Observation and absolute frequency measurements of the $^1S_0-^3P_0$ optical clock transition in ytterbium

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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 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 $518,295,836,593.2 \pm 4.4 \text{ kHz}$. The measured frequency in $^{173}$Yb (F=5/2) is $518,295,857,050.0 \pm 4.4 \text{ kHz}$. Measurements are made with a femtosecond-laser frequency comb calibrated by the NIST cesium fountain clock and represent nearly a million-fold reduction in uncertainty. The natural linewidth of these J=0 to J=0 transitions is calculated to be $\sim 10 \text{ mHz}$, making them well-suited to support a new generation of optical atomic clocks based on confinement in an optical lattice.

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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. Such a clock system comprises a dipole-force optical lattice trap that confines an ensemble of neutral atoms individually to sub-wavelength sites for Doppler- and recoil-free precision spectroscopy. 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. However, until now the absolute frequency of this resonance was known in tables 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.

Using a femtosecond-laser frequency comb, 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, whose performance can potentially surpass that of the best cesium primary standard by orders of magnitude.

The ytterbium $^1S_0 \leftrightarrow ^3P_0$ resonance is an outstanding potential clock transition, in part because of its narrow natural linewidth ($\sim 10 \text{ mHz}$). This transition is strictly dipole-forbidden from spin and orbital angular momentum considerations. An appreciable excitation probability exists, however, through hyperfine mixing of the $^3P_0$ level with nearby states in the odd isotopes. (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, 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, 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 $\geq 10^5$ atoms) the lattice enables long spectroscopic interaction times (1 s) and accompanying narrow-linewidth measurements. These 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. Additionally, if a three-dimensional lattice has an occupation number less than one, collisional frequency shifts are expected to be negligible.

Offering both high stability and high accuracy, this best-of-both-worlds frequency reference scheme was first proposed for $^{87}$Sr. Recently, its analogous clock transition has been observed and measured, and high-resolution Doppler-free spectroscopy of lattice-confined $^{87}$Sr atoms has been demonstrated. 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, but may introduce unwanted experimental issues such as optical pumping and increased sensitivity to lat-
tice polarization for the clock transition \( \frac{1}{2} \). 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 \( ^{171}\)Yb and \(^{173}\)Yb, and researchers have achieved Bose-Einstein condensation of \(^{174}\)Yb by all-optical means \( ^{17}\).

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. \(^{18}\) 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 \(\sim 1\) mm), we drive the clock transition at an estimated Rabi rate of \(\sim 2\pi \times 4\) kHz. A single \(\pi\)-pulse addresses approximately 2\% of the atoms in a \(70\) \(\mu\)K sample (\(\Delta \nu_{\text{Doppler}} \approx 240\) kHz, full-width half-maximum). Adding successive \(\pi\)-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 blue 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 \(\sim 3\) mK for \(^{171}\)Yb and \(^{173}\)Yb, and \(\sim 7\) mK for \(^{174}\)Yb. Similar to sub-Doppler cooling in the alkaline-earths \( ^{15}\), the lower temperatures of the odd isotopes can be explained by the presence of hyperfine ground-state structure \( ^{17}\). The cooling laser source is a system of InGaN laser diodes \( ^{18}\). 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 \( ^{18}\). 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 the atomic beam.

A second slave laser produces \(\sigma^+ - \sigma^-\) trapping beams (red-detuned to \(\delta = \Gamma/1.75 = -2\pi \times 16\) MHz) for the MOT that uses a magnetic field gradient of \(\sim 4.5\) mT/cm. 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 mW and 1 mW, respectively. Under these conditions, up to \(\sim 2 \times 10^6\) atoms can be trapped in the blue MOT.

Atoms from the blue MOT are transferred to a 555.8-nm second-stage MOT with up to 65\% 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 \(\sim 150\)-fold decrease in the Doppler cooling limit with respect to the 398.9 nm transition. Measured temperatures are then as low as 30 \(\mu\)K 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 \( ^{20}\). \(^{21}\). 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 \(\sim 4\) mm. The frequency is red de-tuned 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 \( ^{22}\). Some of the frequency-correction capabilities were removed from this system to maximize power for spectroscopy, resulting in a laser linewidth of \(\sim 5\) kHz. The longitudinal modes of the ul-

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**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.
trastable 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. Ultimately, we 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 lineshape is Stark-shifted by $\sim$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 blue MOT is loaded for 100 ms in the presence of the green MOT beams. The blue 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 $\mu$s resonant green pulse is detected with a photomultiplier tube and integrated to serve as a normalization factor for each measurement. A sequence of 300 $\mu$s, 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 re-thermalize (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 ($\sim$10 kHz/mT). 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.

Figures 2(a) and 2(b) show excitation data for the doubly-forbidden $^1S_0 \leftrightarrow ^3P_0$ transition in $^{171}$Yb and $^{173}$Yb, respectively. The ordinate in the figures indicates the fraction of trapped atoms relative to the off-resonance case. Lineshapes 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 $\mu$K for $^{171}$Yb and 48 $\mu$K 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.

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 [23, 24]. Both the mode spacing and the offset frequency of the self-referenced comb are stabilized to a hydrogen maser that is calibrated by the NIST cesium primary frequency standard. The mode-spacing of the comb is determined by the 998731460 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 518,295,836,593.2 $\pm$ 4.4 kHz. This number represents the mean of six measurements that have a standard deviation of 1072 Hz. The measured frequency of the (6s$^2$)$^1S_0 \leftrightarrow (6s6p)^3P_0$ transition in $^{173}$Yb (F=5/2) is 518,294,576,850.0 $\pm$ 4.4 kHz. This number represents the mean of three measurements that have a standard deviation of 216 Hz. To within the stated uncertainty, the isotope shift between $^{171}$Yb and $^{173}$Yb is 1,259,743.2 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.

Two tests allow us to limit Doppler-related uncertainties associated with a possible atomic cloud drift velocity to 4.0 kHz. First, there is no measurable shift in line centers between traveling- and standing-wave configurations to within the fitting uncertainty. Second, a satura-
tion dip present in higher-resolution, standing-wave line-shapes constrains drift-velocity shifts by indicating the relationship between Doppler line-center and the laser frequency associated with the zero-velocity atoms. We also note the presence of another saturation feature that is likely due to the near-vertical angle of the yellow spectroscopy laser beam. The subtle effects of this geometry complicate accuracy evaluation: during a single probe pulse there is non-negligible atomic gravitational acceleration leading to effective chirping of the laser frequency as seen by the atoms. Further investigation will be required to fully understand the implications of this geometry.

Constrained to 4.0 kHz, the Doppler-related error dominates other systematic factors. Including uncertainties associated with lineshape fitting, ac Stark shifts from residual trapping laser light, and comb measurements, the total measurement uncertainty is 4.4 kHz. This accurate knowledge 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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