Observation of an Electric Octupole Transition in a Single Ion

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The $^2S_{1/2}$-$^2F_{7/2}$ electric octupole ($E3$) transition in ^{172}Yb$^+$ has been detected by observing quantum jumps in a single laser cooled ion, stored in an electrodynamic trap. The transition frequency is 642 116 785.3(0.7) MHz (1σ). Consideration of the transition rate and laser parameters implies a $^2F_{7/2}$ level lifetime of 3700 days. This is the first time an atomic $E3$ transition has been driven. This transition has applications as an optical frequency reference. [S0031-9007(96)02175-8]

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A forbidden transition, in a single atom at rest in free space, is perhaps the ultimate spectral reference. In recent years this ideal has been approached by laser cooling a single ion stored in an electrodynamic trap and observing a weak transition using quantum jumps [1–3]. Electric quadrupole ($E2$) transitions, with linewidths of about 1 Hz, are usually used as the spectral reference. In this Letter we report the observation of an electric octupole ($E3$) transition, with a natural linewidth of 5×10^{-10} Hz. This is the first time an atomic $E3$ transition has been driven.

Alkalilike ions have a strong $^2S_{1/2}$-$^2P_{1/2}$ transition which permits high resonant scattering rates, making them well suited for laser cooling. In particular, ^{199}Hg$^+$ and ^{171}Yb$^+$ have nuclear spin $\frac{1}{2}$; therefore the forbidden transition used for the frequency reference has a component which is free from the linear Zeeman effect, while the overall atomic system has a comparatively simple hyperfine structure. In this work ^{172}Yb$^+$ is used, since the absence of hyperfine structure further simplifies laser cooling. Yb$^+$ has three candidate transitions in the optical spectrum at 411, 435, and 467 nm. The viability of the 411 and 435 nm transitions has already been demonstrated [4,5]. Until now, the $^2S_{1/2}$-$^2F_{7/2}$ transition at 467 nm (see Fig. 1) has eluded detection due to the long lifetime of the $^2F_{7/2}$ level. A previous experiment [6] set a lower limit of 8 days on the lifetime, while theoretical estimates of 650 [7] and 1500 days [8] have been made. The transition therefore has $Q \sim 10^{24}$, and this is potentially the narrowest known resonance in the optical spectrum. This unusual atomic system could have other applications where a very long lived excited state, or an unperturbed well-defined two-level system is desirable (e.g., quantum computing).

The detection of such weak transitions is facilitated by the use of electron shelving or quantum jumps [9]. With this technique, a single atom is monitored by repeatedly driving a strong ($E1$) transition, and observing the resonance fluorescence. A narrow linewidth laser is used to excite the weak reference transition. The two laser radiations are chopped in antiphase to avoid broadening this narrow reference transition. The fluorescence is interrupted when the ion is driven into the metastable state, resuming sometime later when the ion spontaneously decays back to the ground level. The signal thus consists of light and dark periods in the fluorescence, as the atom makes "quantum jumps" into and out of the metastable state. In this work the 369 nm $^2S_{1/2}$-$^2P_{1/2}$ transition is driven to both generate the resonance fluorescence and cool the ion, and the 467 nm $^2S_{1/2}$-$^2F_{7/2}$ transition is probed (see Fig. 1). In the experiment described here a slight variation of the quantum jump scheme is used. After the ion has made a jump into the $^2F_{7/2}$ level, it is returned to the ground state by driving a subsidiary transition, rather than waiting for spontaneous decay.

In order to successfully drive such a weak transition it was necessary to reduce the uncertainty in its spectral position, reduce broadening mechanisms, and use a high light intensity. The tabulated uncertainty of the 467 nm transition frequency is ± 2.6 GHz ($\pm 2\sigma$) [10]. This has been reduced to ± 8 MHz ($\pm 2\sigma$) by measurements, made at the National Physical Laboratory (NPL), of the $^2S_{1/2}$-$^2D_{5/2}$ transition [4,11] and the $^2D_{5/2}$-$^2F_{7/2}$ transition [12] (see
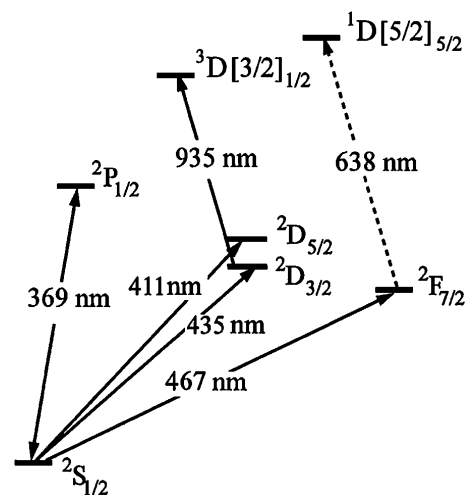

FIG. 1. Partial term scheme of Yb$^+$.

Table I). The Doppler width of the 467 nm transition was eliminated by confining the ion in a region of space much less than the wavelength of the incident light (the Lamb-Dicke regime [13]), by laser cooling the ion in a small electrodynamic trap. A high 467 nm intensity was generated by focusing 10 mW of light with a 350 kHz bandwidth into a spot of radius 12 μm. Quantum "off" jumps were observed in a 1 MHz section of the 16 MHz search region when the ion was interrogated by laser radiation at 467 nm.

A schematic layout of the experimental apparatus is shown in Fig. 2. The trap consists of a ring and two end caps made from tantalum wire of diameter 0.50 mm. The ring has a diameter $2r_0 = 1.00$ mm, and the end cap separation is $2z_0 = 0.94$ mm. An ac voltage of frequency $\Omega = 2\pi \times 11.50$ MHz and amplitude $V_{\text{zero-peak}} = 350$ V is applied to the ring, which generates a pseudopotential well in which the ion is trapped. A dc voltage of 17.4 V is also added to the ring, which modifies the radial and axial oscillation frequencies making them approximately equal, $\omega_r \approx \omega_z = 2\pi \times 1.1$ MHz.

If the ion is moved away from the ac center of the trap by stray fields, it experiences additional micromotion at the drive frequency. This is corrected by applying a small dc voltage to one of the end caps. Using the rf-photon correlation technique [14] as a diagnostic, the intensity of the micromotion sidebands at 467 nm are inferred to be less than 0.01 than that of the main carrier. The trap parameters, along with a calculation of the minimum kinetic energy achieved by laser cooling, predict the intensity of the secular sidebands at 467 nm to be less than 0.05 that of the carrier. The secular sidebands observed on the 467 nm profile had an intensity in accordance with expectations, thus showing the ion was well confined in the Lamb-Dicke regime, eliminating the first order Doppler effect.

A single ion of Yb$^+$ is laser cooled by driving the $^2S_{1/2}$-$^2P_{1/2}$ resonance transition at 369 nm (see Fig. 1). The natural width of the cooling transition is $2\pi \times 19.6$ MHz [15,16], which makes the Doppler cooling limit 0.47 mK. From the $^2P_{1/2}$ state, there is a 0.66% branching ratio [8] for decay into the metastable $^2D_{3/2}$ level. To maintain the cooling cycle, the ion is rapidly returned to the ground state via the $^3D[3/2]_{1/2}$ level by laser radiation at 935 nm [17]. An argon-ion pumped, frequency-doubled Ti:sapphire laser is used to generate the cooling radiation at 369 nm, while an extended cavity

diode emitting at 935 nm is used to depopulate the $^2D_{3/2}$ state. During the loading of the ion, 50 μW of 369 nm and 10 mW of 935 nm light are focused at the trap center into spots with radii 100 μm. During the experiment itself, the 369 nm light is focused into a 12 μm radius spot, and the power used is reduced accordingly. Single ion fluorescence rates of up to 50 kHz are observed.

The earth's ambient magnetic field causes the individual Zeeman components of the $^2S_{1/2}$-$^2F_{7/2}$ transition to be spread over several megahertz. Careful control of the magnetic field is therefore needed to concentrate all of the Zeeman components into a small frequency region. In zero magnetic field, the fluorescence from the ion falls to zero due to optical pumping into the $m_J = \pm 3/2$ state of the $^2D_{3/2}$ level. This effect allows the magnetic field to be nulled to about ± 1.5 μT, using three orthogonal pairs of field coils. A small magnetic field of 11 μT is then applied parallel to the direction of the laser beam. This selects $\Delta m_J = \pm 1$ components of the $^2S_{1/2}$-$^2F_{7/2}$ transition. The line shape is thus made up of four components spaced at ± 110 and ± 230 kHz with relative intensities of 5 to 3. This structure is not resolved in this experiment due to the linewidth of the 467 nm laser.

Light at 467 nm is generated with an argon-ion pumped frequency-doubled Ti:sapphire laser. The frequency of

FIG. 2. The experimental apparatus. AOM, acousto-optic modulator. PBS, polarizing beam splitter.

TABLE I. Frequency measurements made at the NPL.

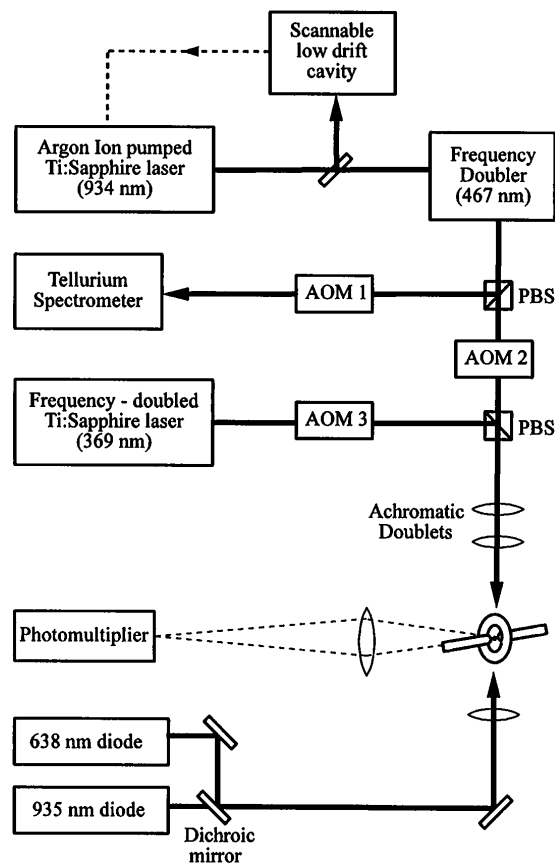| Transition	Frequency (MHz)	Reference
$^2S_{1/2}$-$^2D_{5/2}$	729 476 868.9(0.4)	[4,11]
$^2D_{5/2}$-$^2F_{7/2}$	87 360 087(4)	[12]
$^2S_{1/2}$-$^2F_{7/2}$ (expected)	642 116 781.9(4.1)	
$^2S_{1/2}$-$^2F_{7/2}$ (actual)	642 116 785.3(0.7)	[This work]

the Ti:sapphire laser is stabilized to a scannable reference cavity which reduces the laser bandwidth to 350 kHz, with a drift of 50 kHz/min. The Ti:sapphire output is frequency doubled using an angle-tuned crystal of lithium triborate. The fundamental radiation is enhanced around the crystal in a build up cavity, generating about 15 mW of 467 nm light. Doppler-free absorptions in molecular tellurium (^{130}Te$_2$) are used as local frequency references at 467 nm. Modulation transfer features are generated using the technique described by Ma *et al.* [18], using a cell characterized by Barwood *et al.* [19]. Tellurium features in the vicinity of the search region were measured by comparison with an iodine-stabilized He-Ne laser [20]. The measurements show a systematic shift of +90(1) MHz when compared with the results published by Courteille *et al.* [21]. The tellurium feature closest to the 467 nm search region was measured to be 642 116 513.6(0.6) MHz (1σ).

The 369 and 467 nm laser beams are combined with a polarizing beam splitter, and then focused to a spot with radius $w_0 = 12$ μm, using an achromatic lens system. The 369 nm laser is used as a "tracer" for the 467 nm laser to ensure overlap with the ion. The overlap of the two laser spots is better than ± 2 μm, which is tested outside the trap by focusing the beams onto a 5 μm pinhole and observing the transmitted light.

When the 467 nm radiation is in resonance with the $^2S_{1/2}$-$^2F_{7/2}$ transition, the ion is driven into the $^2F_{7/2}$ level and the resonance fluorescence ceases. The $^2F_{7/2}$ state is depopulated by driving the $^2F_{7/2}$-$^1D[5/2]_{5/2}$ transition at 638.615(1) nm [4,11] with an extended cavity diode laser. This source delivers 3 mW of light with a 2 MHz bandwidth focused in a 150 μm spot. The $^1D[5/2]_{5/2}$ state decays into either the $^2D_{3/2}$ or the $^2D_{5/2}$ level, and from there back into the cooling cycle where resonance fluorescence resumes.

Collisions with background gas can also cause a transition into the $^2F_{7/2}$ level, from the $^2D_{3/2}$ level [4,6,22]. This mechanism is indistinguishable from a stimulated off jump caused by the 467 nm laser, and so contributes a background to the experiment. The base pressure inside the trap is less than 2×10^{-8} Pa. A background off-jump rate of less than 1 per hour is observed.

An initial scan of the search region located the position of the transition. A high resolution scan of the resonance was then performed to produce a line profile. The 467 nm light is split into two portions. The strongest part, about 10 mW, is focused onto the ion, the remainder is used to observe the tellurium reference. The frequency difference between the tellurium feature and the $^2S_{1/2}$-$^2F_{7/2}$ transition is spanned with two acousto-optic modulators (AOM1 and AOM2 in Fig. 2). The 369 and 467 nm laser beams are chopped in antiphase using AOM2 and AOM3. This avoids broadening the ground state during the 467 nm laser interrogation. The cycle consists of

12 ms of 369 nm radiation followed by 30 ms of 467 nm radiation.

The initial observation of the $^2S_{1/2}$-$^2F_{7/2}$ transition was made while repeatedly scanning the 467 nm laser over the 20 MHz search region. The frequency of the laser was varied by applying a voltage ramp to the piezo-scannable reference cavity. The tellurium feature has a dispersion shape with a peak-peak width of 8 MHz. The feature was observed throughout the scan, making it possible to measure the frequency of the 467 nm laser to better than 1 MHz. During a three hour period eight off quantum jumps were observed, six of which were localized in a 1 MHz section of the search region. During this time, an average of three background jumps is probable. After each off event, fluorescence was restored by depopulating the $^2F_{7/2}$ level using the laser at 638 nm. Attempts to induce a stimulated decay from the $^2F_{7/2}$ state with 467 nm radiation were unsuccessful. This was probably due to heating of the ion by elastic collisons with the background gas, which occurs on time scales of order 10 sec. Neither collisional quenching nor molecule formation [23] were observed. This is not suprising, as these inelastic processes are many orders of magnitude less likely than elastic collisions.

Once the location of the 467 nm transition had been determined, a more careful scan of the resonance was undertaken. The laser frequency was set to the center of the tellurium feature, and one of the acousto-optic modulators (AOM1) used to scan over a 3.5 MHz region in steps of 250 kHz. The resulting line profile (Fig. 3) shows the carrier centered at 271.7 MHz from the tellurium feature. It is also possible to discern sidebands spaced at the secular frequency, with an observed intensity in accordance with expectations. Each 250 kHz bin represents a total
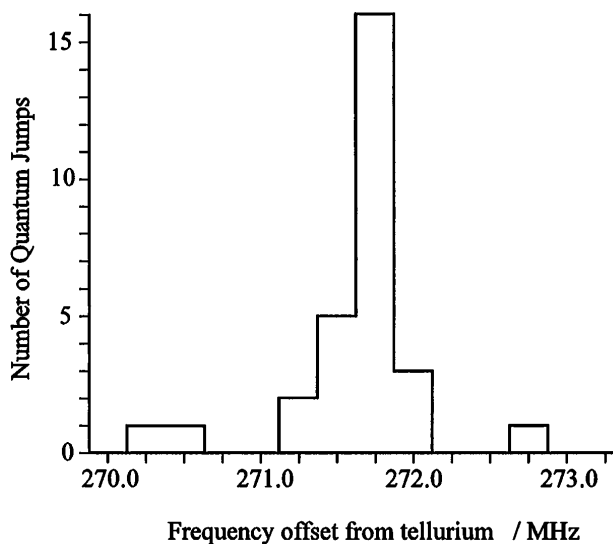

FIG. 3. Profile of the 467 nm $^2S_{1/2}$-$^2F_{7/2}$ transition. Each 250 kHz bin is interrogated for a total of 492 sec by 467 nm radiation.

of 492 sec interrogation by the 467 nm radiation. This scan took two hours to complete, in which time an average background of two jumps is probable.

From the rate at which the transition is driven, taken together with the laser parameters used, an estimate of the $^2F_{7/2}$ lifetime can be made using a simple rate equation analysis. This gives an estimated lifetime of 3700 days, which corresponds to 10.1 years. The standard deviation on this estimate is $+7/-4$ yr (1σ) due to uncertainties in the exact laser parameters used.

The extremely long lifetime of the $^2F_{7/2}$ level makes the 467 nm transition attractive as an optical frequency standard. To use this transition as a high-Q frequency reference will require the probe laser bandwidth to be reduced. Fast servoing of the laser frequency to an extremely high finesse reference cavity, should reduce the bandwidth of the 467 nm laser from 350 kHz to the 1 Hz level. This reduction in bandwidth would lead to a corresponding increase in the quantum jump rate. This would allow a relaxation in the existing power and focusing requirements. It is also important to minimize perturbations to the transition frequency. In particular, to reduce the effects of magnetic fields on the reference transition, it will be desirable to use the 171 isotope of Yb$^+$. Although more difficult to cool [5,24,25], the odd isotope has transitions which have a reduced magnetic field dependence. This will require the 467 nm transition to be located in this isotope, probably by repeating the measurements of the $^2S_{1/2}$-$^2D_{5/2}$ and $^2D_{5/2}$-$^2F_{7/2}$ transitions.

In conclusion, the 467 nm $^2S_{1/2}$-$^2F_{7/2}$ transition in ^{172}Yb$^+$ has been driven. A Doppler-free absorption feature in ^{130}Te$_2$, which has a frequency 642 116 513.6(0.6) MHz (1σ), is used as a local reference. The $^2S_{1/2}$-$^2F_{7/2}$ transition is observed to have a frequency of 642 116 785.3(0.7) MHz (1σ). The maximum jump rate and laser parameters allow an estimate of 10 years to be placed on the $^2F_{7/2}$ lifetime. This is the first time an atomic electric octupole transition has been driven. Previously, magnetic octupole emission in the x-ray region from highly charged ions has been observed [26]. This experiment demonstrates the viability of the $^2S_{1/2}$-$^2F_{7/2}$ electric octupole transition as an optical frequency reference with exceptionally high Q.

[1] H. Dehmelt, IEEE Trans. Instrum. Meas. **31**, 83 (1982).
[2] W. M. Itano, J. C. Bergquist, and D. J. Wineland, Science **237**, 612 (1987).
[3] G. Werth, Metrologia **22**, 190 (1986).
[4] P. Taylor *et al.* (to be published).
[5] D. Engelke and C. Tamm, Europhys. Lett. **33**, 347 (1996).
[6] H. Lemitz, J. Hattendorf-Ledwoch, R. Blatt, and H. Harde, Phys. Rev. Lett. **62**, 2108 (1989).
[7] V. Shevelko (private communication in [6]).
[8] B. C. Fawcett and M. Wilson, At. Data Nucl. Data Tables **47**, 241 (1991).
[9] H. Dehmelt, Bull. Am. Phys. Soc. **20**, 60 (1975).
[10] W. C. Martins, R. Zalubas, and L. Hagen, *Atomic Energy Levels—The Rare Earth Elements* (U.S. GPO, Washington, D.C., 1976).
[11] P. Gill *et al.,* Phys. Rev. A **52**, 909 (1995).
[12] A. S. Bell *et al.,* J. Mod. Opt. **39**, 381 (1992).
[13] R. H. Dicke, Phys. Rev. **89**, 472 (1953).
[14] R. Bümel, C. Kappler, W. Quint, and H. Walther, Phys. Rev. A **40**, 808 (1989).
[15] R. W. Berends, E. H. Pinnington, B. Guo, and Q. Ji, J. Phys. B **26**, 701 (1993).
[16] R. M. Lowe, P. Hannaford, and A. M. Mårtensson Pendrill, Z. Phys. D **28**, 283 (1993).
[17] A. S. Bell *et al.,* Phys. Rev. A **44**, 20 (1991).
[18] L. S. Ma *et al.,* Appl. Phys. B **57**, 159 (1992).
[19] G. P. Barwood *et al.,* Phys. Rev. A **43**, 4783 (1991).
[20] P. Taylor, P. Gill, and W. R. C. Rowley (to be published).
[21] P. Courteille, L. S. Ma, W. Neuhauser, and R. Blatt, Appl. Phys. B **59**, 187 (1994).
[22] A. Bauch, D. Schnier, and C. Tamm, J. Mod. Opt. **39**, 389 (1992).
[23] K. Sugiyama and J. Yoda, in *Proceedings of the Fifth Symposium on Frequency Standards and Metrology,* edited by J. Bergquist (World Scientific, Singapore, 1995), p. 432.
[24] V. Enders *et al.,* Europhys. Lett **24**, 325 (1993).
[25] P. Fisk, M. Lawn, and C. Coles, Appl. Phys. B **57**, 287 (1993).
[26] P. Beiersdorfer, Phys. Rev. Lett. **67**, 2272 (1991).