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Subkilohertz absolute-frequency measurement of the 467-nm electric octupole transition in $^{171}Yb^{+}$

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The $^2S_{1/2}(F=0)\rightarrow{}^2F_{7/2}(F=3,m_F=0)$ transition at 467 nm in a single trapped, laser-cooled ion of $^{171}Yb^{+}$ has been measured to be $f_{Yb^+}=642\,121\,496\,771.26(23)$ kHz using a femtosecond laser frequency comb generator. The measurement is limited by measurement statistics and by the ac Stark shift, both related to the 4.5 kHz linewidth of the probe laser at the time. The systematic shifts of the transition, including ac Stark, second-order Zeeman, quadrupole, and blackbody shifts, have been evaluated.

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I. INTRODUCTION

A forbidden optical transition in a single, cold, trapped atomic ion makes an excellent frequency reference due to its high Q ($\sim 10^{15}$ for a 1-Hz linewidth transition) and the low-perturbation environment offered by an ion trap. There are a number of ions with electric quadrupole transitions with linewidth around 1 Hz being developed as optical frequency standards, including mercury [1], strontium [2,3], and ytterbium [4]. Intercombination transitions in group IIIA ions such as indium [5] are also being developed.

The $^{171}Yb^{+}$ ion is unique among candidate species for frequency standards in that the lowest-lying excited state is the $^2F_{7/2}$ state, which decays to the $^2S_{1/2}$ ground state via an electric octupole ($E3$) transition with a lifetime on the order of 6 yr [6]. Hence the natural linewidth of this transition is extremely narrow (nanohertz) and will never be a limit on the performance of the standard, which will be set by the interaction of the ion with external fields, including the probe laser.

In this paper we describe a measurement of the absolute frequency of the 467 nm electric octupole transition in $^{171}Yb^{+}$ using a femtosecond laser frequency comb generator [7–9]. Such devices have revolutionized optical frequency metrology by providing highly accurate comparisons of microwave and optical frequencies [4,10], enabling direct measurements of optical frequencies relative to the SI second to be made, and opening up the possibility of an ‘‘optical clock’’ [11].

II. EXPERIMENTAL ARRANGEMENT

The ion is trapped in a radio-frequency Paul trap, of the endcap type [12], with tantalum electrodes and an endcap spacing of 0.56 mm [13]. Single-ion confinement times of up to one month have been observed.

The energy levels of $^{171}Yb^{+}$ are shown in Fig. 1. In this experiment, a magnetic field of 310 μT was applied perpendicular to the trap axis, lifting the m_F degeneracy of the levels shown. The field is necessary to avoid a fluorescence null that occurs for low magnetic fields on the S-P transition.

The ion is laser cooled using light at 369 nm (from a frequency-doubled Ti:sapphire laser) on the $^2S_{1/2}(F=1)\rightarrow{}^2P_{1/2}(F=0)$ transition. Microwaves at 12.6 GHz are fed into the trap by an external waveguide to prevent pumping into the $F=0$ ground state via off-resonant excitation of the $^2P_{1/2}(F=1)$ level. From the upper state of the cooling transition, the ion can also decay with a branching ratio of 7×10^{-3} [14] to the $^2D_{3/2}(F=1)$ level, which is metastable with a lifetime of 50 ms [15]. Diode laser light at 935 nm continuously illuminates the ion to pump the ion back to the ground state via the fast-decaying $^3D[3/2]_{1/2}(F=0)$ level. If the ion is excited to the $^2P_{1/2}(F=1)$ state, it can decay to the $^2D_{3/2}(F=2)$ state, which is only very weakly coupled by the 935 nm light. This leads to a dark time in the ion's fluorescence, so it is necessary to monitor the fluorescence signal for a sufficient time to distinguish these dark times from those caused by the ion being in the $^2F_{7/2}$ state.

The ion is excited to the F state in order to perform spectroscopy, but will not return to the ground state for a long time, so diode laser light at 638 nm is used to pump the ion back to the ground state via the $^1D[5/2]_{5/2}$ levels.

The 467-nm transition is driven by another frequency-doubled Ti:sapphire laser, locked by the Pound-Drever-Hall technique [16] to a high-finesse ($\mathcal{F}=250\,000$) optical cavity

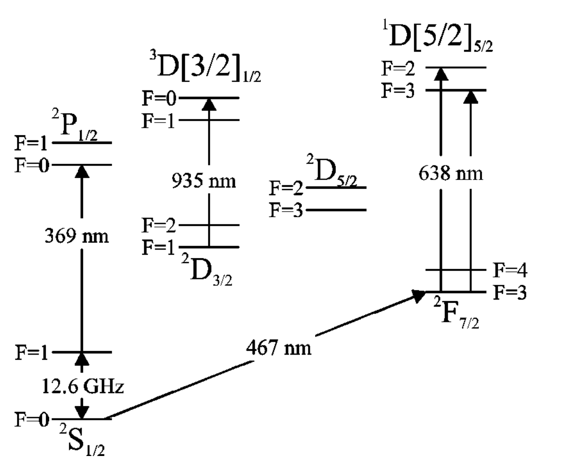

FIG. 1. Energy levels of $^{171}Yb^{+}$.

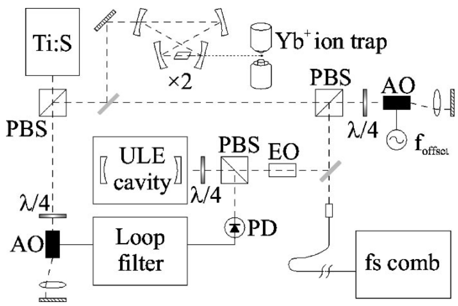

FIG. 2. Schematic of the probe laser system. EO, electro-optic modulator; AO, acousto-optic modulator; PD, photodiode, PBS, polarising beam splitter.

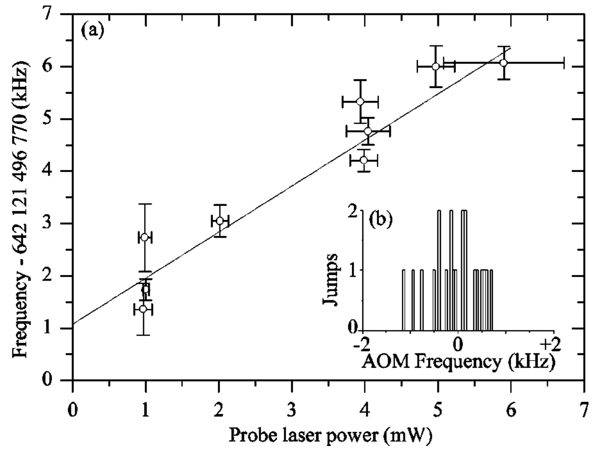

FIG. 3. (a) Frequency of the octupole transition as a function of probe laser power. (b) The spectrum of a single scan of the octupole transition, prior to combination with femtosecond comb data.

constructed from ultralow expansivity (ULE) glass (Fig. 2). The cavity is held inside a temperature-controlled cylinder inside a vacuum chamber, the outer wall of which is also temperature controlled. The frequency drift of the cavity is typically below 1 $\mathrm{Hz\,s^{-1}}$. As the octupole transition is very weak, and the probe light linewidth large [4.5 kHz full width at half maximum (FWHM)], compared to the Fourier-limited transition linewidth, a high-intensity beam is required to see a reasonable rate of quantum jumps. To achieve this, the probe laser is focused to a waist of ≈ 5 μm at the ion.

Light locked to the ULE cavity at 934 nm is picked off from the beam path after the cavity offset acousto-optic modulator and sent to the femtosecond comb via a 50-m optical fibre. The femtosecond laser is a mode-locked Ti:sapphire laser constructed from a kit supplied by KMLabs [17], of a linear cavity design with a repetition rate f_{R} of 87 MHz, and a pulse length of a few tens of femtoseconds. The output of the laser in the frequency domain is a comb of modes, spaced by the repetition rate, and with a span of about 35 nm FWHM. The comb is then broadened to more than an octave in optical frequency in a short length of microstructure optical fiber [18]. The comb modes are offset from integer multiples of f_{R} by the carrier envelope offset frequency f_{ceo}, which is measured by the self-referencing technique [8,9]. The repetition rate beat (after down-conversion by mixing with a synthesized frequency referenced to a hydrogen maser), carrier envelope offset beat and the beat of the comb with the probe laser (f_{beat}) are counted with frequency counters referenced to the 10 MHz output of a hydrogen maser. This forms part of the clock ensemble used to generate the NPL(UTC) time scale, providing traceability to the SI second at the 2.5×10^{-14} level. The repetition rate is also stabilized using the maser as a reference. The frequency of the light locked to the cavity is then determined by the equation $f_{\mathrm{laser}}=mf_{\mathrm{R}}\pm f_{\mathrm{ceo}}\pm f_{\mathrm{beat}}$, where m is the mode number of the nearest comb mode to the probe laser. The mode number and the signs of the beats can be determined provided that the frequency of the probe laser is already known to within a few MHz, as is the case here.

III. EXPERIMENTAL PROCEDURE

To align the 467-nm probe laser on the ion, a tracer beam of 369 nm light, focused to a 5-μm waist, is first aligned on the ion by monitoring the fluorescence signal. The 467-nm beam is then aligned with the tracer using an achromatic doublet lens system.

With the tracer beam aligned along the probe direction, micromotion (the driven motion of the ion when it does not sit at the center of the trap) is minimized by adjusting the voltage on two pairs of compensation electrodes. The rf-photon correlation technique [19] is used to minimize the micromotion along the probe direction and along the direction used for Doppler cooling. A third beam and compensation voltage are available but were not used in this experiment—the effect of this is discussed in Sec. IV.

To make a measurement of the transition frequency, "quantum jump" spectra of the ion are taken. First, a Doppler cooling pulse is applied, then the microwaves are switched off 60 ms before the cooling light, in order to pump into the $F=0$ ground state by off-resonant excitation of the $^2P_{1/2}(F=1)$ level. The probe laser then illuminates the ion for 330 ms. The direction of the magnetic field is set to maximize the $m_F=0\rightarrow m_F=0$ transition probability. The cooling beam and microwaves are then switched back on, and the fluorescence signal monitored for 300 ms to determine whether a quantum jump has occurred. The efficiency of this detection is very close to unity. Care is taken to ensure that occasional short term fluorescence changes due to elastic collisions are not identified as quantum jumps. The result of this is an essentially background-free quantum jump spectrum. Most scans were over a range of 20 kHz at 467 nm, in 100 steps, with five probe-pulse and detection cycles at each frequency, taking around 10 min to complete.

The femtosecond comb measures the frequency of the light at 934 nm locked to the ULE cavity, which must be combined with data from the ion spectrum as a function of the offset AOM frequency (see Fig. 3), in order to perform a measurement of the transition frequency. The data acquisition systems for the ion trap and the femtosecond comb both record the time as measurements are taken, and the data are processed afterwards. The comb measurements and the ion measurements are not necessarily exactly coincident in time. However, the cavity drift is smooth and predictable, and linear interpolation is used between comb measurements to de-

TABLE I. Estimated size of systematic frequency shifts and their standard uncertainties for this measurement. Blackbody corrections are to 0 K.

Systematic shift	Correction (Hz)	Uncertainty (Hz)
Second-order Zeeman	+185	34
Maser frequency	0	16
369 nm ac Stark	−0.6	0.3
935 nm ac Stark	+0.5	0.1
Quadrupole	±0.6	0.6
dc Stark	+0.08	0.03
Second-order Doppler	+0.05	0.02
Blackbody Stark	+0.09	0.03
Blackbody second-order Zeeman	+0.013	0.004
Total	+186	38

termine the cavity frequency at the time of each quantum jump, with an uncertainty of around 50 Hz. The statistical uncertainty on an individual comb measurement (typically 300 measurements of 1 s gate time) is about 20 Hz, insignificant compared to the frequency uncertainty of about 1 kHz (from taking a simple mean of the observed jumps) on a single scan of the octupole transition.

The largest systematic shift in the experiment is the ac Stark shift due to the probe light. A measurement of this shift as a function of probe laser power is shown in Fig. 3. The slope is consistent with the previously measured value of 42(12) μHz W^{-1}m^2 [13]. The diameter of the beam waist is stable to a higher degree of accuracy that can be measured using a micrometer-mounted knife edge. To avoid introducing errors related to measuring the beam waist, the frequency of the transition is measured as a function of probe laser power and extrapolated to zero power. A significant part of the error in this measurement is due to uncertainty in the intensity experienced by the ion. As the focus of the probe beam is so tight, the light intensity at the ion can change significantly as the alignment of optical components drifts. The alignment procedure was repeated after around every four scans of the ion to keep a check on alignment drift. Two-dimensional micromotion compensation was also performed at these intervals. The power of the 467-nm beam also drifted over the length of the scan, typically by 5%, so the power was recorded immediately before and after each scan.

Each point in Fig. 3 is derived from a collection of scans made on a particular day (42 scans were taken over four days) at a particular probe laser power. The frequency error bars are limited by statistics (the laser linewidth and the number of jumps observed), while the power error bars are the estimated power uncertainties. A weighted straight line fit to the (ungrouped) data gives an intercept of $f_0 = 642\,121\,496\,771.07(23)$ kHz (1σ).

IV. SYSTEMATIC SHIFTS

The dominant systematic shifts for the measurement are shown in Table I. After the ac Stark shift, the second-order Zeeman shift is the largest systematic shift owing to the applied magnetic field of 310 μT. The shift has a measured coefficient of 1.9(3) mHz$(\mu\mathrm{T})^{-2}$ [13]. It is possible to reduce the size of this systematic shift to well below 1 Hz by "polarization spinning" the 369-nm cooling beams [20] and working in a much smaller magnetic field of a few microtesla.

The 369-nm cooling light is switched off using an AOM, but some light leaks through the AOM in its "off" state. As this is close to resonance on the S-P transition [12.6 GHz detuned from the $^2S_{1/2}(F=0)$ level], this leads to an ac Stark shift, though this is insignificant for the current measurement. Further, the 935-nm repumper light is not switched off during the probe periods, leading to a small ac Stark shift. This is small due to the large detuning of this light from resonant transitions involving the $^2S_{1/2}$ and $^2F_{7/2}$ levels, and again is insignificant in the present arrangement. Both of these shifts can be removed by the use of mechanical shutters on the beams.

The quadrupole shift arises from the interaction of any dc component of the quadrupolar trapping field with the electric quadrupole moment of the $^2F_{7/2}$ state. Following [21], the quadrupole shift is 0.6 Hz V^{-1} for the magnetic field perpendicular to the trap axis. No additional voltage is being applied to the endcaps to equalize the trap frequencies in the radial and axial directions, and it is assumed that the source of the stray potential causing the quadrupole shift is on the endcaps of the trap. The magnitude of the quadrupole field is not well known, but is unlikely to be larger than ± 1 V. This gives a maximum quadrupole shift of ± 0.6 Hz. The quadrupole shift measured for three mutually perpendicular magnetic-field directions sums to zero [21] for constant field strength, and so the quadrupole shift can be measured and eliminated to a high degree of accuracy, limited by the precision of the measurement and knowledge of the magnitude and direction of the magnetic field. It may also be possible to reduce the stray quadrupole field in the trap by observing the secular frequencies of the trap and applying compensation voltages to the endcap electrodes [22].

The dc Stark and second-order Doppler shifts are both directly related to the micromotion of the ion. The dc Stark shift occurs as the ion experiences a nonzero electric field away from the center of the trap, while the second-order Doppler shift is a function of the speed of motion of the ion. The corrections shown in the table are calculated from typical measured micromotion amplitudes for the ion during the experiment at intervals of four scans. Minimizing micromotion more often and in three dimensions would reduce these shifts substantially.

The blackbody Stark and blackbody second-order Zeeman shifts arise from the interaction of the ion with the electric and magnetic components of the blackbody radiation field.

V. CONCLUSION

Combining the measured offset with the systematic shifts gives a value for the absolute frequency of the transition of $f_{\mathrm{Yb}^+} = 642\,121\,496\,771.26(23)$ kHz, where the 1σ uncertainty corresponds to a relative accuracy of 3.6×10^{-13}. This is in good agreement with (and four times more accurate than) the first, preliminary, measurement using the femtosecond comb of $f_{\mathrm{Yb}^+} = 642\,121\,496\,772.6(1.3)$ kHz [23] and is almost 3000 times more accurate than the previous published value of $f_{\mathrm{Yb}^+} = 642\,121\,498.1(0.8)$ MHz [6]. By reducing the probe laser linewidth (which also reduces the power required to drive the transition, and hence the ac Stark shift) to the 1 Hz level, and by working in a reduced magnetic field by using polarization spinning, it should be possible to increase this accuracy by at least another three orders of magnitude. To match this improvement in the trapped ion standard, improvements to the femtosecond comb will also be necessary, including referencing the comb to the NPL caesium fountain [24].

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[1] R.J. Rafac, B.C. Young, J.A. Beall, W.M. Itano, D.J. Wineland, and J.C. Bergquist, Phys. Rev. Lett. **85**, 2462 (2000).
[2] H.S. Margolis, G. Huang, G.P. Barwood, S.N. Lea, H.A. Klein, W.R.C. Rowley, and P. Gill, Phys. Rev. A (to be published).
[3] J.E. Bernard, A.A. Madej, L. Marmet, B.G. Whitford, K.J. Siemsen, and S. Cundy, Phys. Rev. Lett. **82**, 3228 (1999).
[4] J. Stenger, C. Tamm, N. Haverkamp, S. Weyers, and H. Telle, Opt. Lett. **26**, 1589 (2001).
[5] T. Becker, J. von Zanthier, A.Y. Nevsky, C. Schwedes, M.N. Skvortsov, H. Walther, and E. Peik, Phys. Rev. A **63**, 051802(R) (2001).
[6] M. Roberts, P. Taylor, W.R.C. Rowley, G.P. Barwood, and P. Gill, Phys. Rev. A **62**, 020501(R) (2000).
[7] T. Udem, J. Reichert, R. Holzwarth, and T.W. Hänsch, Phys. Rev. Lett. **82**, 3568 (1999).
[8] D.J. Jones, S.A. Diddams, J.K. Ranka, A. Stentz, R.S. Windeler, J.L. Hall, and S.T. Cundiff, Science **288**, 635 (2000).
[9] R. Holzwarth, T. Udem, T.W. Hänsch, J.C. Knight, W.J. Wadsworth, and P. St J. Russell, Phys. Rev. Lett. **85**, 2264 (2000).
[10] T. Udem, S.A. Diddams, K.R. Vogel, C.W. Oates, E.A. Curtis, W.D. Lee, W.M. Itano, R.E. Drullinger, J.C. Bergquist, and L. Hollberg, Phys. Rev. Lett. **86**, 4996 (2001).
[11] S.A. Diddams, *et al.*, Science **293**, 825 (2001).
[12] C.A. Schrama, E. Peik, W.W. Smith, and H. Walther, Opt. Commun. **101**, 32 (1993).
[13] S.A. Webster, P. Taylor, M. Roberts, G.P. Barwood, and P. Gill, Phys. Rev. A **65**, 052501 (2002).
[14] B.M. Fawcett and M. Wilson, At. Data Nucl. Data Tables **47**, 241 (1991).
[15] C. Gerz, J. Roths, F. Vedel, and G. Werth, Z. Phys. D: At., Mol. Clusters **8**, 235 (1988).
[16] R.W.P. Drever, J.L. Hall, F.V. Kowalski, J. Hough, G.M. Ford, A.J. Munley, and H. Ward, Appl. Phys. B: Photophys. Laser Chem. **B31**, 97 (1983).
[17] Kapteyn-Murnane Laboratories, 4699 Nautilus Court South, #204, Boulder, CO 80301, USA.
[18] J.K. Ranka, R.S. Windeler, and A.J. Stentz, Opt. Lett. **25**, 25 (2000).
[19] D.J. Berkeland, J.D. Miller, J.C. Bergquist, W.M. Itano, and D.J. Wineland, J. Appl. Phys. **83**, 5025 (1998).
[20] D.J. Berkeland and M.G. Boshier, Phys. Rev. A **65**, 033413 (2002).
[21] W.M. Itano, J. Res. Nat'l Inst. Stand. Technol. **105**, 829 (2001).
[22] P.J. Blythe, S.A. Webster, K. Hosaka, and P. Gill, J. Phys. B: At. Mol. Opt. Phys. (to be published).
[23] S.A. Webster, P. Taylor, M. Roberts, G.P. Barwood, P. Blythe, and P. Gill, in *Proceedings of 6th Symposium on Frequency Standards and Metrology*, edited by P. Gill (World Scientific, Singapore, 2002), pp. 115–122.
[24] D. Henderson, S.N. Lea, K. Szymaniec, and P. Whibberley, in *Proceedings of 15th European Frequency and Time Forum* (FSRM, Neuchatel, 2001), pp. 398–401.