PHYSICAL REVIEW A, VOLUME 62, 020501(R)

Observation of the $^2S_{1/2}$-$^2F_{7/2}$ electric octupole transition in a single $^{171}Yb^+$ ion

M. Roberts, P. Taylor, G. P. Barwood, W. R. C. Rowley, and P. Gill
National Physical Laboratory, Queens Road, Teddington, Middlesex TW11 0LW, United Kingdom
(Received 7 March 2000; published 7 July 2000)

The $^2S_{1/2}(F=0)$-$^2F_{7/2}(F=3)$ electric octupole transition in $^{171}Yb^+$ has been detected by observing quantum jumps in the fluorescence of a single ion held in an electrodynamic trap. The transition frequency has been measured as 642 121 498.1 (0.8) MHz. The extreme weakness of this transition makes it ideally suited as an optical frequency reference of subhertz linewidth. The absolute frequencies of a series of Doppler-free absorptions in tellurium vapor are also reported. One of these absorptions was used as a local frequency reference while locating the electric octupole transition.

PACS number(s): 32.80.Pj, 33.20.−t, 06.30.Ft, 42.50.Lc

I. INTRODUCTION

It has been suggested that the ultimate limit to the reproducibility of an ion trap optical frequency standard could approach one part in 10^{18} [1]. For most ion trap systems currently being pursued [2], the reference transition has a natural linewidth of around 1 Hz, implying an averaging time of around one month to reach a part in 10^{18} stability [3]. Reducing the natural linewidth by a factor of 100 reduces the required averaging time 100 fold, to only a few hours. Hence it is desirable to use a reference transition with a subhertz natural linewidth.

One such transition is the electric octupole $^2S_{1/2}$-$^2F_{7/2}$ 467-nm transition in the Yb^+ system. The $^2F_{7/2}$ level in Yb^+ has a lifetime of around 10 yr [4], giving the $^2S_{1/2}$-$^2F_{7/2}$ electric octupole transition a natural linewidth in the nHz region. The removal of the natural linewidth barrier to the standard's stability means that in practice the stability will most likely be limited by the reference laser linewidth. The current state-of-the-art laser linewidth is 0.16 Hz [5], and it should be possible (though difficult) to build a laser with a linewidth of 0.1 Hz or less. The electric octupole transition in Yb^+ therefore offers the potential to make measurements at the part in 10^{18} level within a reasonable averaging time.

The electric octupole transition has been observed in the 172 isotope of ytterbium [4]. However, to construct an optical frequency standard of the highest reproducibility, it is desirable to use the 171 isotope, as this has $m_F=0-m_F=0$ transitions that are free from the linear Zeeman effect. The hyperfine structure of this isotope makes laser cooling and state preparation more difficult, which is the reason for originally working in the 172 isotope. This switch in isotope has made it necessary to relocate the $^2S_{1/2}$-$^2F_{7/2}$ 467-nm transition.

Given the extreme weakness of the octupole transition, it is necessary to have a good knowledge of its spectral position prior to an attempt to observe it. The isotope shift and hyperfine splitting calculations in this system are not of sufficient accuracy to enable a reliable prediction of the transition frequency. It has therefore proved necessary to measure the $^2S_{1/2}(F=0)$-$^2D_{5/2}(F=2)$ 411-nm [6] and $^2F_{7/2}(F=3)$-$^2D_{5/2}(F=2)$ 3.43-μm [7] transitions in the $^{171}Yb^+$ isotope (see Fig. 1 and Table I). The electric octupole frequency is the difference in these two values.

This Rapid Communication reports an observation of the electric octupole transition in the $^{171}Yb^+$ isotope. A single $^{171}Yb^+$ ion is monitored by repeatedly driving the $^2S_{1/2}$-$^2P_{1/2}$ cooling transition, and observing resonance fluorescence. The fluorescence is interrupted when the ion is driven into the $^2F_{7/2}$ state by laser light at 467 nm. This quantum jump signal is used to build up a transition profile as the 467-nm laser frequency is stepped over the transition frequency. Due to the long lifetime of the $^2F_{7/2}$ state, the ion is returned to the cooling cycle by driving a subsidiary transition at 638 nm rather than waiting for spontaneous decay. The quantum jump profile allowed for a direct measurement of the octupole transition frequency.

II. EXPERIMENTAL ARRANGEMENT

This work was performed in a variation of the electrodynamic (Paul) trap, known as an endcap trap, which was de-

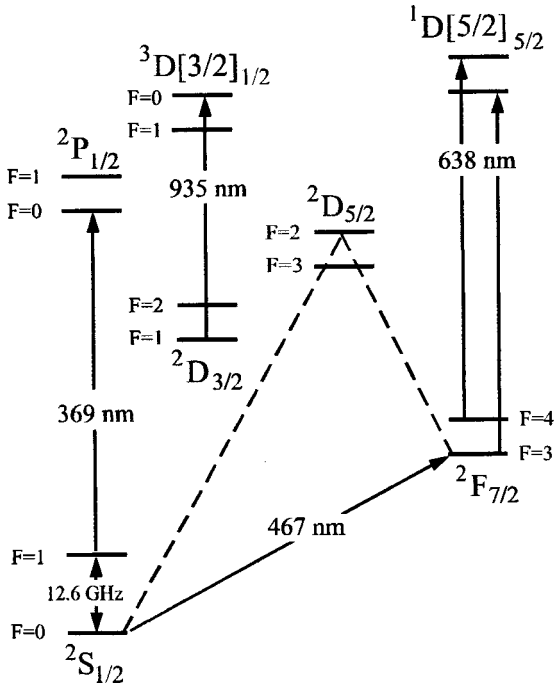

FIG. 1. Partial term scheme of $^{171}Yb^+$. The 369-nm, 935-nm, and 12.6-GHz transitions are used for laser cooling. The 638-nm transition is used to return the ion to the cooling cycle, once the 467-nm transition has been driven.

TABLE I. Frequency measurements of $^{171}Yb^{+}$ transitions.

Transition	Frequency (MHz)
$^2S_{1/2}(F=0)$-$^2D_{5/2}(F=2)$	729 487 779.566(0.153)
$^2F_{7/2}(F=3)$-$^2D_{5/2}(F=2)$	87 366 282.258(0.091)
$^2S_{1/2}(F=0)$-$^2F_{7/2}(F=3)$ (predicted)	642 121 497.308(0.178)
$^2S_{1/2}(F=0)$-$^2F_{7/2}(F=3)$ (observed)	642 121 498.1(0.8)

veloped by Schrama *et al.* [8]. The National Physical Laboratory (NPL) trap has the same electrode geometry as described in Ref. [8], with an endcap separation of 0.56 ± 0.02 mm. The trap is driven by an ac voltage (zero peak) of 290 V at a frequency of 12.8 MHz, and has a measured efficiency of 0.63 ± 0.03. In this experiment, an additional dc voltage of -14.8 V is also applied to the endcaps to equalize the radial and axial secular frequencies to a frequency of approximately 1.3 MHz. Single-ion confinement times of up to a month have been observed.

The levels involved in the laser-cooling scheme are shown in Fig. 1, and a schematic layout of the experiment is shown in Fig. 2. Details of the laser systems have been discussed previously [6]. The single ion is laser cooled by repeatedly driving the $F=1-F=0$ component of the $^2S_{1/2}$-$^2P_{1/2}$ transition with laser radiation at 369 nm. The resonance fluorescence from this transition is used to monitor the state of the ion. From the $^2P_{1/2}(F=0)$ state there is a small probability for decay into the metastable $^2D_{3/2}(F=1)$ state. To maintain the cooling cycle, the $^2D_{3/2}$ level is rapidly depopulated by a laser at 935 nm, returning the ion to the $F=1$ ground state via the $^3D[3/2]_{1/2}(F=0)$ level. This

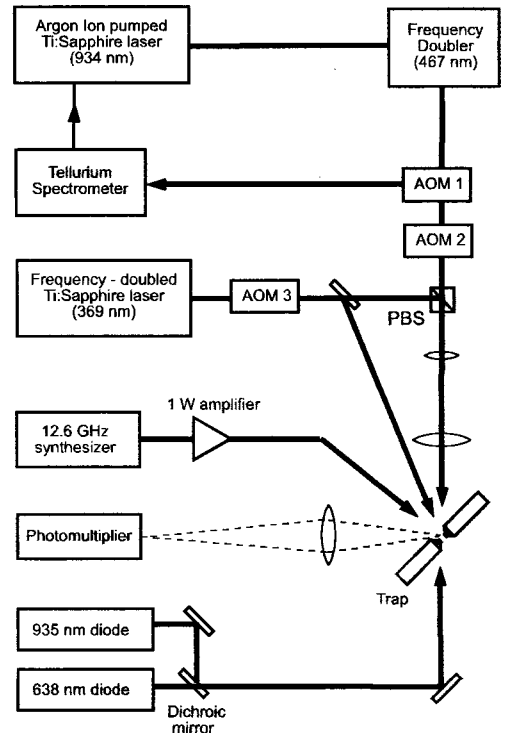

FIG. 2. Schematic layout for the experiment. AOM, acousto-optic modulator; PBS, polarizing beam splitter.

cooling cycle forms a closed loop, but nonresonant excitation of the $^2P_{1/2}(F=1)$ state by the cooling laser leads to optical pumping into the $F=0$ ground state. Therefore to achieve effective cooling, the $F=0$ ground state must be coupled to the cooling cycle. In this work, this is done by driving the $F=0-F=1$ ground-state transition with microwave radiation at 12.6 GHz. Any off-resonant population of the $^2D_{3/2}(F=2)$ state is off-resonantly depopulated by the intense 935-nm source.

To build up a quantum jump profile, a number of frequency scans over the transition frequency are performed. For each scan, the probe-laser frequency is stepped over the relevant region and several interrogation cycles are performed at each point. The interrogation cycle is as follows: At each step the ion is cooled and detected with radiation at 369 nm for 200 ms. Next, the ion is prepared in the $F=0$ ground state by switching the microwave radiation off (60 ms); this allows the $F=0$ state to become off-resonantly populated by decay from the $^2P_{1/2}(F=1)$ level [9]. The cooling radiation is then also switched off, to prevent broadening of the ground state. Finally, a 360-ms pulse of 467-nm radiation interrogates the ion.

In the event of the $^2S_{1/2}$-$^2F_{7/2}$ transition being driven, the fluorescence from the ion ceases. The 467-nm laser frequency is noted and the $^2F_{7/2}$ state is depopulated by driving the $^2F_{7/2}$-$^1D[5/2]_{5/2}$ transition at 638 nm [6]. From the upper state of the 638-nm transition, the ion returns to the cooling cycle via either the $^2D_{3/2}$ or $^2D_{5/2}$ levels. A number of 467-nm scans are performed to build up a profile, as typically only every other scan yields a quantum jump, due to the extreme weakness of the octupole transition.

Collisions with background gas can also cause a transition into the $^2F_{7/2}$ level from the $^2D_{3/2}$ level. This mechanism causes a laser frequency independent background to the experiment. The rate of pressure-induced jumps depends on the base pressure in the trap, which in this case is less than 2×10^{-8} Pa. The background jump rate is less than 0.3 h.

III. THE PROBE-LASER SYSTEM

Light at 467 nm is generated with a frequency-doubled Ti:sapphire laser, which has been described previously [4]. The frequency of the 467-nm output is controlled by stabilizing it to Doppler-free absorptions in tellurium vapor. The frequency offset between the tellurium absorption frequency and the electric octupole frequency is bridged with a double-passed acousto-optic modulator (AOM1) driven at around 250 MHz. The frequency shifted beam is used to observe the tellurium features with 1 mW of light, and the unshifted beam is sent to the trap where 2.5 mW is used for the experiment. A second modulator (AOM2) is used to chop the 467-nm beam, which gives a fixed 60-MHz shift to the probe-laser frequency.

Molecular $^{130}Te_2$ vapor at around 500 °C exhibits a series of absorptions that are used as a local frequency reference. The Doppler-free features are obtained using the modulation transfer technique, with a setup that closely follows that of Ma and co-workers [10,11]. The Opthos $^{130}Te_2$ cell is 75 mm long, 25 mm in diameter, and has a 50-mm sidearm. The

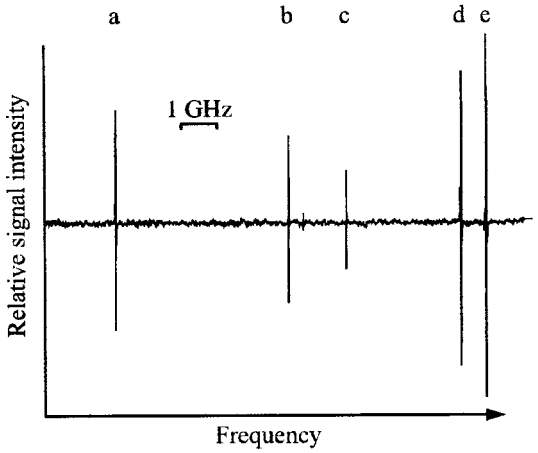

FIG. 3. Doppler-free molecular absorptions in tellurium vapor in the vicinity of 467 nm. Transition ''b'' is used as a frequency reference when locating the octupole transition in $^{171}Yb^+$.

body of the cell is heated in an oven to 500 °C and the sidearm is controlled at 470 °C to within ±2 °C. A spectra of the tellurium features over the frequency region of interest is shown in Fig. 3. The absolute frequencies for the features marked a–e in Fig. 3 are given in Table II. These were measured with 0.6-MHz $(1-\sigma)$ uncertainty by comparison with a 633-nm iodine-stabilized He-Ne laser, via the NPL 1-m etalon wavelength comparator [12].

The intensities of the absorptions observed in this work differ from those of Refs. [10,11]. In addition, the absolute frequencies of the features given in Table II are consistently 90(1) MHz higher than those reported in Ref. [11]. There are a number of reasons to believe that the frequencies reported in Table II are the correct values: the NPL wavelength comparator is a well established device and is capable of accuracies of one part in 10^{10}; the tellurium frequencies were also measured independently with a wave meter of 10-MHz accuracy, giving values consistent with those in Table II; and the predicted and observed measurements of the octupole frequency in this work, and previously [4], agree within the quoted uncertainties, which is the most critical test of the measurement accuracy.

The $^2S_{1/2}$-$^2F_{7/2}$ electric octupole transition was expected to lie 501.9 (0.6) MHz below the frequency of the tellurium absorption marked ''b'' in Fig. 3 and Table II. This frequency region could be scanned in a repeatable manner by

TABLE II. Frequencies of tellurium absorptions. Feature ''b'' was used as a frequency reference to locate the $^2S_{1/2}$-$^2F_{7/2}$ transition in $^{171}Yb^+$.

Absorption	Frequency (MHz)
a	642 116 513.6 (0.6)
b	642 121 999.2 (0.6)
c	642 123 750.1 (0.6)
d	642 127 041.9 (0.6)
e	642 127 720.2 (0.6)

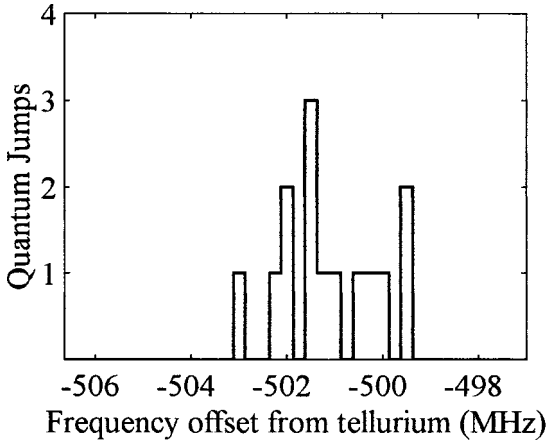

FIG. 4. Quantum jump profile of the electric octupole $^2S_{1/2}(F=0)$-$^2F_{7/2}(F=3,m_F=0)$ transition in $^{171}Yb^+$.

locking the double-passed output of AOM1 to the tellurium absorption and scanning the acoustic modulation frequency. A rather poor signal-to-noise ratio of the tellurium feature limited the laser linewidth to around 1 MHz.

To drive the electric octupole transition on a reasonable time scale requires a high 467-nm intensity. The 467 and 369-nm beams are overlapped via a polarizing beam splitter and focused to a tight spot ($w_0 \approx 7\ \mu$m) at the trap center with an achromatic lens system. This allows the cooling laser to be used as an alignment beam, ensuring the 467-nm light is overlapped with the ion. The lateral overlap between the two beams is tested outside the vacuum chamber with a 5-μm diameter pinhole showing the foci to be overlapped to better than 2μm. The focal planes of the two beams were shown to coincide to within one fifth of a Rayleigh range using a knife-edge test. To ease the location of fluorescence each day, a second 369-nm laser beam path was available with a larger focus of $w_0 \sim 50\ \mu$m. This beam was also used in the main experiment to cool the ion during the cooling pulses. The tightly focused 369-nm laser beam was blocked at these times.

IV. OBSERVATION OF THE OCTUPOLE TRANSITION

The initial observation of the $^2S_{1/2}$-$^2F_{7/2}$ transition was performed in a small magnetic field (<10 μT). This reduced the octupole transition Zeeman structure to within a MHz or so. A 10-MHz scan was performed around the frequency region of interest and a small number of jumps were observed near the center of that region.

Once an approximate location of the transition had been found, a high field of 500 μT was applied to split the Zeeman structure. The direction of this field was chosen to preferentially select the $m_F=0-m_F=0$ transition (60% of the total transition probability). Figure 4 shows the resulting quantum jump profile of the $^2S_{1/2}(F=0)$-$^2F_{7/2}(F=3,m_F=0)$ transition. The $\Delta m=1$, 2, and 3 Zeeman components are separated by 10, 20, and 30 MHz at this magnetic field, and hence are not taken in on this scan. The profile is built up from 40 separate scans, each consisting of 40 frequency steps of 250 kHz. At each step there are 15 probe interroga-

tions of 320 ms, giving a total interrogation time for the duration of the experiment of 7680 s. The profile in Fig. 4 consists of a carrier and two secular sidebands spaced at 1.3 MHz. The sidebands are unresolved in this picture due to the relatively large linewidth of the 467-nm laser.

The center frequency of the transition is obtained from the mean of the data of Fig. 4 (-501.125 MHz), and the absorption frequency of the tellurium component ''b'' (see Table II). This gives a center frequency of 642 121 498.1 MHz. There are four components to the error of this measurement: the uncertainty in the tellurium absorption frequency (0.6 MHz), the standard error of the mean of the data in Fig. 4 (0.4 MHz), possible asymmetry in the observed data (0.1 MHz), and an uncertainty due to the second-order Zeeman shift (0.2 MHz). Using simple perturbation theory, the second-order Zeeman shift is estimated to be 75 kHz for a magnetic field of 500 μT, which is in agreement with previous measurements of this effect on the $^2S_{1/2}$-$^2D_{5/2}$ transition [6]. However, as the shift has not actually been measured for the octupole transition, a conservative uncertainty of 0.2 MHz is included in the error budget for this effect. At the Doppler cooling limit, the sidebands are expected to be small (10% of the carrier) and approximately equal in intensity [4]. Hence the sidebands will not cause a significant asymmetry in the observed line profile. As a conservative estimate, an asymmetry of 10% of the standard deviation of the data is taken, giving an asymmetry contribution to the error bar of 0.1 MHz. The $^2S_{1/2}(F=0)$-$^2F_{7/2}(F=3,\ m_F=0)$ transition therefore has a frequency of 642 121 498.1 (0.8) MHz. This is within one standard deviation of the indirect measurement of the transition frequency (see Table I).

The laser power, spot size, laser linewidth, and jump rate near line center allow an estimate of the $^2F_{7/2}$ state lifetime to be made. This yields a result of 1.7×10^8 s or 5.4 yr. The large standard uncertainty on this measurement of $+9.3/-3.6$ yr is due to the uncertainties in the exact parameters used, in particular the poor statistics of the line center jump rate. This value is consistent with the 10 $(+7, -4)$ yr lifetime obtained for the $^2F_{7/2}$ state in $^{172}Yb^+$ [4]. The previous measurement has a slightly smaller percentage error, due primarily to the better statistics of that data.

V. CONCLUSION

The $^2S_{1/2}(F=0)$-$^2F_{7/2}(F=3)$ electric octupole transition has been observed in a single ion of $^{171}Yb^+$. The transition frequency has been directly measured as 642 121 498.1 (0.8) MHz. This is within 1-σ of the indirect measurement of 642 121 497.308 (0.176) MHz that was used to enable the location of the octupole transition. A Doppler-free absorption in molecular tellurium was used as a local frequency reference for this measurement. The frequencies of a number of such absorptions have been reported, each with a frequency uncertainty of 0.6 MHz.

From the laser power, spot size, laser linewidth, and jump rate at line center it has been possible to deduce the lifetime for the $^2F_{7/2}$ level to be 5.4 yr $(+9.3/-3.6$ yr). This is consistent with the previous measurement [4] of the $^2F_{7/2}$ lifetime of 10 $(+7, -4)$ yr.

The inconveniently low quantum jump rate of this experiment will increase linearly as the laser linewidth is decreased. For example, with a 1-kHz linewidth laser tuned to line center, a jump rate of over 10 per second would be achieved for the current laser power and spot size. With a smaller laser linewidth, it will also be possible to resolve the Zeeman structure of the octupole transition at a much lower magnetic field. This will drastically reduce the systematic shift to the transition frequency caused by the second-order Zeeman effect.

This is only the second time that an optical electric octupole transition has been observed in an atomic system, the first being in the $^{172}Yb^+$ isotope. This experiment paves the way for using the electric octupole transition in $^{171}Yb^+$ as an optical frequency reference with unprecedented stability and reproducibility.

[1] H. Dehmelt, IEEE Trans. Instrum. Meas. **31**, 83 (1982).

[2] A. Madej and J. Bernard, in *Frequency Measurement and Control, Topics in Applied Physics*, edited by A. Luiten (Springer Verlag, Berlin, 2000).

[3] D. J. Wineland, J. C. Bergquist, D. Berkeland, J. J. Bollinger, F. C. Cruz, W. M. Itano, B. M. Jelenkovic, B. E. King, D. M. Meekhof, J. D. Miller, C. Monroe, M. Rauner, and J. N. Tan, in *Proceedings of the 5th Symposium on Frequency Standards and Metrology*, edited by J. C. Bergquist (World Scientific, Singapore, 1996), pp. 11–19.

[4] M. Roberts, P. Taylor, G. P. Barwood, P. Gill, H. A. Klein, and W. R. C. Rowley, Phys. Rev. Lett. **78**, 1876 (1997).

[5] B. Young, R. Rafac, F. Cruz, W. Itano, D. Wineland, and J. Bergquist, in *Proceedings of the XIV International Conference on Laser Spectroscopy*, edited by R. Blatt (World Scientific, Singapore, 1999), pp. 61–70.

[6] M. Roberts, P. Taylor, S. V. Gateva-Kostova, R. B. M. Clarke, W. R. C. Rowley, and P. Gill, Phys. Rev. A **60**, 2867 (1999).

[7] P. Taylor, M. Roberts, G. M. Macfarlane, G. P. Barwood, W. R. C. Rowley, and P. Gill, Phys. Rev. A **60**, 2829 (1999).

[8] C. A. Schrama, E. Peik, W. W. Smith, and H. Walther, Opt. Commun. **101**, 32 (1993).

[9] D. Engelke and C. Tamm, Europhys. Lett. **33**, 347 (1996).

[10] L. S. Ma, P. Courteille, G. Ritter, W. Neuhauser, and R. Blatt, Appl. Phys. B: Photophys. Laser Chem. **57**, 159 (1992).

[11] P. Courteille, L. S. Ma, W. Neuhauser, and R. Blatt, Appl. Phys. B: Lasers Opt. **59**, 187 (1994).

[12] G. P. Barwood, W. R. C. Rowley, P. Gill, J. L. Flowers, and B. W. Petley, Phys. Rev. A **43**, 4783 (1991).