

## Measurement of the ${}^2S_{1/2}$ - ${}^2D_{5/2}$ 411-nm interval in laser-cooled trapped ${}^{172}\text{Yb}^+$ ions

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The  $4f^{14}6s\ {}^2S_{1/2}$ - $4f^{14}5d\ {}^2D_{5/2}$  411-nm transition in  ${}^{172}\text{Yb}^+$  has been measured to be 729 476 868(11) MHz. The frequency was determined by probing a cold cloud of a few  ${}^{172}\text{Yb}^+$  ions held in a radio-frequency trap. The frequency measurement is part of a program to locate the  $4f^{14}6s\ {}^2S_{1/2}$ - $4f^{13}6s^2\ {}^2F_{7/2}$  467-nm ultranarrow transition in laser-cooled  $\text{Yb}^+$ . The  $4f^{13}6s^2\ {}^2F_{7/2}$ - $4f^{13}5d6s\ {}^1D[K=5/2]_{J=5/2}$  transition at 638 nm has been driven in order to depopulate the  $4f^{13}6s^2\ {}^2F_{7/2}$  state.

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### I. INTRODUCTION

Laser-cooled trapped ions offer great potential as future optical frequency standards [1]. Laser cooling reduces Doppler effects, and confinement of the ions allows long interaction times, thereby eliminating transit time broadening. The  $\text{Yb}^+$  ion offers potential standards in the visible, infrared, and microwave regions [2-8].  $\text{Yb}^+$  has a  ${}^2S_{1/2}$ - ${}^2P_{1/2}$  resonance transition at 369 nm [9], which is used for laser cooling (see Fig. 1). An ion excited into the  ${}^2P_{1/2}$  state can decay back to either the ground state or into the metastable  ${}^2D_{3/2}$  state with a branching ratio of 0.48% [10]. It is therefore necessary to clear out the  ${}^2D_{3/2}$  state, so that the cooling cycle can be maintained. This is achieved with radiation at 935 nm [11]. Temperatures of  $\sim 100$  mK have been reached.

The  $4f^{13}6s^2\ {}^2F_{7/2}$  metastable state in  $\text{Yb}^+$  has a calculated lifetime of 1533 days [12] and an observed lifetime in excess of 8 days [13]. The  $4f^{14}6s\ {}^2S_{1/2}$ - $4f^{13}6s^2\ {}^2F_{7/2}$  electric octopole transition at 467 nm therefore should have an ultranarrow natural linewidth ( $\ll 1$  Hz). Observation of this transition could lead to an optical frequency reference of extremely high  $Q$ . Estimates suggest that with a cw laser of 10 mW in a 1-kHz bandwidth, it would take a few seconds to drive the transition. Up until the present time, the uncertainty in the  ${}^2S_{1/2}$ - ${}^2F_{7/2}$  frequency has been 1.3 GHz [14], and this is too great for a search for the transition to be made with a realistic chance of success. In an attempt to reduce the uncertainty of the transition frequency, the National Physics Laboratory has undertaken a program to measure the  ${}^2F_{7/2}$ - ${}^2D_{5/2}$  3.43- $\mu\text{m}$  [15] and the  ${}^2S_{1/2}$ - ${}^2D_{5/2}$  411-nm transitions. Both the 3.43- $\mu\text{m}$  and the 411-nm transitions have themselves been proposed as potential frequency standards [2,6].

This paper reports a direct measurement of the 411-nm transition using a cloud of a few laser-cooled  ${}^{172}\text{Yb}^+$  ions. The fluorescence from these ions was monitored as the 411-nm laser frequency was scanned over the resonance. Ions driven into the  ${}^2D_{5/2}$  state by the 411-nm radiation can

decay into the long-lived  ${}^2F_{7/2}$  state and become trapped, resulting in a decay of the fluorescence signal. The 411-nm transition line shape is extracted from this decay profile.

### II. EXPERIMENTAL ARRANGEMENT

Figure 2 shows a schematic diagram of the experiment. The rf trap ( $r_0 = \sqrt{2}z_0 = 5.0$  mm) and its operation are described elsewhere [9]. The trap was loaded with small numbers of  ${}^{172}\text{Yb}^+$  ions under an ultrahigh vacuum of  $10^{-8}$  Pa. The cooling radiation at 369 nm was generated using LD 700 dye in a Coherent 699-21 ring laser with an intracavity lithium iodate doubling crystal. Approximately 150  $\mu\text{W}$  of this uv radiation was focused to a 150- $\mu\text{m}$  waist at the trap center, in order to drive the  $\text{Yb}^+$   $4f^{14}6s\ {}^2S_{1/2}$ - $4f^{14}6p\ {}^2P_{1/2}$  resonance transition. The resultant fluorescence

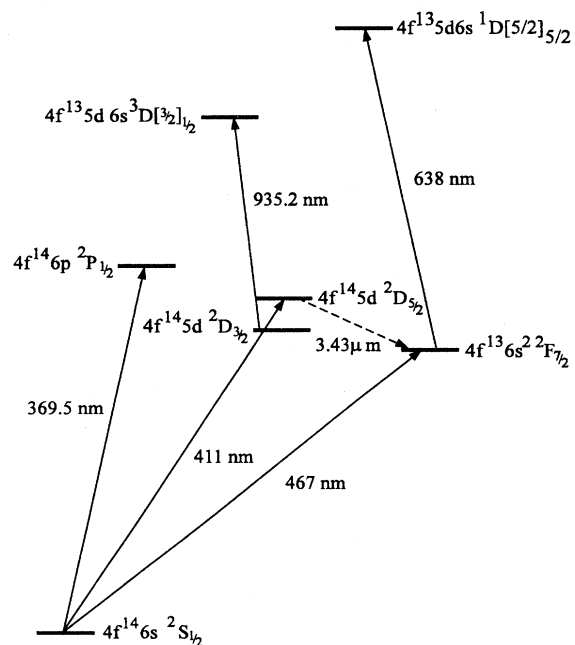


FIG. 1. Partial term scheme of  ${}^{172}\text{Yb}^+$ . The ion is cooled by the absorption and reemission of 369-nm radiation.

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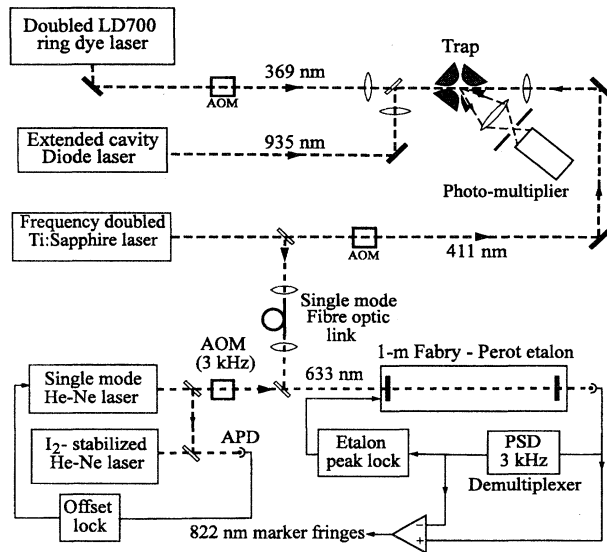


FIG. 2. Experimental arrangement for the frequency measurement of the  $^2S_{1/2}$ - $^2D_{5/2}$  interval in  $^{172}\text{Yb}^+$ . AOM, acousto-optic modulator; APD, avalanche photodiode; PSD, phase-sensitive detector.

from the ions was focused through a  $400\text{-}\mu\text{m}$  pinhole onto a solar blind photomultiplier.

Single-frequency radiation at 935 nm was generated by an SDL-6310-C diode in an extended cavity arrangement [16]. Approximately 12 mW of this light was focused into a  $150\text{-}\mu\text{m}$  spot so as to drive the  $4f^{14}5d^2D_{3/2}$ - $4f^{13}5d6s^3D[3/2]_{1/2}$  transition. This prevents ions from becoming trapped in the metastable  $D_{3/2}$  level.

Light at 411 nm was generated by frequency doubling the output from a modified Schwarz ring Ti:sapphire laser. The laser was modified with the addition of a pair of Brewster plates and a single solid intracavity étalon. The étalon's transmission was locked to the laser frequency by piezodithering it at 6 kHz and using phase-sensitive detection [17]. The laser was stabilized to an external piezotunable reference cavity of finesse 30. The power of the fundamental light was resonantly enhanced in a separate ring cavity around an angle-tuned Brewster-cut lithium triborate crystal. The enhancement cavity was kept in resonance with the Ti:sapphire output by the FM sideband reflection technique [18]. The generated 411-nm radiation was attenuated and propagated through the trap in the opposite direction to the cooling radiations. Approximately  $100\text{ }\mu\text{W}$  of 411-nm radiation focused into a  $150\text{-}\mu\text{m}$  spot was used to drive the transition. The frequency of the 411-nm radiation was monitored by sending a portion of the fundamental radiation (at 822 nm) into an evacuated 1-m Fabry-Pérot étalon [19], frequency-locked to the transmission fringe of a tunable 633-nm He-Ne laser, which was itself offset-locked to an NPL 633-nm iodine-stabilized He-Ne laser via a crystal oscillator. The latter He-Ne laser was stabilized to a saturated component of known frequency [20] of the  $B-X$  11-5  $R(127)$  transition in  $^{127}\text{I}_2$ . The étalon transmission fringes for both 633 and 822 nm were monitored by a single detector and separated by acousto-optic amplitude modulation of the 633-nm light at 3

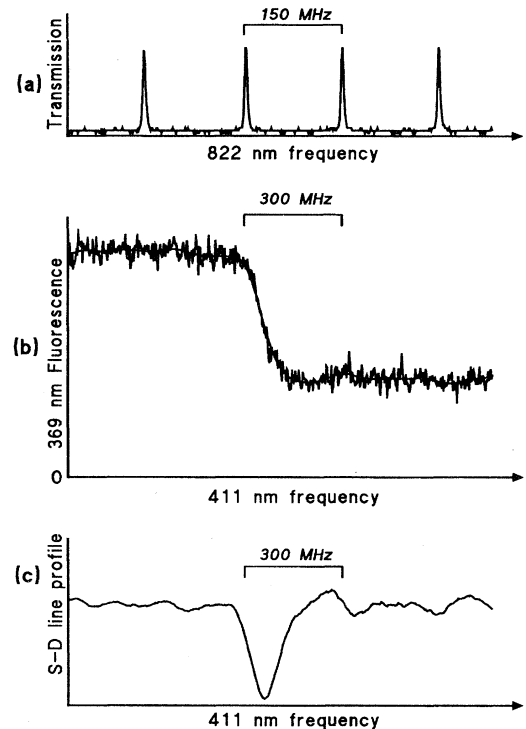


FIG. 3. A set of data for a typical 411-nm decay. (a) 1-m étalon fringes at 822 nm; (b) 369-nm fluorescence decay and smoothed data; (c) 411-nm line profile extracted from (b).

kHz, with subsequent demultiplexing by means of a subsidiary phase-sensitive detector. The 822-nm 1-m étalon fringes and the detected fluorescence from the ions were recorded simultaneously on a personal computer, which was also used to control the scan of the 822-nm laser. To assign the 822-nm 1-m étalon order number unambiguously, the Ti:sapphire laser frequency was measured using a fringe-fractioning wavemeter with an accuracy of 10 MHz (standard deviation). The 369- and 411-nm radiations were chopped in antiphase by the use of acousto-optic modulators, the 411-nm radiation being on for 5-ms pulses spaced by 100 ms, the 369-nm radiation being off at these times. This was in order to avoid any shift of the  $^2S_{1/2}$ - $^2D_{5/2}$  transition frequency due to the ac Stark effect, resulting from the 369-nm cooling radiation, which was coupled to the 411-nm transition via the ground state.

### III. MEASUREMENT OF THE 411-nm TRANSITION

The Ti:sapphire laser was scanned over the 411-nm line, and the 369-nm fluorescence level and a set of five marker fringes from the 1-m Fabry-Pérot étalon were recorded (see Fig. 3). When driven into the  $^2D_{5/2}$  state, the ions can decay back to the ground state or become trapped in the  $^2F_{7/2}$  state indefinitely, whereby a decay in fluorescence is observed. The number of ions in the ground state is directly proportional to the fluorescence level. When the laser is scanned over the 411-nm transition from a frequency  $\nu$  to  $\nu + \delta\nu$ , the fluorescence level  $I$  will change by an amount  $\delta I$ , given by

$\delta I \propto -I g(\nu) \delta \nu$ , where  $g(\nu)$  is the 411-nm line shape; therefore

$$g(\nu) = \frac{k}{I} \frac{\partial I}{\partial \nu},$$

where  $k$  is a constant.

The scan rate was set as slow as possible (approximately 15 MHz/s) to collect sufficient counts to achieve a good signal-to-noise ratio. However, the scan rate was fast enough so that the experimental conditions did not change significantly during the scan; in particular, the frequencies of the two cooling lasers. Scans were taken in both frequency directions. The power level of the 411-nm radiation was set such that the fluorescence fell to approximately 50% of its original value for each scan. This gave a sufficient signal-to-noise ratio on the data, and allowed a series of decays to be taken from each cloud of ions.

#### IV. RESULTS, ANALYSIS, AND DISCUSSION

In order to extract a 411-nm line profile for each decay, the fluorescence data were first smoothed using a moving window least-squares smoothing routine, and then differentiated. The ensuing profile was then weighted for the intensity of the light. The derived profile from the decay in Fig. 3(b) is shown in Fig. 3(c). The center frequency of the 411-nm profile was measured relative to the adjacent 822-nm 150-MHz marker fringe [Fig. 3(a)]. A total of 50 411-nm decays were observed on a variety of clouds containing different numbers of ions. The cooled cloud 411-nm linewidths (measured from the derived profile) varied from  $\sim 100$  to 400 MHz, dependent on the number of ions in the cloud, since large clouds cannot be cooled as effectively as small ones. Corresponding cloud temperatures were  $\sim 1.6$  and 25 K, respectively. For scans on the colder clouds (those with linewidths less than 200 MHz) there was a good signal-to-noise ratio, and a narrow profile could be derived. For the less cold clouds, asymmetry in the line shape was more noticeable, and there was an increased scatter in the data points. Hence, our result is taken from the 24 scans on the colder clouds. When all the data are included, the mean is shifted by less than 3 MHz. There was a difference of 11.4 MHz in the means of the forward and reverse scans. This is thought to be due to asymmetry in the 411-nm line shape, which may be caused by the heating of the cloud during the scan, as ions enter the  $F$  state and leave the cooling cycle. This would account for a shift of the line center in the direction of the scan.

The transition frequency was obtained from the mean frequency difference between the center of the decay profile and the 411-nm frequency corresponding to the adjacent 822-nm 1-m étalon fringe. This mean was determined using the 24 colder data scans, and corrected for the 60-MHz frequency downshift due to the acousto-optic modulator in the 411-nm beam. This gave the result of 729 476 868 MHz with a standard deviation of 11 MHz under our experimental conditions. Using this result, together with the measurement of

the 3.43- $\mu\text{m}$  transition of 87 360 087(4) MHz, we can predict that the 467-nm transition frequency will be 642 116 781(12) MHz. A second experiment was carried out with neither the 369-nm nor the 411-nm light being chopped, to determine any light shift. From the nine pieces of data taken, a shift of only 5 MHz was observed. As the standard deviation for these data was 20 MHz, this indicated the absence of any light shift within the uncertainty of the measurement.

The measurement of the 411-nm transition in this work employed a signal differentiation technique. This was used to generate a line profile, as there was no method available at the time of depopulating the long-lived  ${}^2F_{7/2}$  state, where the ions remain indefinitely under our trap conditions. Subsequent to this measurement, the transition at 638 nm has been driven. This excites ions trapped in the long-lived  $4f^{13}6s^2 {}^2F_{7/2}$  state into the high-lying  $4f^{13}5d6s {}^1D[5/2]_{5/2}$  state [21] from which they decay back to the ground state (see Fig. 1). Single-frequency radiation at 638 nm was generated by a Philips CQL-840/D diode in an extended cavity arrangement. Approximately 0.3 mW of light focused into a 250- $\mu\text{m}$  spot was used to drive the transition. The regeneration of fluorescence has been seen for large clouds and also for a few ions. Quantum jumps have been observed in a few-ion system by irradiating the trap with 411- and 638-nm light simultaneously. Using this laser at 638 nm, a single-ion quantum jump version of this experiment could be performed. This should lead to a smaller uncertainty in the 411-nm transition frequency, as the cold single-ion linewidth should be considerably narrower than that of this measurement.

#### V. CONCLUSION

This paper has described a direct measurement of the  ${}^{172}\text{Yb}^+ 4f^{14}6s {}^2S_{1/2}$ - $4f^{14}5d {}^2D_{5/2}$  transition at 411 nm. Small clouds of ions were driven into the  $4f^{13}6s^2 {}^2F_{7/2}$  state and resulting fluorescence decay profile recorded. The frequency of this transition was found to be 729 476 868 MHz, with a one standard deviation of 11 MHz.

This measurement could be improved by the use of 638-nm diode radiation to empty the  ${}^2F_{7/2}$  state, thereby allowing the measurement of a 411-nm profile constructed from single-ion quantum jump data. A 638-nm diode has already been used to empty the  $F$  state.

Taking the measurement of the 411-nm transition together with the previously measured frequency of the 3.43- $\mu\text{m}$  transition, an indirect measurement of the 467-nm transition has been achieved, yielding a transition frequency of 642 116 781(12) MHz. This result now makes it feasible to attempt a search for the ultranarrow 467-nm transition, which may offer an unrivaled high  $Q$  optical reference.

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