Observation and Absolute Frequency Measurements of the ${}^1S_0$-${}^3P_0$ Optical Clock Transition in Neutral Ytterbium

C. W. Hoyt,[*] Z. W. Barber,[†] C. W. Oates, T. M. Fortier,[‡] S. A. Diddams, and L. Hollberg

National Institute of Standards and Technology, 325 Broadway, Boulder, Colorado 80305, USA
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We report the direct excitation of the highly forbidden $(6s^2)^1S_0 \leftrightarrow (6s6p)^3P_0$ optical transition in two odd isotopes of neutral ytterbium. As the excitation laser frequency is scanned, absorption is detected by monitoring the depletion from an atomic cloud at $\sim 70\ \mu$K in a magneto-optical trap. The measured frequency in ^{171}Yb (F = 1/2) is 518 295 836 591.6 ± 4.4 kHz. The measured frequency in ^{173}Yb (F = 5/2) is 518 294 576 847.6 ± 4.4 kHz. Measurements are made with a femtosecond-laser frequency comb calibrated by the National Institute of Standards and Technology cesium fountain clock and represent nearly a 10^6-fold reduction in uncertainty. The natural linewidth of these $J = 0$ to $J = 0$ transitions is calculated to be ~ 10 mHz, making them well suited to support a new generation of optical atomic clocks based on confinement in an optical lattice.

Work is underway to realize a high-performance optical clock that combines the best features of state-of-the-art single-ion and neutral atom optical frequency standards [1]. Such a clock system comprises a dipole-force optical lattice trap that confines an ensemble of neutral atoms individually to subwavelength sites for Doppler- and recoil-free precision spectroscopy [2,3]. Crucial to this scheme is an atom with a ''clock'' transition that has both a narrow linewidth and an insensitivity to lattice perturbations. Several groups have recognized ytterbium as an excellent candidate and are pursuing lattice-based optical clocks that will use its narrow ${}^1S_0 \leftrightarrow {}^3P_0$ transition [3–6]. However, until now the absolute frequency of this resonance was known in tables [7] to only a few gigahertz. We report the direct excitation of the doubly forbidden $(6s^2)^1S_0 \leftrightarrow (6s6p)^3P_0$ optical transition at 578.4 nm in two odd isotopes of ytterbium [8]. Using a femtosecond-laser frequency comb [9], we make precision absolute frequency measurements with an uncertainty of 4.4 kHz—an improvement of nearly 10^6. This accurate frequency knowledge will expedite the pursuit of an ytterbium-based optical clock. The short-term stability of an ytterbium optical clock will surpass that of the best cesium primary standard [10] by orders of magnitude, thereby opening the possibility for exceptional accuracy.

The ytterbium ${}^1S_0 \leftrightarrow {}^3P_0$ resonance is an outstanding potential clock transition, in part because of its narrow natural linewidth (~ 10 mHz [3]). This transition is strictly dipole-forbidden from spin and orbital angular momentum considerations. An appreciable excitation probability exists in the odd isotopes, however, through hyperfine mixing of the ${}^3P_0$ level with nearby states. (See the diagram in Fig. 1.)

In an optical atomic clock based on confinement to a lattice, the lattice laser wavelength is chosen such that the ac Stark shift of the upper clock energy state matches the

shift of the ground state. This results in a vanishing net perturbation to the clock frequency. The shift-canceling wavelength for ytterbium is calculated to be 752 nm [3], readily accessible by high-power cw titanium-sapphire laser systems. Furthermore, the $J = 0$ to $J = 0$ transitions in Yb reported here are expected to depend minimally on lattice polarization [3], enabling straightforward implementation of two- and three-dimensional lattice geometries. However, experimental measurements of the shift-canceling lattice wavelength are required to rule out possible deleterious effects of higher-order processes.

A lattice-based neutral atomic frequency reference has several important features. In addition to a high signal-to-noise ratio (due to averaging signals from $\gtrsim 10^5$ atoms) the lattice enables long spectroscopic interaction times (1 s) and accompanying narrow-linewidth measurements. These
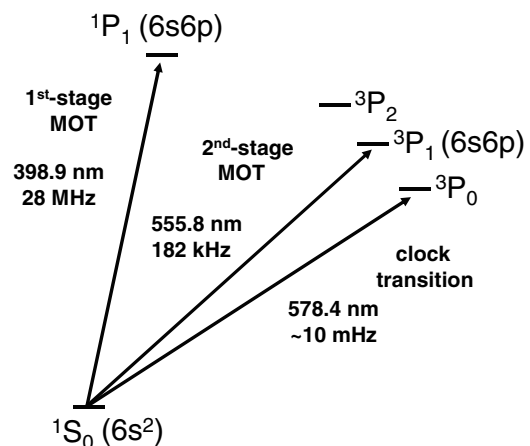

FIG. 1. Ytterbium atomic energy levels. Wavelengths and natural linewidths are indicated for the relevant cooling, trapping, and clock transitions. The fine-structure splitting is not to scale. The hyperfine structure for the ^{171}Yb (I = 1/2) and ^{173}Yb (I = 5/2) isotopes is ignored for clarity.

two factors lead to high short-term stability. Also, the high accuracy of an ion system may be reached since its motional state control can be mimicked for each neutral atom in its respective lattice site. With each atom tightly confined to a fraction of the lattice wavelength (i.e., in the Lamb-Dicke regime), systematic frequency uncertainties associated with the Doppler shift are essentially eliminated [11]. Additionally, if a three-dimensional lattice has an occupation number less than one, collisional frequency shifts are expected to be negligible [2].

The use of a lattice-insensitive $J = 0$ to $J = 0$ narrow clock transition was first proposed for [87]Sr [12]. Recently, its analogous clock transition has been observed and measured [13], and high-resolution Doppler-free spectroscopy of lattice-confined [87]Sr atoms has been demonstrated [14,15]. One of the primary differences between Sr and Yb is their hyperfine structure. The nuclear spins of [87]Sr, [171]Yb, and [173]Yb are I = 9/2, 1/2, and 5/2, respectively. The larger total angular momentum of [87]Sr leads to lower temperatures [5,16], but may introduce unwanted experimental issues such as optical pumping and increased sensitivity to lattice polarization for the clock transition [2]. Ytterbium offers two odd isotopes with reasonable natural abundance ([171]Yb: 14%, [173]Yb: 16%) and the possibility to explore the relatively simple $I = 1/2$ spin system. Ytterbium has been laser cooled and trapped [4,5,17] and researchers have achieved Bose-Einstein condensation of [174]Yb by all-optical means [18].

As a first step toward a lattice-based system, we investigated the ultranarrow clock transition in a cloud of cold atoms. Ytterbium can be cooled to low temperatures because its energy level structure (see Fig. 1) is amenable to two successive stages of laser cooling and trapping as described below. Lower temperatures aid in the search for absorption on the ultranarrow clock transition because smaller Doppler widths lead to higher-contrast signals. Courtillot *et al.* [13] used a novel approach to find the $(5s^2)^1S_0 \leftrightarrow (5s5p)^3P_0$ clock transition in [87]Sr by monitoring the trap fluorescence from 2 mK atoms in a first-stage magneto-optical trap (MOT). The clock transition was observed as a 1% depletion in fluorescence when the clock laser frequency was on resonance. Here we cool an Yb atomic sample to tens of microkelvins with the addition of a second-stage MOT using the 555.8 nm transition shown in Fig. 1. Then, with 20 mW of power at 578.4 nm focused to a spot slightly larger than the size of the atomic cloud ($1/e^2$ beam radius of ~1 mm), we drive the clock transition at an estimated Rabi rate of ~$2\pi \times 4$ kHz. A single π pulse addresses approximately 2% of the atoms in a 70 μK sample ($\Delta\nu_{\text{Doppler}} \approx 240$ kHz, full-width half-maximum). Adding successive π pulses during the lifetime of the trap, we are able to address more atoms and increase trap depletion to over 80%.

As shown in Fig. 1, a violet transition at 398.9 nm offers a broad transition (natural linewidth: $\Gamma = 2\pi \times 28$ MHz)

for a first-stage MOT. We obtain temperatures of ~3 mK for [171]Yb and [173]Yb, and ~7 mK for [174]Yb. Similar to sub-Doppler cooling in the alkaline-earth metals [16], the lower temperatures of the odd isotopes can be explained by the presence of hyperfine ground-state structure [5]. The cooling laser source is a system of InGaN laser diodes [4,19]. Two slave diodes are injection locked by a master external-cavity diode laser. The frequency of the light from each slave laser is controlled independently through the use of acousto-optic modulators. The master laser is locked to the desired isotope by saturation spectroscopy using an Yb hollow-cathode lamp [20]. The output from one of the slave lasers is red detuned from resonance by $\delta \equiv \omega_{\text{laser}} - \omega_0 \approx -4\Gamma$ and acts as a slowing beam as it counter propagates with respect to an ytterbium atomic beam generated by an oven at ~777 K. A second slave laser produces $\sigma^+ - \sigma^-$ trapping beams (red detuned to $\delta = \Gamma/1.75 = -2\pi \times 16$ MHz) for the MOT. The trap is formed in the atomic beam in a standard manner by intersecting three retro-reflected beams at the zero point of a ~4.5 mT/cm magnetic field gradient: two at 90° in the horizontal plane and one normal to the horizontal plane. All of the beams are fiber coupled to the vacuum chamber region to provide spatial filtering. The slowing beam power is 4 mW, and the powers of the horizontal and vertical trapping beams are 2 and 1 mW, respectively. Each trapping beam has a $1/e^2$ radius of ~4 mm. Under these conditions, up to ~2×10^6 atoms can be trapped in the violet MOT.

Atoms from the violet MOT are transferred to a 555.8 nm second-stage MOT with up to 70% efficiency. As shown in Fig. 1, the natural linewidth of this $^1S_0 \leftrightarrow {}^3P_1$ transition is $\Gamma = 2\pi \times 182$ kHz, which implies a ~150-fold decrease in the Doppler cooling limit with respect to the 398.9 nm transition. Measured temperatures are then as low as 30 μK for [173]Yb, with trap lifetimes as long as 3 s. The second-stage cooling laser light is provided by a narrow linewidth fiber laser system (1 W at 1111.6 nm) that is frequency doubled. Second-harmonic light at 555.8 nm with a power of 30 mW is generated in a single pass through a 5 cm MgO-doped periodically poled lithium niobate crystal [21,22]. This light is then split into trapping, probe, and spectroscopy (locking) beams. After fiber coupling to the chamber area, up to 4.5 mW is measured in each trapping beam with $1/e^2$ radii of ~4 mm. The frequency is red detuned several linewidths from resonance. The frequency-modulated spectroscopy beam intersects the atomic beam at normal incidence downstream from the trapping area. Phase-sensitive detection of the green fluorescence is used to stabilize the IR master oscillator to the desired isotope.

The yellow excitation light at 578.4 nm is provided by a highly stable dye laser. The laser is spectrally narrowed using a tunable high-finesse reference cavity, which is in turn locked to an ultrastable cavity with a finesse of greater

than 150 000 [23]. Some of the frequency-correction capabilities were removed from this system to maximize power for spectroscopy, resulting in a laser linewidth of ~5 kHz. The longitudinal modes of the ultrastable cavity drift at a rate of a few hundred hertz/day and therefore serve as an effective reference in the search for the extremely narrow $^1S_0 \leftrightarrow {}^3P_0$ clock transition. We first found the resonance in both ^{171}Yb and ^{173}Yb by applying the 578.4 nm clock radiation while the atoms were in the green MOT. However, the absorption line shape was Stark shifted by ~300 kHz due to the presence of the green trapping beams.

To avoid this large Stark shift, we used a method based on a series of excitation pulses that follows the demonstration and suggestions contained in Ref. [13]. The violet MOT is loaded for 100 ms in the presence of the green MOT beams. The violet beams are then turned off and the green MOT ($dB/dz \approx 1.5$ mT/cm) is allowed to settle for 30 ms. After turning off the green MOT beams the fluorescence from a 200 μs resonant green pulse is detected with a photomultiplier tube and integrated to serve as a normalization factor for each measurement. A sequence of 300 μs, 578.4 nm yellow pulses (traveling or standing wave) is applied in a nearly vertical direction while the trapping beams are off. The pulses are separated in time by 1 ms, during which time the green MOT is on. This allows the remaining trapped atoms to rethermalize (i.e., refill velocity holes) and the excited atoms to gravitationally accelerate downward and out of resonance with the next pulse. The green MOT magnetic field gradient is on during the excitation pulses, although its effect on the $J = 0$ transition is negligible for this measurement (~10 kHz/mT [3]). After the prescribed number of yellow pulses (typically 50), the integrated fluorescence from a final resonant green pulse serves to read the atom depletion relative to the first normalization green pulse.

Fig. 2 shows excitation data for the doubly forbidden $^1S_0 \leftrightarrow {}^3P_0$ transition in ^{171}Yb and ^{173}Yb. The ordinate in the figures indicates the fraction of trapped atoms relative to the off-resonance case. Line shapes are determined primarily by the velocity distributions of the atoms in the second-stage MOT, and are fit well by Gaussians. The corresponding temperatures, 84 μK for ^{171}Yb and 48 μK for ^{173}Yb, agree well with time-of-flight measurements. The difference in temperatures most likely results from the difference in angular momenta for the two isotopes [5,16].

The absolute frequencies of these resonances were determined from a beat frequency between the stabilized cw excitation laser and specific modes of an optical frequency comb. The comb was generated from a broadband Ti:sapphire femtosecond-laser oscillator, which enables the measurement of the comb offset frequency without the use of highly nonlinear fiber [24,25]. Both the mode spacing and the offset frequency of the self-referenced
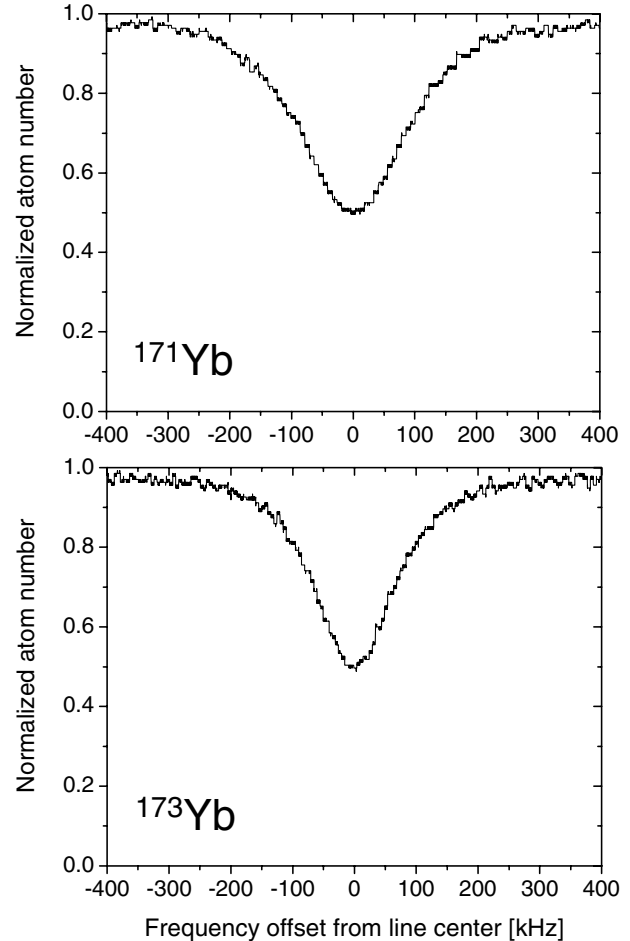

FIG. 2. Line shape data for the ^{171}Yb $^1S_0 \leftrightarrow {}^3P_0$ and ^{173}Yb $^1S_0 \leftrightarrow {}^3P_0$ transitions, averaged 4 times and taken with 50 traveling-wave pulses at each 7.5 kHz step. Each step lasts ~200 ms. The Doppler-broadened widths correspond to temperatures of 84 μK for ^{171}Yb and 48 μK for ^{173}Yb.

comb are stabilized to a hydrogen maser that is calibrated by the National Institute of Standards and Technology cesium primary frequency standard. The mode spacing of the comb is determined by the 998 731 460 Hz repetition rate of the laser. The measured frequency of the $(6s^2)^1S_0 \leftrightarrow (6s6p)^3P_0$ transition in ^{171}Yb (F = 1/2) is 518 295 836 591.6 ± 4.4 kHz. The measured frequency of the $(6s^2)^1S_0 \leftrightarrow (6s6p)^3P_0$ transition in ^{173}Yb (F = 5/2) is 518 294 576 847.6 ± 4.4 kHz. The measured isotope shift between ^{171}Yb and ^{173}Yb is 1 259 744.0 kHz. This is comparable to the corresponding isotope shift of 1 271 600 kHz measured for the $^1S_0 \leftrightarrow {}^3P_1$ transition at 555.8 nm [26].

The major source of uncertainty for these measurements was a potential Doppler shift due to a systematic drift velocity of the atomic cloud upon release from the MOT. In order to test for such a drift we compared measurements using traveling- and standing-wave excitation laser configurations. Since these measurements were made with the excitation beam at near-vertical incidence, we needed to

account for the gravitational acceleration of the atoms during the 300 μs excitation pulses. This leads to an effective chirping of the laser frequency as seen by the atoms and causes a ~2.5 kHz shift for traveling-wave excitation. With this correction to the traveling-wave results, the difference between traveling- and standing-wave results was less than the measurement-to-measurement scatter ($\sigma \approx 700$ Hz). However, this comparison does not isolate the effects of a possible cloud drift velocity from an intensity imbalance in the counter-propagating beams.

In principle, a more reliable constraint on the Doppler-related uncertainty can be set by our observation of a saturation dip in standing-wave line shapes, since it corresponds to zero-velocity atoms. Using the maximum observed separation between the dip and Doppler line center, we conservatively set Doppler-related uncertainty at 4.0 kHz.

Constrained to 4.0 kHz, the Doppler-related error dominates other systematic factors. Including uncertainties associated with line shape fitting, ac Stark shifts from residual trapping laser light, and comb measurements, the total measurement uncertainty is 4.4 kHz. This level of uncertainty of the absolute clock transition frequencies in ytterbium will expedite Doppler-free spectroscopy in a lattice—an important step in the development of a new high-performance optical atomic clock.

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*Electronic address: hoycha@boulder.nist.gov
†Also at University of Colorado, Boulder, CO 80309, USA.
‡Also at Los Alamos National Laboratory, Physics Division, MS H803, Los Alamos, NM 87545, USA.

[1] S. A. Diddams, J. C. Bergquist, S. R. Jefferts, and C. W. Oates, Science **306**, 1318 (2004).
[2] H. Katori, M. Takamoto, V. G. Pal'chikov, and V. D. Ovsiannikov, Phys. Rev. Lett. **91**, 173005 (2003).
[3] S. G. Porsev, A. Derevianko, and E. N. Fortson, Phys. Rev. A **69**, 021403(R) (2004).
[4] C. Y. Park and T. H. Yoon, Phys. Rev. A **68**, 055401 (2003).
[5] R. Maruyama, R. H. Wynar, M. V. Romalis, A. Andalkar, M. D. Swallows, C. E. Pearson, and E. N. Fortson, Phys. Rev. A **68**, 011403(R) (2003).
[6] T. Hong, C. Cramer, W. Nagourney, and E. N. Fortson, Phys. Rev. Lett. **94**, 050801 (2005).
[7] W. C. Martin, R. Zalubas, and L. Hagan, *Atomic Energy Levels: The Rare-Earth Elements* (U.S. Dept. of Commerce, National Bureau of Standards, Washington, D.C., 1978).
[8] Independent observations of these transitions will appear in T. Hong, C. Cramer, E. Cook, W. Nagourney, and E. N. Fortson, physics/0504216 [Opt. Lett. (to be published)].
[9] T. Udem, J. Reichert, R. Holzwarth, and T. W. Hänsch, Phys. Rev. Lett. **82**, 3568 (1999).
[10] S. R. Jefferts, T. P. Heavner, and E. A. Donley, Jpn. J. Appl. Phys. **43**, 2803 (2004).
[11] J. C. Bergquist, W. M. Itano, and D. J. Wineland, Phys. Rev. A **36**, R428 (1987).
[12] H. Katori, in *Proceedings of the 6th Symposium on Frequency Standards and Metrology*, edited by P. Gill (World Scientific, Singapore, 2002), p. 323.
[13] I. Courtillot, A. Quessada, R. P. Kovacich, A. Brusch, D. Kolker, J.-J. Zondy, G. D. Rovera, and P. Lemonde, Phys. Rev. A **68**, 030501(R) (2003).
[14] M. Takamoto, F. L. Hong, R. Higashi, and H. Katori, Nature (London) **435**, 321 (2005).
[15] Andrew D. Ludlow, Martin M. Boyd, T. Zelevinsky, Seth M. Foreman, Sebastian Blatt, Mark Notcutt, Tetsuya Ido, and Jun Ye, physics/0508041.
[16] X. Xu, T. H. Loftus, J. W. Dunn, C. H. Greene, J. L. Hall, A. Gallagher, and J. Ye, Phys. Rev. Lett. **90**, 193002 (2003).
[17] T. Loftus, J. R. Bochinski, and T. W. Mossberg, Phys. Rev. A **61**, 061401 (2000).
[18] Y. Takasu, K. Maki, K. Komori, T. Takano, K. Honda, M. Kumakura, T. Yabuzaki, and Y. Takahashi, Phys. Rev. Lett. **91**, 040404 (2003).
[19] K. Komori, Y. Takasu, M. Kumakura, Y. Takahashi, and T. Yabuzaki, Jpn. J. Appl. Phys. **42**, 5059 (2003).
[20] J. I. Kim, C. Y. Park, J. Y. Yeom, E. B. Kim, and T. H. Yoon, Opt. Lett. **28**, 245 (2003).
[21] N. Pavel, I. Shoji, T. Taira, K. Mizuuchi, A. Morikawa, T. Sugita, and K. Yamamoto, Opt. Lett. **29**, 830 (2004).
[22] J.-P. Wallerand, in *2004 Conference on Precision Electromagnetic Measurements Digest, IEEE* (IEEE, Piscataway, NJ, 2004), p. 50.
[23] B. C. Young, F. C. Cruz, W. M. Itano, and J. C. Bergquist, Phys. Rev. Lett. **82**, 3799 (1999).
[24] A. Bartels and H. Kurz, Opt. Lett. **27**, 1839 (2002).
[25] T. M. Ramond, S. A. Diddams, L. Hollberg, and A. Bartels, Opt. Lett. **27**, 1842 (2002).
[26] D. L. Clark, M. E. Cage, D. A. Lewis, and G. W. Greenlees, Phys. Rev. A **20**, 239 (1979).