

Precision spectroscopy using a partially stabilized frequency comb

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We present a simple method for precision spectroscopy using an optical frequency comb. One mode of a 1 GHz repetition rate mode-locked Ti:sapphire laser is offset-locked to an Rb-stabilized diode laser. This partially stabilized frequency comb stays locked, unattended, for hours at a time. Using the measured offset frequency and repetition rate, we calculate the frequency of each comb mode with absolute uncertainty of about 10 kHz in a 10 s measurement window. We demonstrate the capabilities and limitations of this approach with measurements in Rb, Cs, and Ca. © 2014 Optical Society of America

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1. Introduction

Optical frequency combs based on mode-locked lasers have greatly simplified precision laser frequency measurements [1–3]. Complicated frequency chains connecting laser frequencies to primary frequency standards [4] are now replaced routinely with compact table-top experiments [5,6]. This reduction in size, cost, and complexity opens new opportunities for metrology [7], atomic clocks [8,9], communications [10], laser spectroscopy [11], and astronomy [12].

That much being said, high-accuracy frequency combs can be temperamental. Laser experiments using self-referenced frequency combs often require considerable effort just to maintain the comb, which is a sometimes aggravating situation when the comb is supposed to be a tool and not a research project in itself. This temperamental nature is partially due to the stringent operating requirements for the comb. In a typical application, a mode-locked laser is broadened so that its frequency spectrum spans an octave [13–16]. In some cases, the laser is broadened by four-wave mixing in a photonic crystal fiber or microresonator.

In others, the broadening occurs by self phase modulation in the Ti:sapphire cavity itself. Regardless of how it is done, the broadening mechanism is a critical requirement, and it is often less robust than one might hope.

In self-referenced operation, the broadened comb output is inserted into an $f - 2f$ interferometer. The low-frequency portion of the comb is frequency doubled and compared with the high-frequency portion. This comparison makes it possible to extract the carrier-envelope offset frequency, f_{ceo} , which can then be controlled.

In the frequency comb, each mode is accurately determined using the formula:

$$f_n = f_{\text{ceo}} + n f_{\text{rep}}, \quad (1)$$

where n is an integer and f_{rep} is the repetition rate of the mode-locked laser. For the highest accuracy, the frequencies f_{ceo} and f_{rep} are stabilized to global positioning system (GPS) disciplined rf synthesizers or H-masers or some other high-precision low-noise frequency reference.

This stabilization represents another point in which the frequency combs are less robust than one might hope. For example, the repetition rate is

often stabilized by feeding back to the cavity length (although other feedback paths are sometimes used [17]). The frequency range over which the repetition rate can be stabilized is necessarily limited by the range of the piezo actuators. The carrier-envelope offset frequency is stabilized by feeding back to the pump power. However, the transfer function between pump power and f_{ceo} is not monotonic. Over some ranges of the pump power, it increases with pump power. Over others, it decreases [18].

For the highest-accuracy laser frequency measurements, these difficulties must be taken in stride. There simply is no other way to obtain the same accuracy. To be fair, these difficulties are small compared with those of the complicated frequency chains of just a few years ago [4]. In some applications, such as using cross-phase modulation with a well-characterized continuous wave (cw) laser, only the repetition rate needs to be stabilized [19]. Fiber laser frequency combs (FLFCs) tend to be more robust than Ti:sapphire-based combs. These have been successfully deployed in remote locations and rugged environments [12,20].

Yet there are many applications in which Ti:sapphire-based frequency combs can be used with much less difficulty. If somewhat lower accuracy is allowable, say, 10 kHz in the near infrared, then nearly all of the difficulties mentioned above can be avoided. In this paper, we describe a partially stabilized frequency comb-based spectroscopy method that can be used from 750 to 865 nm. We demonstrate the accuracy and versatility of this method by measuring transition frequencies in Rb, Cs, and Ca.

2. Experiment

We generate a frequency comb using a commercial 1 GHz repetition rate mode-locked femtosecond (fs) laser oscillator [21]. This passively mode-locked laser uses a six-mirror dispersion-compensated cavity, a thin Ti:sapphire gain crystal, and no other intracavity optical elements. The comb is pumped using 4 W from a frequency-doubled single-frequency Nd:YVO laser.

In our experiment, we measure three different RF frequencies. A schematic diagram of the laser

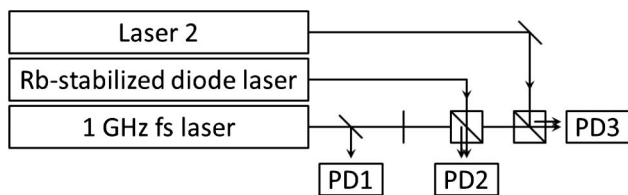


Fig. 1. Diagram of the laser measurement system. The 1 GHz repetition rate femtosecond (fs) laser generates our frequency comb. The frequency comb is offset-locked to the Rb-stabilized diode laser, providing excellent stability for one of the comb modes. The beat note between an additional laser, Laser 2, can be measured or locked to the frequency comb. PD1, PD2, and PD3 are all high-speed photodiodes used to measure RF beat-note frequencies. The experimental setup uses a number of amplifiers, which, for the sake of clarity, are not shown in this figure.

measurements is shown in Fig. 1. A beam splitter is used to direct a few percent of the fs laser beam power onto a fast photodiode (PD1 in Fig. 1). This photodiode measurement is bandpass filtered at 1 GHz, mixed down to 1.5 MHz, and counted with mHz precision, as sketched in Fig. 2. All of our counters and frequency synthesizers are referenced to a GPS-disciplined 10 MHz Rb oscillator.

A portion of the fs laser beam power is used for the Rb laser offset lock. We stabilize a diode laser to the $^{87}\text{Rb } F = 2 \rightarrow F' = 2/3$ crossover transition using saturated absorption. This 780 nm laser provides an absolute frequency reference for our frequency comb [22]. One mode of the comb is offset-locked to the diode laser using a microwave interferometer, as described in [23]. The output from the diode laser and a portion of the frequency comb are overlapped on a high-speed photodiode (PD2 in Fig. 1). This signal is filtered ($f_{\text{HPF}} > 50$ MHz and $f_{\text{LPF}} < 450$ MHz) and amplified and mixed down into the range of 5–30 MHz. This mixed-down RF signal is sent to a “microwave interferometer” (gray box in Fig. 2), comprised simply of a power splitter, two different lengths of coaxial cable, and a frequency mixer. Changing the interferometer input frequency by 40 MHz changes the output phase by 2π . The output of the interferometer mixer is low-pass filtered ($f_{\text{LPF}} < 2$ MHz) and used as an error signal for the offset lock.

This offset lock error signal feeds back to a piezoelectric transducer upon which one of the fs laser cavity mirrors is mounted. Applying voltage here changes the frequency of the beat note between the diode laser and the nearest comb mode. The

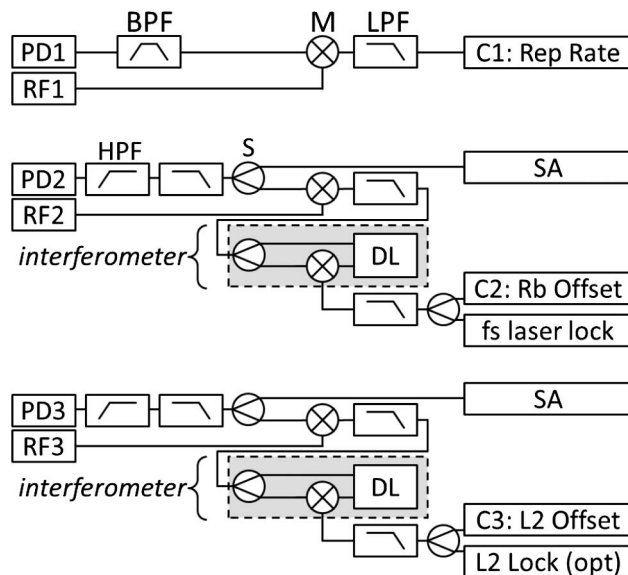


Fig. 2. Schematic diagram of the RF frequency measurements. The photodiodes PD1, PD2, and PD3 are those indicated in Fig. 1. BPF, bandpass filter; LPF, low-pass filter; M, frequency mixer; S, rf power splitter; SA, rf spectrum analyzer; DL, delay line. RF1, RF2, and RF3 are RF frequency synthesizers used to mix down the RF frequencies measured by the photodiodes to ranges that are more easily measured by the counters C1, C2, and C3.

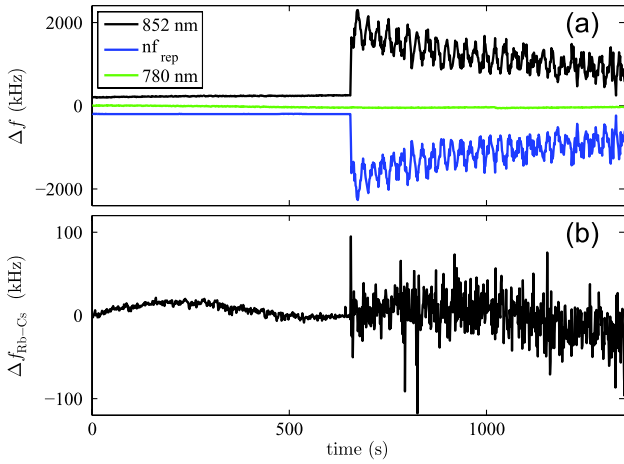


Fig. 3. (a) Changes in the measured beat notes and repetition rate. (b) Changes in the calculated frequency interval between the Cs D2 $F = 3 \rightarrow F' = 3/4$ crossover transition at 852 nm and the Rb D2 $F = 2 \rightarrow F' = 2/3$ crossover transition at 780 nm. Laser 2 is locked to the Cs transition. As discussed in the text, the repetition rate lock is turned off at $t = 650$ s, leaving the comb partially stabilized with only one comb mode actively locked to the Rb-stabilized diode laser. The slow drift in $\Delta f_{\text{Rb-Cs}}$ is due to a drift in the saturated absorption lock point. In this configuration, the frequency comb operates unattended for hours at a time. The counters are nearly synchronized, and the counter gate time is 1 s.

output of the interferometer gives a smooth error signal with very low phase noise. An integral-gain feedback circuit adjusts the fs laser cavity length so that the interferometer output is zero. This locks the offset frequency between the diode laser and the nearest comb mode to 26 MHz, which corresponds to one of the zero crossings of the interferometer output.

The microwave interferometer lock point drifts slowly. The interferometer phase is influenced primarily by temperature variations (a few kHz/ $^{\circ}\text{C}$), beat-note amplitude variations, and fluctuations in the DC offset voltage due to a variety of sources such as capacitive pickup. A frequency counter is used to monitor the beat note between the diode laser and the comb when the beat-note frequency is locked. The measured frequency variation is typically 1 kHz in a 1 s measurement interval, with larger variations over longer time scales.

A portion of the remaining fs laser power is used either to measure or control the frequency of a cw Ti:sapphire laser, indicated as Laser 2 in Fig. 1. A laser beam containing a few percent of the power from L2 and also from the fs laser are overlapped on PD3 (see Fig. 1). This signal is split, amplified, and mixed in a manner similar to the signal from PD2. Depending on the measurement (see below), Laser 2 can either be offset-locked to the comb or locked to a different atomic frequency reference, and the comb can be used to determine the laser's frequency.

The frequency of Laser 2 is then

$$f_{\text{laser}} = f_{\text{Rb}} \pm f_{\text{b2}} + nf_{\text{rep}} \pm f_{\text{b3}}, \quad (2)$$

where f_{Rb} is the transition frequency of the ^{87}Rb $F = 2 \rightarrow F' = 2/3$ crossover transition [22], $f_{\text{b2}} = f_{\text{RF2}} \pm f_{\text{C2}}$ is the beat note between the Rb-stabilized diode laser and the nearest frequency comb mode, $f_{\text{b3}} = f_{\text{RF3}} \pm f_{\text{L2}}$ is the beat note between Laser 2 and the nearest frequency comb mode, and n is an integer. By comparing the frequencies measured using the spectrum analyzers, the counters C2 and C3, and the RF synthesizers RF2 and RF3, we can determine the values of f_{b2} and f_{b3} . Changing the cavity length *before* the Rb offset-lock is engaged and monitoring how the beat notes change makes determining the plus or minus signs in Eq. (2) unambiguous.

Using this method, we don't have to lock either the repetition rate or the carrier-envelope offset frequency. Simply anchoring one of the comb modes and then measuring the repetition rate and beat notes gives us the necessary information to determine the frequencies of all the other comb modes [24]. As we show below, for integration times on the order of 10 s, the frequency uncertainty is in the 10 kHz range. In principle, if the frequency comb was stable enough, we would not need to stabilize anything. Simply counting the different beat notes would be sufficient [25]. However, the technical noise in our frequency comb gives unacceptably large uncertainties in the measured beat note frequencies.

Many studies reported in the literature have utilized fs laser-based frequency combs referenced to saturated absorption. It was used in the first demonstrations of optical frequency combs [2,26]. Some versions of direct frequency comb spectroscopy [27] have used this (see, for example, [28]). A careful comparison of comb-locking methods showed that referencing to saturated absorption in a vapor cell limits the ultimate accuracy of a comb [28]. However, if an accuracy on the order of 10 kHz is sufficient, the simplified method we describe can be extremely useful.

3. Results

For comparison purposes, we have inserted an AOM into the fs laser's pump beam. This makes it possible to lock the repetition rate by adjusting the pump power [17]. When the repetition rate is locked and one of the comb modes is offset-locked to the Rb diode laser, the frequency comb is fully stabilized [24].

We illustrate the performance of the comb by measuring frequency intervals between the Rb-stabilized diode laser and three other transitions: the Cs D2 $F = 3 \rightarrow F' = 3/4$ crossover transition at 852 nm, the ^{87}Rb D1 $F = 3 \rightarrow F' = 2$ transition at 794 nm, and the ^{40}Ca $4s^21S_0 \rightarrow 4s4p^1P_1^{\circ}$ transition at 423 nm.

A. Cs D2 Transition

In Fig. 3, we show the drift in our measured Cs D2 frequency. For this measurement, Laser 2 is locked to the Cs D2 $F = 3 \rightarrow F' = 3/4$ crossover transition at 852 nm using saturated absorption. For the first 650 s of the measured signal, the fs laser repetition rate is locked. After 650 s, we unlock the repetition

rate. As expected, the noise in the measured frequency interval increases [Fig. 4(b)].

In some sense, this data represents a “worst-case” scenario for our mode-locked laser because of the 70 nm interval between the lasers locked to saturated absorption in Rb and Cs. Small variations in the repetition rate are multiplied by a large mode number n [see Eq. (2)].

When the repetition rate is not locked, high-frequency noise in our frequency comb around 2.5 kHz increases the variability in the counted repetition rate. In addition, a somewhat large frequency excursion with a ~ 30 s period is displayed. This corresponds to the time constant of the temperature feedback loop in the water chiller used to control the fs laser baseplate temperature. We see the effect of the baseplate temperature fluctuations from time to time, depending on the environmental conditions in our lab. However, this variation is slow enough that frequency counting removes it from our measurement of the Rb-Cs frequency interval. Due to this thermal fluctuation, the repetition rate itself is changing by roughly ± 12 Hz ($df/f \sim 10^{-8}$) on a 30 s time scale. When this variation is multiplied by the mode number, $n \approx 33000$ for the Rb-Cs frequency interval, the repetition rate instability amounts to ± 400 kHz at 852 nm. Still, this large variation is accurately accounted for in the measured beat notes when the repetition rate is not locked. The fractional Allan deviation of the first 650 s of data in Fig. 3 falls below 4×10^{-12} at about 5 s of integration. For the unlocked data, the fractional Allan deviation falls below 20×10^{-12} at about 20 s.

B. Rb D1 Transition

In Fig. 4, we show the drift in our measured Rb D1 transition frequency. We lock a cw Ti:sapphire laser to the ^{85}Rb D1 $F = 3 \rightarrow F' = 2$ transition at 794 nm.

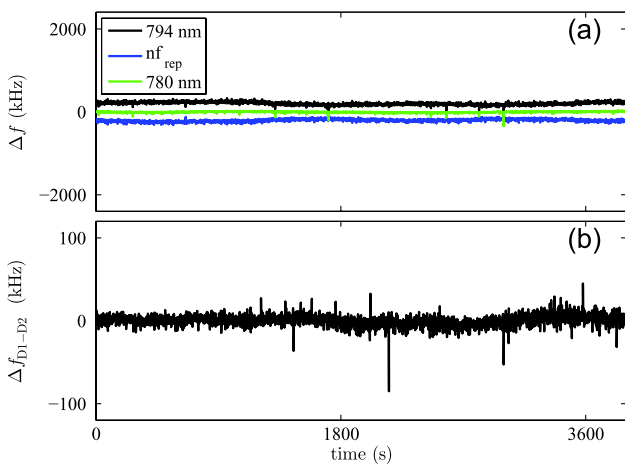


Fig. 4. (a) Changes in the measured beat notes and repetition rate. (b) Changes in the calculated frequency interval between the ^{85}Rb D1 $F = 3 \rightarrow F' = 2$ transition at 794 nm and the ^{87}Rb D2 $F = 2 \rightarrow F' = 2/3$ crossover transition at 780 nm. For this data, only the offset lock between one comb mode and the Rb-stabilized diode laser is engaged. The counter gate time is 1 s.

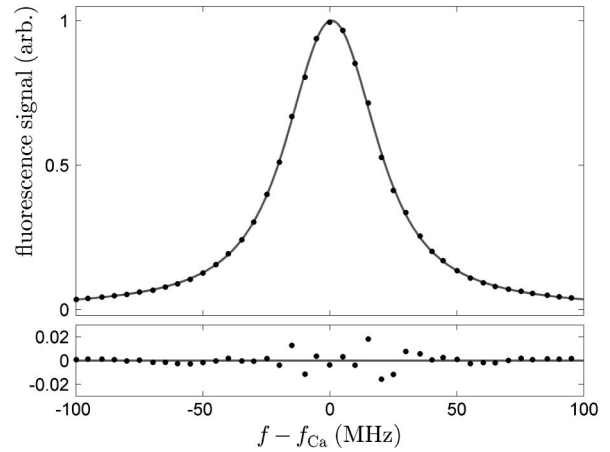


Fig. 5. Fluorescence signal collected from a collimated atomic calcium beam (top panel). A laser at 846 nm is offset-locked to the frequency comb. It is frequency doubled to 423 nm and used to excite the ^{40}Ca $4s^2\text{S}_0 \rightarrow 4s4p^1P_1^o$ transition. The black dots are the data; the gray line is a Voigt profile fit. The Lorentzian FWHM is 35.3 MHz; the Gaussian RMS width is 7.1 MHz. The bottom panel shows the residuals of the fit.

For these measurements, the repetition rate is not locked. The thermal loading visible in the previous figure is not present. Even if it were, the much smaller mode number corresponding to the Rb D1 to Rb D2 frequency interval $n \approx 7000$ would make it insignificant. The variation in repetition rate counting does not contribute appreciably to the uncertainty budget in the measured interval.

C. ^{40}Ca $4s^2\text{S}_0 \rightarrow 4s4p^1P_1^o$ Transition

In Fig. 5, we show the measured line shape of the ^{40}Ca $4s^2\text{S}_0 \rightarrow 4s4p^1P_1^o$ transition. In this case, Laser 2 at 846 nm is frequency doubled to 423 nm. The 423 nm laser beam is attenuated and aligned to be perpendicular to a collimated Ca atomic beam. The divergence of the atomic beam is measured to be less than 0.01 rad. The laser beam is retro-reflected back onto itself to minimize the transition frequency’s Doppler shift. Fluorescence from the atomic beam is collected using a fast optical system, measured using a photomultiplier tube, and digitized using an oscilloscope.

The laser at 846 nm is offset-locked to the frequency comb as described previously. However, by adjusting the frequency of the synthesizer RF3, we can adjust the frequency of Laser 2 with high precision [23]. The frequency of an RF synthesizer is changed over a ± 50 MHz interval, shifting the frequency of the 423 nm laser by ± 100 MHz. This method to scan the frequency of the 423 nm laser is facilitated by the 1 GHz wide mode spacing of the frequency comb. The mode spacing is wide enough that Laser 2 remains offset-locked to the same comb mode during the laser frequency scan. Each data point in Fig. 5 corresponds to a measurement time of 30 s. At each frequency, the fluorescence from the atomic beam is measured. In order to minimize the influence due to systematic drifts in

either laser beam or atomic beam intensity, the offset frequencies at which the fluorescence is measured are chosen at random from a predetermined list of frequencies.

Also shown in Fig. 5 is a fit to a Voigt line shape. The fitted Lorentzian width is 35.3 MHz, which is in good agreement with the experimental value of 35.2 (6) MHz [29]. The fitted Gaussian width is 7.1 MHz, which is in good agreement with the measured divergence of the atomic beam.

Comparing our measured value of the ^{40}Ca $4s^21S_0 \rightarrow 4s4p^1P_1^\circ$ transition frequency to the value of 709 078 373.01(35) MHz from [30] shows good agreement, with our value 0.82 MHz higher. Our measurement cannot be considered definitive because the laboratory magnetic fields are not canceled. This could lead to a frequency error as large as 0.4 MHz. In addition, errors related to the lock point offset in our Rb diode laser are on the 0.2 MHz level. We are undertaking to eliminate both of these errors at the present time.

4. Conclusion

In this paper, we have demonstrated the performance of a partially stabilized frequency comb. In principle, the entire comb could be used in a completely unstabilized mode. The beat notes and repetition rate could all be measured, and frequency intervals could be thereby computed. Unfortunately, the passive stability of our lasers is not good enough for this to be desirable. Partially stabilizing the comb dramatically reduces the main noise components, which makes it possible for us to measure and stabilize lasers with ~ 10 kHz uncertainty with less than 30 s of averaging. We have shown how to lock and to continuously scan lasers over a few hundred MHz with this level of uncertainty. The unbroadened frequency comb is wide enough that this method can be used to control, measure, and scan lasers with wavelengths ranging from 750 to over 865 nm.

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