Sub-Hertz Optical Frequency Comparisons between Two Trapped $^{171}Yb^+$ Ions

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We compare the frequencies of the $6s\,^2S_{1/2}(F=0) \rightarrow 5d\,^2D_{3/2}(F=2)$ reference transition in $^{171}Yb^+$ for two single ions stored in independent traps. The quadrupole moment of the $5d\,^2D_{3/2}$ state is measured to be $9.32(48) \times 10^{-40}$ C m^2 and from the quadratic Stark shift the relevant scalar and tensor polarizabilities are determined to be $\alpha_S(S_{1/2}) - \alpha_S(D_{3/2}) = -6.9(1.4) \times 10^{-40}$ J m^2/V^2 and $\alpha_T(D_{3/2}) = -13.6(2.2) \times 10^{-40}$ J m^2/V^2, respectively. In the absence of external perturbations we find a mean frequency difference between the two trapped ions of 0.26(42) Hz, corresponding to a relative difference of $3.8(6.1) \times 10^{-16}$. This is comparable to the agreement found in the most accurate comparisons between cesium fountain clocks.

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It has long been pointed out that frequency standards based on narrow optical transitions in laser-cooled ions stored in radio-frequency traps have the potential to reach instabilities and systematic uncertainties beyond the capabilities of the best microwave standards [1]. Several transitions in different ions have been investigated so far [2–7] and a relative uncertainty of 3.4×10^{-15} has been demonstrated in a frequency measurement of a reference transition in $^{88}Sr^+$ relative to a cesium fountain clock [8]. Similar measurements of optical transition frequencies in $^{199}Hg^+$ [2] and $^{171}Yb^+$ [9] have been used recently to derive the so far most stringent limit on the present temporal variation of the fine structure constant [10]. Studies of the systematic frequency shifts of an optical frequency standard by measurements of its frequency relative to established microwave standards are inherently limited by the instabilities and uncertainties of the latter. These limitations can be overcome by comparing the optical frequency standard with a second one as we demonstrate in this Letter by comparing two single-ion frequency standards based on a reference transition in $^{171}Yb^+$. Here we focus on measurements of the frequency shifts of the reference transition due to the interaction of the ion with externally applied electric fields. From these measurements we infer the electric quadrupole moment of the $5d\,^2D_{3/2}$ state of $^{171}Yb^+$ and the atomic polarizabilities determining the quadratic Stark shift. Electric quadrupole moments relevant to single-ion frequency standards have also recently been measured in $^{88}Sr^+$ [4] and $^{199}Hg^+$ [11]. In the absence of externally applied perturbations we find a relative difference between our frequency standards of $3.8(6.1) \times 10^{-16}$.

The reference transition considered here is the $6s\,^2S_{1/2}(F=0, m_F=0) \rightarrow 5d\,^2D_{3/2}(F=2, m_F=0)$ electric quadrupole transition with a natural linewidth of 3.1 Hz at 688 THz (436 nm) in $^{171}Yb^+$. We use two Paul traps of identical design with ring electrode diameters of 1.4 mm. Using a radio-frequency trap drive voltage of $U_{ac} \simeq 600$ V at a frequency $\Omega \simeq 2\pi \times 16$ MHz, the ions are confined by a cylindrically symmetric pseudopotential with an axial depth of 17 eV and with secular frequencies $\omega_z \simeq \omega_x + \omega_y \simeq 2\pi \times 1.3$ MHz. In order to compensate any electric stray field, the position of the ion is monitored with an imaging resolution of better than 2 μm while the trapping potential is strongly reduced in the axial (radial) direction by adding a positive (negative) constant voltage U_{dc} to U_{ac}. Stray field compensation voltages that are applied between the trap end caps and to two additional electrodes are adjusted so that the ion does not detectably change its position when the potential depth is lowered to less than 0.25 eV. The remaining stray field induced displacement of the ion under normal operating conditions is then calculated to be less than 100 nm.

The ion is laser cooled to the Lamb-Dicke regime by a frequency doubled diode laser at 371 nm. The reference transition can be resolved with a linewidth of 10 Hz [10]. A detailed description of the experimental setup and of the spectroscopy of the reference transition is given in [6,12]. The setup used for the frequency comparisons is shown in Fig. 1. Both trap systems use the same cooling and probe lasers. Two independent frequency shift and servo systems

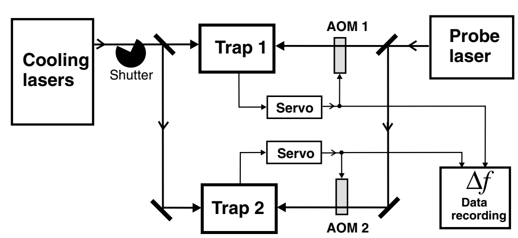

FIG. 1. Schematic of the experimental setup used to compare two $^{171}Yb^+$ optical frequency standards. Two acusto-optical modulators (AOMs) are used to independently shift the frequencies of the two probe beams interacting with the ions. A mechanical shutter blocks the cooling lasers while the probe beams interrogate the ions.

are employed to stabilize the probe beam frequencies to the line centers of the reference transitions in the two trapped ions. In order to minimize servo errors due to drift of the probe laser frequency, second-order integrating servo algorithms are used. For times larger than the servo time constants (~10 s), the optical frequencies probing the two ions can be regarded as independent and determined solely by the respective atomic transition frequencies. The frequency difference between the two servos $\Delta f = f(\mathrm{AOM2}) - f(\mathrm{AOM1})$ is recorded once per second and averaged for typically 500–700 s.

The ions are interrogated every 90 ms by 30 ms pulses. The atomic resonances show essentially Fourier limited linewidths of approximately 30 Hz and maximum excitation probabilities of about 0.6. The observed relative instability of the frequency difference Δf measured by the Allan standard deviation $\sigma_y(\tau)$ under these conditions is $\sigma_y(500\,\mathrm{s}) \simeq 7 \times 10^{-16}$, corresponding to a statistical uncertainty of the measured frequency averages of 0.5 Hz. For averaging times $\tau > 100$ s, $\sigma_y(\tau)$ follows the expected $\tau^{-1/2}$ behavior. The observed instability is independent of the temporal overlap of the probe pulses applied to the two ions and is in good agreement with numerical simulations of the servo action for the case that the fluctuations of the atomic resonance signals are dominated by quantum projection noise [13].

The limiting systematic uncertainty for most single-ion frequency standards has so far been the quadrupole shift due to the interaction of the atomic quadrupole moment with the electric stray field gradient which is usually unknown and not compensated. For an electric field gradient described in its principal axis frame by the general quadrupole potential

$$\Phi = A[(1+\epsilon)x^2 + (1-\epsilon)y^2 - 2z^2], \tag{1}$$

the quadrupole shift of the reference transition is up to first order in perturbation theory given by [14]

$$h\Delta\nu_Q = A\Theta(D_{3/2})g(\alpha, \beta), \tag{2}$$

$$g(\alpha, \beta) = 3\cos^2\beta - 1 - \epsilon\sin^2\beta\cos2\alpha. \tag{3}$$

Here, $\Theta(D_{3/2})$ is the quadrupole moment of the $5d\,^2D_{3/2}$ state, h is Planck's constant, and α, β are the first two of the Euler angles that relate the principal axes of the field gradient to the reference frame where the z axis is the quantization axis as defined by the direction of the applied static magnetic field. ϵ describes the deviation of the potential from cylindrical symmetry. The quadrupole potential (1) in the vicinity of the ion is composed of the trap potential and a contribution due to the electric stray field. For the trap potential, $\epsilon = 0$ can be assumed since ω_x and ω_y are degenerate to within 1%. The trap potential can therefore be written as

$$\Phi_{\mathrm{trap}} = (A_{\mathrm{dc}} + A_{\mathrm{ac}}\cos\Omega t)(x^2 + y^2 - 2z^2), \tag{4}$$

where $A_{\mathrm{dc}} = U_{\mathrm{dc}}/d^2$ and $A_{\mathrm{ac}} = U_{\mathrm{ac}}/d^2$. The quantity d is a characteristic length that depends on the trap geometry and can be determined experimentally from the secular frequencies. The contribution of A_{ac} to the quadrupole shift averages to zero to first order, but could lead to a considerable contribution via the second-order quadrupole shift [15]. However, the second-order shift is proportional to m_F [16] and therefore exactly zero in our case. Thus, the trapping field only causes a quadrupole shift if $A_{\mathrm{dc}} \neq 0$.

Two methods have been proposed which allow one to eliminate the quadrupole shift without *a priori* knowledge of the electric stray field gradient [8]. One method [14] exploits the fact that the sum over $g(\alpha, \beta)$ in Eq. (3), and thus the sum over the first-order quadrupole shifts, is equal to zero for any set of three mutually orthogonal directions of the magnetic field. This relation does not hold for the second-order quadrupole shift. The other method [17] uses the fact that the Hamiltonian for the quadrupole shift is traceless, which implies that the sum of the quadrupole shifts of all magnetic sublevels for a given transition is equal to zero up to any order of perturbation theory. Both schemes also eliminate the tensor part of the quadratic Stark shift, which depends on m_F and on the orientation of the magnetic field in a way analogous to the quadrupole shift (see below).

In order to investigate the quadrupole shift of the reference transition, we apply a voltage U_{dc} to the ring electrode of one trap and measure the resulting frequency shift relative to the other trap, which is kept at $U_{\mathrm{dc}} = 0$. If A_{dc} is much larger than the electric stray field gradient, we can set $A = A_{\mathrm{dc}}$ and $\epsilon = 0$. If the orientation of the magnetic field relative to the trap axis is known, the quadrupole moment $\Theta(D_{3/2})$ can be calculated from the observed frequency shift using Eq. (2). Figure 2 shows several measurements for different combinations of magnetic field orientations in the two traps. The field orientations 1, 2, 3 were established with estimated uncertainties of 20° (trap 1) and 10° (trap 2) by observing the Zeeman pattern of the $^2D_{3/2}$ $(F = 2)$ manifold for various linear polarizations of the probe beam. The absolute value of the magnetic field, determined from the Zeeman splitting, was 1.4–3.0 μT depending on the orientation used. The data shown in Fig. 2 are corrected for the resulting quadratic Zeeman shift of 0.10–0.47 Hz [12]. For the measurement of the quadrupole moment $\Theta(D_{3/2})$, the magnetic field orientation was adjusted such that $|\Delta f|$ was maximized to within the statistical measurement uncertainty in the vicinity of $\beta = 90°$. This measurement is labeled $(1, \underline{4})$ in Fig. 2. Taking into account a correction due to a 5° uncertainty in β, we find for the electrical quadrupole moment $\Theta(D_{3/2}) = 9.32(48) \times 10^{-40}$ C m^2 $= 2.08(11)ea_0^2$ (e: elementary charge, a_0: Bohr radius). This is consistent with our previous less precise measurement [6] and in good agreement with the result 9.13×10^{-40} C m^2 of a recent calculation [11].

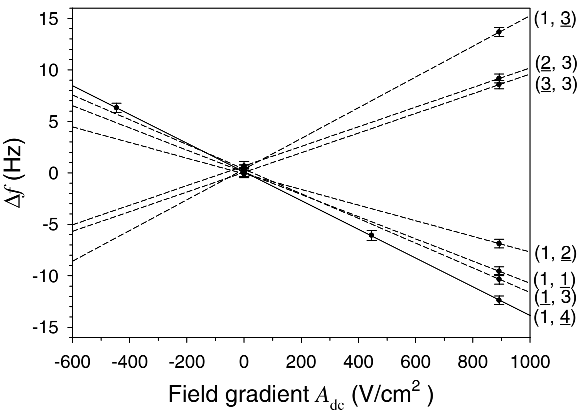

FIG. 2. Quadrupole shift as a function of applied field gradient for different orientations of the magnetic fields in trap 1 and trap 2. The error bars represent the statistical uncertainties of the measurements and the lines are the results of linear regressions. The data points at $A_{dc} = 0$ are displayed with higher resolution in Fig. 4. The numbers in brackets label the combinations of magnetic field orientations used (trap 1, trap 2), and the underlines indicate in which trap the field gradient was applied. The magnetic field orientations 1, 2, 3 are approximately orthogonal sets for both traps, while orientation 4 is perpendicular to the trap axis. The quadrupole moment of the $^2D_{3/2}$ state was determined from the $(1, \underline{4})$ measurement (solid line).

The quadratic Stark shift of the $^{171}\mathrm{Yb}^+$ reference transition is given by [14]

$$h\Delta\nu_S = [2\Delta\alpha_S + \alpha_T(3\cos^2\beta - 1)]\frac{E^2}{4}, \qquad (5)$$

where $\Delta\alpha_S = \alpha_S(^2S_{1/2}) - \alpha_S(^2D_{3/2})$ is the difference of the scalar electric polarizabilities of the ground state and the $^2D_{3/2}$ state, $\alpha_T = \alpha_T(^2D_{3/2})$ is the tensor polarizability of the $^2D_{3/2}$ state, and β is the angle between the electric and the magnetic field. To measure the polarizabilities, we displace one of the ions by a distance Δz from the trap center by adding a variable offset to the compensation voltage applied between the end cap electrodes of the trap [15]. The ion is then subject to the quadratic Stark shift caused by the time average of the ac trapping field at the displaced position. The quantities $\Delta\alpha_S$ and α_T can be determined from at least two measurements with different angles β. Figure 3 shows the dependence of Δf on the displacement Δz for the magnetic field orientation (1, 3) in Fig. 2. For this orientation $\beta = 32(3)°$ is inferred from the measured quadrupole shift. The data points taken at $\Delta z = \pm 0.7\ \mu$m and $\Delta z = 1.1\ \mu$m include corrections for the calculated second-order Doppler shifts of -0.16 Hz and -0.35 Hz, respectively. A second Stark shift measurement was done for the magnetic field orientation (1,4), where $\beta = 90(5)°$. Here no significant shifts were observed, indicating an accidental cancellation of the scalar and tensorial contributions to the Stark shift at this angle. Combining these measurements we obtain $\Delta\alpha_S = -6.9(1.4) \times 10^{-40}\ \mathrm{J m^2/V^2}$ and $\alpha_T(D_{3/2}) = -13.6(2.2) \times 10^{-40}\ \mathrm{J\,m^2/V^2}$. The experimental result is close to values calculated from available oscillator strength data [18], $\Delta\alpha_S = -4.4 \times 10^{-40}\ \mathrm{Jm^2/V^2}$ and $\alpha_T = -11.5 \times 10^{-40}\ \mathrm{J\,m^2/V^2}$ [19].

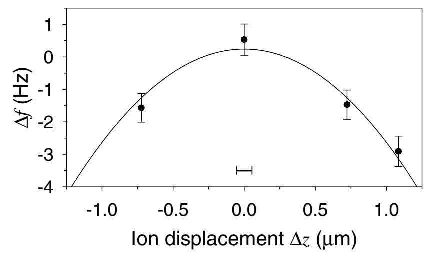

FIG. 3. Quadratic Stark shift as a function of the displacement Δz of the ion from the saddle point of the quadrupole trap potential. Δz was calculated from the voltage applied between the trap end caps. The solid line is a least-squares fit of a parabola centered at $\Delta z = 0$. The horizontal bar in the lower part represents the uncertainty range of the position $\Delta z = 0$ which remains after the compensation of the electric stray field.

Figure 4 shows measurements of the frequency difference between the ions for $U_{dc} = 0$ and $\Delta z = 0$ in both traps. The weighted mean frequency difference of all eight measurements is 0.26 Hz and the average statistical uncertainty of the individual measurements is 0.42 Hz. The contribution of the quadratic Zeeman shift correction to the systematic uncertainty of the measurement is below 0.05 Hz. Using a stray field compensation as described above, the total uncertainty contribution from quadratic Stark shift and second-order Doppler shift is below 0.01 Hz. The blackbody ac Stark shift expected from the measured polarizability $\Delta\alpha_S$ is about -0.4 Hz at 300 K and should be equal to within 0.02 Hz for both trap systems.

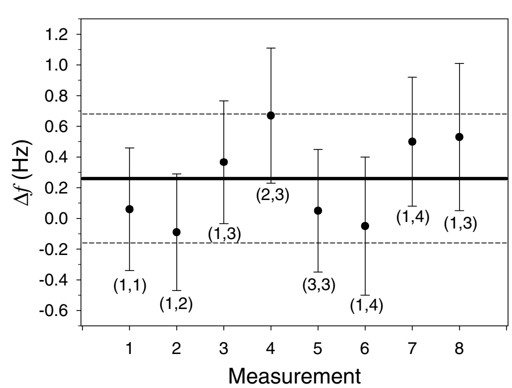

FIG. 4. Frequency difference Δf in the absence of external perturbations. Shown are the measurements at $A_{dc} = 0$ (1–6) from Fig. 2 with two more measurements added. The data were taken within two days and are displayed in temporal order. The solid line is the weighted average of the data, the dashed lines mark the average statistical uncertainty of the data points.

From measurements of the secular frequencies one can derive an estimate of the electric stray field gradients in the traps [4]. Together with the measured quadrupole moment this yields an upper limit for the quadrupole shift of $\Delta\nu_Q \leq 1.4$ Hz for both traps. Assuming that the scatter of the data in Fig. 4 is due to the orientation dependence of the quadrupole shift and of the quadratic Stark shift, and that the magnetic field orientations 1, 2, 3 are exactly orthogonal, the difference frequency Δf_0 between the ions corrected for quadrupole and tensor quadratic Stark shifts is given by

$$\Delta f_0 = \frac{1}{3}(\Delta f_{11} + \Delta f_{12} - \Delta f_{13} + \Delta f_{23} + \Delta f_{33}), \quad (6)$$

where Δf_{ij} is the frequency difference measured for the combination of magnetic field orientations (i, j). From the estimates of other systematic frequency shifts made above, one expects Δf_0 to be consistent with zero. Using measurements 1–5 from Fig. 4, we find $\Delta f_0 = 0.1(6)$ Hz, where the uncertainty is dominated by the uncertainties of the magnetic field orientations.

In order to analyze the data from Fig. 4 without any assumptions on the origin of the scatter of the data, a χ^2 test was performed for the hypothesis that the data sample represents one constant value equal to the mean of the data set. We find a probability of 84% that a random set of eight values with the same mean value as the data would give the same or a higher χ^2. Thus, within the statistical uncertainty of our measurements the data yield no evidence for frequency shifts of the reference transition that depend on the orientation of the magnetic field. For an uncertainty estimate on the observed mean frequency difference of 0.26 Hz, it therefore appears justified to regard the scatter of the data as random. Nevertheless, as it is difficult to determine the contribution of systematic shifts to the scatter of the data, we do not assume the data to represent one constant value of Δf. We thus conservatively estimate the uncertainty of the mean frequency difference as the average statistical uncertainty of the individual measurements, which is 0.42 Hz.

The relative difference that we observe between the two $^{171}Yb^+$ frequency standards is comparable to the best result found in a comparison of cesium fountain clocks [20], which are the most accurate clocks at present. This result represents an improvement by more than 2 orders of magnitude over previous comparisons between single-ion frequency standards [21]. In the future we expect to be able to investigate the interactions between a trapped ion and its environment with further reduced uncertainty. One important step in this direction will be a more accurate control of the magnetic field. By interrogation of the reference transition with longer probe pulses and by increasing the averaging time, the two $^{171}Yb^+$ frequency standards can be compared with much lower statistical uncertainties than demonstrated here. It seems therefore possible to test the performance of optical frequency standards at an accuracy level exceeding that of the best cesium clocks in the near future.

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