Kilohertz-resolution spectroscopy of cold atoms with an optical frequency comb

T.M. Fortier, Y. Le Coq, J.E. Stalnaker, D. Ortega, S.A. Diddams, C.W. Oates and L. Hollberg

Los Alamos National Laboratory, P-23 Physics Division MS H803, Los Alamos NM 87545
National Institute of Standards and Technology, Time and Frequency Division, MS 847 Boulder CO 80305
Gleb Wataghin Physics Institute, State University of Campinas (UNICAMP) Brasil

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 coherent optical bandwidth provided from frequency combs based on modelocked femtosecond lasers provide the optical-to-microwave division necessary to directly count optical frequencies, which has lead to significant advances in optical frequency measurement. With the development of ultra-broadband titanium-doped sapphire (Ti:S) lasers and the coherent broadening possible with nonlinear optical fibers, 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. Previous measurements with the direct output of a frequency comb have been performed by stabilization of the comb to microwave references and studying transitions with megahertz linewidths. A greater potential can be realized by transferring the stability of a narrow optical reference to the comb.

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 for nonlinear spectroscopy of very narrow optical transitions with low optical powers. Here we work with a forbidden intercombination transition in cold neutral calcium (Ca) and resolve features as narrow as 1.2 kHz. In order compensate for the relatively high residual temperatures of our atoms, we amplify the teeth centered at the transition wavelength in a laser diode. Additionally, we demonstrate the comb’s wavelength versatility by stabilizing the comb relative to a high-finesse cavity at 534 nm, but perform high-resolution spectroscopy at 657 nm. The application of direct optical measurements with an optical frequency comb to optical clocks that rely on both very cold atoms and optical frequency combs. 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 modelocked femtosecond laser based on Ti:S, with a repetition rate around 1 GHz. 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, \( f_{\text{rep}} \), 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. The laser spectrum is composed of more than \(10^5\) optical frequencies, \(\nu_n\), each of which is described absolutely by the equation, \(\nu_n = n \times f_{\text{rep}} + f_0\), where \(n\) is the mode number (\(\sim 10^3\)) that multiplies \(f_{\text{rep}}\) up from microwave into optical frequencies. The laser offset frequency is stabilized using a standard \(f\)-to-2\(f\) interferometer, which uses a self-referencing technique 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). Stabilization of \(f_0\) is obtained by modulation of the Ti:S pump laser power (solid state pump source at 532 nm) via an acousto-optic modulator. The octave bandwidth necessary for stabilization of \(f_0\) is obtained via intra-cavity continuum generation in the Ti:S crystal. The resulting power per mode of the Ti:S 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 modelocked 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. Because the optical cavity is located in a different part of the building, a 300 m long optical fiber delivers the light from the cavity-stabilized laser fiber to the femtosecond laser frequency comb. The fiber length fluctuations are Doppler-cancelled using standard techniques. We obtain a heterodyne beat signal between the comb and the fiber laser with a signal to noise ratio of \(\sim 40\) dB in a 300 kHz resolution bandwidth. This radio frequency 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. Delivery of the comb light to the Ca experiment via a 20-m long polarization maintaining optical fiber without Doppler noise cancelation results in a degradation of the optical linewidth to several hundred Hertz. Noise cancelation on the fiber is imperative for achieving sub-kilohertz resolution measurements.

We use the stabilized comb to study the narrow 657-nm inter-combination line of the Ca atomic clock developed at NIST. 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 (see Fig. 2). Using standard trapping and cooling techniques described in detail in Ref. 15, we obtain a Ca sample with 6 \times 10^7\) atoms cooled to 2 \text{mK} in 2 ms. By taking the direct output of the comb at 657 nm we have approximately 100 nW per mode. This power is sufficient for measurement of the Doppler profile, but insufficient for saturated absorption spectroscopy, which permits higher-resolution, sub-Doppler spectroscopy of the Ca clock transition. For higher optical powers, we use a similar technique as that described in Ref. 14 by injecting 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 \(\sim 0.5\) 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 uniform ten-fold optical amplification across of the injected comb light.

The amplified comb light is delivered to the Ca atoms via optical fiber (Fig. 1) 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 counter-propagating 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 \(\mu\)s and an optical power of \(\sim 2.7\) \(\mu\)W that are 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, whereby fluorescence from the strongly allowed \(^3P_1\) \(\rightarrow\) \(^3P_0\) 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). Figure 3a) shows a measurement of the sub-Doppler photon recoil splitting of 23 kHz taken with the optical frequency comb. The right peak is the standard saturated absorption peak (located at the center of the Doppler profile), while the left peak is due to stimulated emission during the second pulse and is centered at exactly one photon recoil from the absorption peak. Note that the atomic resonance should be exactly half way between the two peaks. Given the natural linewidth of the transition and the optical linewidth
FIG. 3: Saturation 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.

of the comb light we expect a saturation linewidth on the order of 2 kHz for a 100 µs pulse. The broader observed linewidth of 5.9 kHz, measured from a fit to the double-structure, results from power broadening of the transition consistent with the applied optical power.

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 diode laser 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 up to 10 percent of the amplified power, yielding ~1000 times amplification of that particular mode. This amplification factor is inferred from the power broadening measured in the data shown in Fig. 3b). We independently confirmed this behavior by heterodyning the amplified comb light against a stable CW 657 nm optical frequency reference. 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 does 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 850 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 $\nu_0$ is the resonant optical frequency $23$. At a diode laser current of 65 mA the pulse duration necessary for $\pi$-pulse would have exceeded that of the upper state lifetime. Figure 4 shows the fringe contrast obtained for a fixed pulse duration of 7 µs (required for a $\pi/2$-pulse with 850 µW of optical power) with varying pulse separations, $T/2$. 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 atoms participate in the measurement yielding a low signal to noise ratio (S/N) of ~4 after 150 ms of averaging for our narrowest linewidth, $\Delta\nu$ of 1.2 kHz. Lower residual atoms temperatures and higher confine-
ment would narrow the velocity distribution of the atoms, resulting in significantly higher signal to noise ratios. The above technique can be used for a high-resolution absolute measurement of the line center since the AC stark shift due to other comb components should be negligible. For a completely asymmetric distribution of comb lines with equal amplitude, we estimate the AC stark shift to be $\pm 1 \text{ mHz}$.

In summary, we have used the amplified output of a femtosecond optical frequency comb to demonstrate 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 observe two amplification regimes when injecting a 657-nm diode laser with 0.5 nm of comb light. Near threshold, uniform amplification of the comb light is observed, whereas at much higher diode laser currents, preferential amplification results in a hundred-fold increase in amplification at the transition wavelength. With our relatively high atom temperatures, the greater amplification possible with the second regime is necessary for higher resolution spectroscopy of the Ca clock transition with Bordé-Ramsey fringes. 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 ultra-stable combs opens the potential for using a frequency comb stabilized to a single optical reference and performing spectroscopy at frequencies difficult to reach otherwise. One particular example would be to explore neutral Yb confined in an atomic lattice at NIST [25]. The extremely high lattice confinement and nanokelvin residual 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 ultra-stable probe laser at a challenging wavelength (578 nm).

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[1] T. H. Udem et al., Nature, 416, 233 (2002)
[2] D. J. Jones et al., Science, 288, 635 (2000)
[3] R. Ell et al., Opt. Lett., 26, 373 (2001)
[4] A. Bartels et al., Opt. Lett., 27, 1839 (2002)
[5] T. M. Fortier, D. J. Jones and S. T. Cundiff, Opt. Lett., 28, 2198 (2003)
[6] J. K. Ranka et al., Opt. Lett., 25, 25–7 (2000)
[7] R. Teets, J. Eckstein and T. W. Hansch, Phys. Rev. Lett., 38, 760 (1977)
[8] M. J. Snadden, et al., Opt. Comm., 25, 70 (1996)
[9] J. N. Eckstein et al., Phys. Rev. Lett., 48, 847 (1978)
[10] Y.V. Bakanov and V. P. Chebotayev, Appl. Phys., 12, 97 (1977)
[11] A. Marian et al., Phys. Rev. Lett., 95, 023001 (2005)
[12] V. Gerginov et al., Opt. Lett., 30, 1734–1737 (2005)
[13] A. Bartels et al., Opt. Lett., 29, 1081 (2002)
[14] F. Cruz, M. C. Stowe and J. Ye, Opt. Lett., 30, 1337 (2006)
[15] T. M. Fortier, A. Bartels and S. A. Diddams, Opt. Lett., 31, 1011 (2006)
[16] B. Young et al., Phys. Rev. Lett. 82, 3799–3802 (1999)
[17] B. Young et al., in proceedings XIV International Conference on Laser Spectroscopy, edited by R. Blatt (World Scientific, Austria, 1999), p. 61
[18] C.W. Oates et al., Eur. Phys. J. D 7 449–460 (1999)
[19] C. Degenhardt et al., Phys. Rev. A., 72 062111 (2005)
[20] K. Sengstock et al., Opt. Lett. 5, 245 (1980)
[21] J. L. Hall, C. J. Bordé and K. Uehara, Phys. Rev. A., 37, 1339 (1986)
[22] C.W. Oates et al., Phys. Rev. A., 71 (2005)
[23] Ch. J. Bordé et al., Phys. Rev. A., 30, 1836 (1984)
[24] The slowly varying envelope is due to a combination of the Doppler profile, finite duration of $\pi/2$ pulses, and recoil effect [22]
[25] Z. Barber et al., Phys. Rev. Lett. 96, (2006)