Kilohertz-Resolution Spectroscopy of Cold Atoms with an Optical Frequency Comb

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We have performed sub-Doppler spectroscopy on the narrow intercombination line of cold calcium atoms using the amplified output of a femtosecond laser frequency comb. Injection locking of a 657-nm diode laser with a femtosecond comb allows for two regimes of amplification, one in which many lines of the comb are amplified, and one where a single line is predominantly amplified. The output of the laser in both regimes was used to perform kilohertz-level spectroscopy. This experiment demonstrates the potential for high-resolution absolute-frequency spectroscopy over the entire spectrum of the frequency comb output using a single high-finesse optical reference cavity.

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The broad coherent optical bandwidth from frequency combs based on mode-locked femtosecond lasers makes possible the optical-to-microwave division necessary for direct counting of optical frequencies. These advances have led to significant simplification in optical frequency measurement [1,2]. With the development of ultrabroadband titanium-doped sapphire (Ti:sapphire) lasers [3–5] and the coherent broadening possible with nonlinear optical fibers [6], the optical bandwidth of comb generators extends from near infrared into visible frequencies. The subsequent stabilization of such frequency combs allows for absolute optical measurements over hundreds of terahertz making them an ideal tool for spectroscopy. Already, frequency combs have been used for direct spectroscopy of allowed two-photon and one-photon transitions [7–13]. Previous measurements with the direct output of a frequency comb have been performed by stabilizing the comb to microwave references and studying transitions with linewidths on the order of a few megahertz. A greater potential can be realized by transferring the stability of a narrow optical reference to the comb allowing the study of much narrower atomic resonances.

A new paradigm is attained by combining the extremely precise optical synthesis of the comb with cold atomic samples that allow long interaction times and permit nonlinear spectroscopy of very narrow optical transitions with low optical powers. Here we work with a forbidden intercombination transition in cold neutral calcium (^{40}Ca) and resolve features as narrow as 1.2 kHz. Because of the relatively high residual temperatures of our atoms, we amplify the comb teeth centered at the transition wavelength in a laser diode [14]. Additionally, we demonstrate the comb's wavelength versatility by stabilizing the comb to a cw laser referenced to a high-finesse cavity at 534 nm, while performing high-resolution spectroscopy at 657 nm. Direct spectroscopy with an optical comb could potentially yield access to transition frequencies that would be difficult to reach otherwise.

The laser that is used in the experiments is a ring cavity mode-locked femtosecond laser based on Ti:sapphire, with a repetition rate around 1 GHz [15]. The laser produces an optical spectrum with rigorously spaced and coherently related optical frequency components that are characterized by two rf frequencies. The first frequency is the laser repetition rate, f_{rep}, which sets the mode spacing and is determined by the laser cavity length. The second is the carrier-envelope offset frequency, f_0, which defines the absolute comb position and is determined by dispersion in the laser cavity [1]. The laser spectrum is composed of more than 10^5 optical frequencies ν_n, each of which is described absolutely by the equation, $\nu_n = n f_{\mathrm{rep}} + f_0$, where n is the mode number ($\sim 10^5$) that multiplies f_{rep} up from microwave into optical frequencies. The laser offset frequency is stabilized using a standard f-to-$2f$ interferometer, which uses a self-referencing technique [2] that compares frequency-doubled comb lines on the low frequency end of the comb to fundamental light on the high frequency end of the comb (Fig. 1). This comparison yields f_0 as an rf beat frequency that is stabilized to a synthesized reference frequency. The error signal between the two frequencies servos f_0 via feedback to an acousto-optic modulator that adjusts the Ti:sapphire pump laser power (solid state pump source at 532 nm), which in turn controls the dispersion in the laser cavity. The octave bandwidth necessary for stabilization of f_0 is obtained via intracavity continuum generation in the Ti:sapphire crystal [15]. The resulting power per mode of the Ti:sapphire laser spectrum is sufficient for both stabilization of the comb and for spectroscopy at 657 nm without additional broadening in nonlinear fibers.

To use the mode-locked laser as the local oscillator for high-precision spectroscopy requires narrow optical comb lines. To this end, the frequency of one mode of the comb is stabilized to a fiber laser at 1068 nm. Part of this fiber laser output is frequency doubled and referenced to an optical cavity at 534 nm with a finesse of 16 000 and a drift rate of less than 1 Hz/s [16]. Because the optical cavity is located

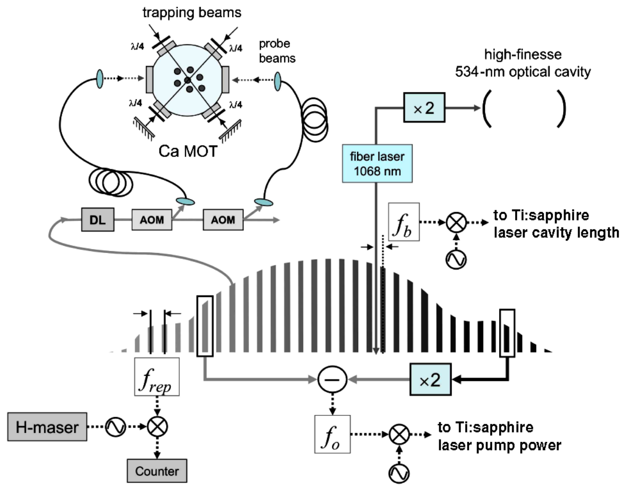

FIG. 1 (color online). Experimental depiction for high-precision optical spectroscopy of cold Ca atoms using a femtosecond laser frequency comb. Acousto-optic modulators (AOM) switch light between two optical fibers that deliver counter-propagating 657-nm pulses to the Ca magneto-optical trap (MOT) (the 423-nm shelving detection beams are left out for simplicity). An f-to-$2f$ self-referencing technique allows phase stabilization of the laser offset frequency, f_0. Phase stabilization of the laser repetition rate, $f_{\rm rep}$, is obtained by phase locking the comb to a fiber laser at 1068 nm that is stabilized to a high-finesse optical cavity. Measurement of the laser repetition rate vs a hydrogen maser permits for frequency calibration in the measurements.

in a different part of the building, a 300 m long optical fiber delivers the light from the cavity-stabilized fiber laser to the femtosecond laser frequency comb. The fiber length fluctuations are Doppler canceled using standard techniques [17]. We obtain a heterodyne beat signal between the comb and the fiber laser with a signal-to-noise ratio of ≈ 40 dB in a 300 kHz resolution bandwidth. This heterodyne beat is phase locked to a synthesized reference frequency via feedback to a piezoelectric actuator that adjusts the laser cavity length. To perform spectroscopy, we scan the synthesized reference and hence the optical lines of the comb. With both the frequency of a single mode and the offset frequency stabilized, we obtain an optical linewidth of $\sim$3 Hz for every comb line spanning the entire spectrum of the laser [15,18]. Delivery of the comb light to the clock experiment via a 20-m long polarization maintaining optical fiber without Doppler noise cancellation results in a degradation of the optical linewidth to several hundred Hertz. Noise cancellation on the fiber would be required to achieve resolutions significantly less than 1 kHz.

We use the stabilized comb to study the narrow 657-nm intercombination line of the Ca atomic clock developed at NIST [19]. The one-photon transition between the $4s^2\,{}^1S_0$ $(m = 0)$ and $4s4p\,{}^3P_1$ $(m = 0)$ levels, which is forbidden in the L-S coupling approximation, has a narrow natural linewidth of 374 Hz [20] (see Fig. 2). Using standard trapping and cooling techniques described in detail in Ref. [19], we obtain a Ca sample with 6×10^7 atoms

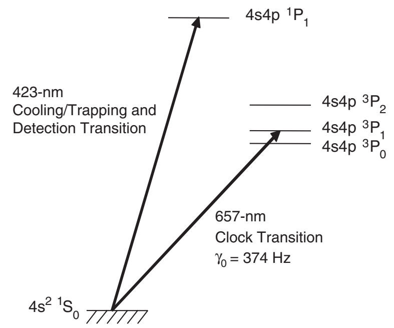

FIG. 2. Relevant ^{40}Ca energy levels.

cooled to 2 mK in 2 ms. By taking the direct output of the comb at 657 nm we have approximately 2.4 μW per mode. This power would be sufficient for saturated absorption spectroscopy but delivery of the comb light via single-mode optical fiber to the Ca atoms and additional losses due to optical components results in a reduction of power, leaving $\sim$400 nW per mode at 657 nm. To compensate for this loss, we employ a similar technique as that described in Ref. [14] and inject comb light into an antireflection coated 657-nm diode laser. The diode laser is injected with comb light centered at 657 nm, which is narrowed to $\simeq$0.5 nm using a 2400 grooves/mm diffraction grating and a single-mode optical fiber. For a diode laser current of 65 mA (just above the self-lasing threshold) this arrangement yields a nearly eight-times optical amplification at 657 nm. The total power delivered to the atoms is 1.6 mW at 65 mA, with $\sim$3 μW in the comb line at the transition wavelength.

The amplified comb teeth are delivered to the Ca atoms via optical fiber whereby acousto-optic modulators act as switches to control the time duration and the separation time of the optical pulses delivered to the atoms (see Fig. 1). Successive counterpropagating pulses from the comb are delivered to the Ca atoms, which allows for a Doppler-free saturated absorption on the optical transition. Typically, we deliver pulses with a duration of 100 μs whose frequency is tuned through the resonance of the $4s^2\,{}^1S_0 \rightarrow 4s4p^3P_1$ transition. Excitation of the Ca sample is measured using a shelving detection scheme [19,21], whereby fluorescence from the strongly allowed ${}^1S_0 \rightarrow {}^1P_1$ transition (423 nm), measured before and after 657-nm excitation, reveals the ground state depletion due to excitation to the ${}^3P_1$ state (see Fig. 2). The 423-nm normalization pulse applied before the 657-nm excitation light is 10 μs long and the 423-nm probe pulse is 30 μs long. Figure 3(a) shows a measurement of the sub-Doppler photon recoil splitting of 23 kHz [22,23] taken with the optical frequency comb. The right dip in the figure is the standard saturated absorption dip (located at the center of the Doppler profile). The left dip is due to stimulated emission from the second pulse and is shifted one-photon recoil from the absorption dip. Note that the unperturbed atomic resonant frequency should be exactly halfway be-

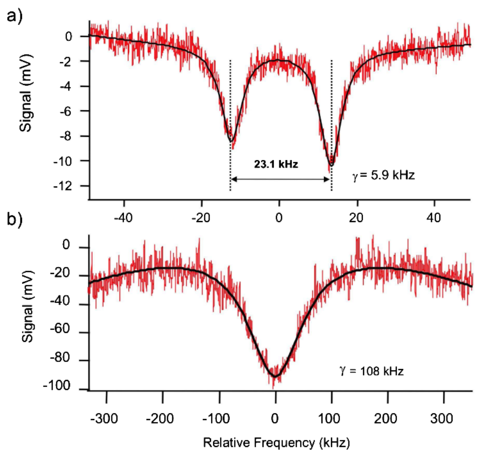

FIG. 3 (color online). Saturated absorption dip observed on the Doppler profile of the Ca clock transition using two counter-propagating pulses from a comb-injected slave laser with a current of (a) 65 mA and (b) 98 mA. The double peak observed in the case of low power broadening [case (a)] is the recoil doublet. In case (b), amplification of preferred comb lines leads to greater power broadening (linewidth = 108 kHz), thereby making the double structure indistinguishable. Each point in the plots is the result of 150 ms of averaging. The y axis is offset for convenience and 1 mV corresponds to ~600 atoms, which is estimated from the measured fractional excitation and the trapped atom number.

tween the two dips [23]. Based on simulations similar to those presented in Ref. [23], we expect a Fourier-limited linewidth of 5.8 kHz for a 100 μs square pulse, consistent with that observed in Fig. 3(a).

Figure 3(b) shows the saturation dip when the laser diode current is increased to 98 mA (i.e., much higher than the threshold current). At this current we observe preferential amplification of particular comb teeth. By careful adjustment of the diode temperature and current, the comb line resonant with the transition can be made to contain more than 10% of the amplified power, yielding ~2000 times amplification of that particular mode leading to a power per mode of ~800 μW. This amplification factor is inferred from the power broadening measured in the data shown in Fig. 3(b). We independently confirmed this behavior by heterodyning the amplified comb light against a stable cw 657 nm optical frequency reference. A consequence of the increased amplification is slightly higher amplitude noise on the 657-nm probe light, as seen in Fig. 3. The comb light alone as measured using a fast photodetector results in an rf spectrum with harmonics of the comb repetition rate. The heterodyne beat between the cw laser and the individual comb lines is observed as sidebands on these repetition rate harmonics. At low diode laser currents, near 65 mA, we observe sidebands with uniform amplitude, which do not exhibit strong dependence with current and temperature. However, at 98 mA, careful adjustment of diode laser current allows for enhancement or suppression of the amplitude of particular sidebands.

With 800 μW of amplified light at the transition wavelength we have sufficient optical power for observation of Bordé-Ramsey fringes, allowing for higher resolution spectroscopy of the Ca clock transition. The technique consists of applying two $\pi/2$ pulses from one direction (separated by a time $T/2$) and then two $\pi/2$ pulses from the opposite direction (separated by the same time $T/2$). The excitation probability exhibits (on top of a slowly varying envelope [24]) a sine wave pattern proportional to $\sin[\pi(\nu - \nu_0)/(2\Delta\nu)]$, where $\Delta\nu = 1/(2T)$ and ν_0 is the resonant optical frequency [25]. With the optical amplification observed for a diode laser current of 65 mA, the pulse duration necessary for $\pi/2$ pulse would not be compatible with implementing the Bordé-Ramsey technique. Figure 4 shows the fringe contrast obtained for a fixed pulse duration of 7 μs (required for a $\pi/2$ pulse with 800 μW of optical power) with varying pulse separations, $T/2$. The noise results from contributions of amplitude noise on the 657-nm probe and shot noise due to the short duration of the 423-nm normalization and detection pulses (limited due to heating of the atomic sample). The relatively long pulse duration due to our limited optical power, coupled with the broad Doppler width (~3 MHz) on the Ca clock transition, allows interaction with only a very narrow velocity class of atoms. As a result only 3% of the

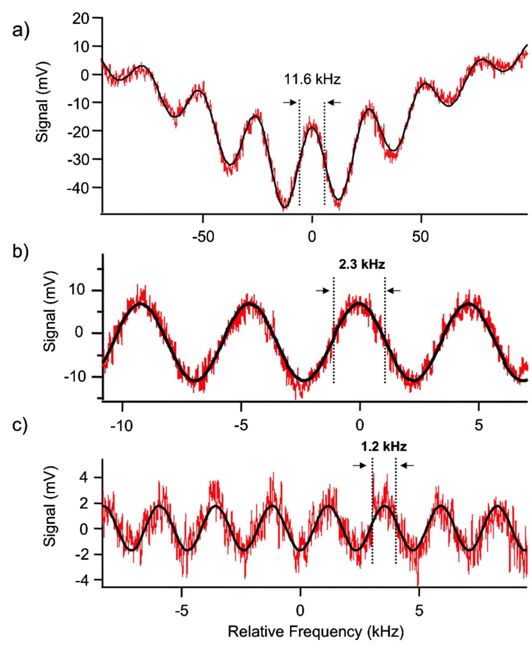

FIG. 4 (color online). Time-resolved optical Bordé-Ramsey fringes for pulse lengths of 7 μs and varying pulse separations of (a) 14 μs, (b) 104 μs, and (c) 203 μs. The decrease in the signal-to-noise ratio from (a) to (c) is the result of spontaneous population decay from the excited state and residual noise on the 657-nm light (see text for details). Each point in the plots is the result of 150 ms of averaging. The y axis is offset for convenience and 1 mV corresponds to ~600 atoms, which is estimated from the measured fractional excitation and the trapped atom number.

atoms participate in the measurement yielding a signal to-noise-ratio (S/N) of $\sim$4 after 150 ms of averaging for our narrowest linewidth of 1.2 kHz. The above techniques can be used for high-resolution absolute measurement of the line center because the weak dipole coupling of the clock transition yields a negligible ac Stark shift due to other comb components. Even for the unlikely case of a completely asymmetric distribution of comb lines with equal amplitude, we estimate this shift to be <1 mHz.

In summary, we have used the amplified output of a femtosecond optical frequency comb to perform kilohertz-level spectroscopy of the $4s4s\,^1S_0(m=0)\rightarrow 4s4p\,^3P_1(m=0)$ transition in a cold sample of atomic Ca. We take advantage of the comb's wavelength versatility by using an optical cavity at 534 nm as a local oscillator, while performing spectroscopy at 657 nm. An additional simplification can be made by stabilizing the comb directly to a high-finesse optical cavity, removing the need for a cw transfer oscillator [26]. We observe two amplification regimes when injecting a 657-nm diode laser with 0.5 nm of comb light. Close to threshold, near uniform amplification of the comb light is observed. At higher diode laser currents, preferential amplification results in a significant increase in optical power at the transition wavelength. Use of an atomic sample with higher confinement and lower residual temperature, however, could potentially allow for measurement of narrower transitions with a single unamplified comb line.

The marriage between cold atoms and ultrastable combs opens the potential for using a frequency comb stabilized to a single optical reference and performing spectroscopy at frequencies difficult to reach otherwise. An interesting example would be to explore neutral Yb confined in an atomic lattice at NIST [27]. The high lattice confinement and nanokelvin temperatures will allow for near-unity excitation of the atoms with a 10 Hz resolution for less than 1 μW of optical power. Using the unamplified output of a single comb line for probing the Yb sample would remove the necessity to build a separate ultrastable probe laser at a wavelength of 578 nm.

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