carlo simulation of the state detection and locking scheme we verified that for the case without local oscillator noise the employed lock algorithm indeed leads for times \(\tau T_c\) to an instability as given by (1) with the numerical value for the constant \(C\) being approximately equal to one. For our experimental parameters \((T_c=0.09\ s, \nu=688\ \text{THz}, \ C=1, \ \text{a maximum excitation probability of about 0.5 at resonance})\) and with the lower resolution \(\Delta \nu=100\ \text{Hz}\) of the trap #2 system, we find \(\sigma^y_\nu(640s) = 1.6 \times 10^{-15}\). This agrees very well with the experimental value, so we conclude that the instability of the measurement was projection noise limited by the trap #2 system with the lower spectroscopic resolution. From the numerical simulations, we expect the model given by (1) to be valid also for higher resolutions \(\Delta \nu\), so improving the resolution in the trap #2 system to the level achieved in the trap #1 system is expected to improve the stability of future comparisons by more than a factor of three.

IV. QUADRUPOLE SHIFT

For the measurement shown in Fig. 2, both traps were operated under identical conditions. During a second measurement an additional dc voltage \(V_0\) between the ring and the endcap electrodes of trap #2 was applied in order to produce a static electric field gradient along the symmetry axis of the trap. This gradient interacts with the electric quadrupole moment of the \(^2D_{3/2}\) level, which leads to a frequency shift of the clock transition proportional to the applied voltage. Fig. 4 shows the resulting frequency shift of trap #2 with respect to trap #1. The data point at zero voltage corresponds to the mean value derived from the data in Fig. 2. The other points represent the frequency difference \(\Delta \nu\) with respect to the long term mean value, averaged over...
approximately five minutes with error bars representing the corresponding standard deviations. According to [6], $\Delta \nu$ can be expressed as

$$
h \Delta \nu = A \cos^2(\beta) \cdot \Theta(2D_{3/2})$$

(2)

where $h$ is Planck’s constant, $A$ is the electric field gradient along the symmetry axis of the trap, $\beta$ is the angle between the trap axis and the quantization axis defined by the direction of the magnetic field at the point of the ion, and $\Theta(2D_{3/2})$ is the electric quadrupole moment of the $^2D_{3/2}$ state. For the $^2D_{3/2}(F=2, m_F=0)$ state of $^{171}$Yb$^+$ the factor $K(I,J,F,m_F)$ is equal to one. The value of $A$ can be derived from the applied dc voltage and the geometry of the trap. The direction of the magnetic field is determined from the signals of a three-axis magnetic field sensor close to the trap. The signal offsets arising from the different locations of the sensors and the ion were determined by evaluating spectra of the linear $\Delta m_F=\pm 1$ and $\Delta m_F=\pm 2$ Zeeman components of the clock transition.

Assuming good compensation of the ion’s micromotion the application of a static electric field gradient does not change the position of the ion in the trap. This means that the contribution of the quadratic Stark shift to the frequency difference measured is not changed with respect to the measurement with $V_0=0$ and we can use (2) to calculate the electric quadrupole moment of the $^2D_{3/2}$ state. With the angle $\beta=44\pm5^\circ$ the value obtained for the quadrupole moment is $\Theta(2D_{3/2})=(3.9\pm1.9)e a_0^2$ in atomic units, with $e$ being the elementary charge and $a_0$ the Bohr radius. The uncertainty of the measured quadrupole moment is determined by the statistical uncertainty of the measured frequency shifts and the precision to which the angle $\beta$ could be determined. Future measurements at different orientations of the magnetic fields will allow a more precise determination of the electric quadrupole moment of the $^2D_{3/2}$ state and help to reduce the contribution of the quadrupole shift to the uncertainty of a future frequency standard [6].

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