

# Direct fiber comb stabilization to a gas-filled hollow-core photonic crystal fiber

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**Abstract:** We have isolated a single tooth from a fiber laser-based optical frequency comb for nonlinear spectroscopy and thereby directly referenced the comb. An 89 MHz erbium fiber laser frequency comb is directly stabilized to the P(23) (1539.43 nm) overtone transition of <sup>12</sup>C<sub>2</sub>H<sub>2</sub> inside a hollow-core photonic crystal fiber. To do this, a single comb tooth is isolated and amplified from 20 nW to 40 mW with sufficient fidelity to perform saturated absorption spectroscopy. The fractional stability of the comb, ~7 nm away from the stabilized tooth, is shown to be  $6 \times 10^{-12}$  at 100 ms gate time, which is over an order of magnitude better than that of a comb referenced to a GPS-disciplined Rb oscillator.

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**OCIS codes:** (300.6460) Spectroscopy, saturation; (120.3930) Metrological instrumentation.

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## 1. Introduction

Optically referenced combs demonstrate superior short-term instability [1] limited by that of the optical reference. Typically, optically referenced frequency combs require a stable cw laser locked to an optical cavity and/or an atomic or molecular reference. However, direct comb spectroscopy eliminates the need for a cw laser, which is advantageous for portable, practical systems, for which fiber-laser-based combs are well-suited [2, 3]. Hu *et al.* [4] have directly optically referenced an Er fiber laser-based frequency comb to a Rb cell by generating its second harmonic and stabilizing to the two-photon transition in Rb. Heinecke *et al.* [5] have demonstrated an optically-referenced Ti:sapphire comb by stabilizing a single tooth to a Rb transition using saturated absorption spectroscopy (SAS). However, application of this technique to a low repetition rate fiber frequency comb in the near IR offers unique challenges. In particular, sufficient power per tooth of the comb must be obtained, either by increasing the repetition rate of the laser or by amplifying the frequency comb, or both.

Fiber lasers with repetition rates of 1-10 GHz have been created [6–10], but are difficult to fully stabilize. In 2012, Chao *et al.* for the first time successfully phase stabilized a ~1 GHz erbium fiber laser [11] based on a linear cavity. However, this laser required extreme amplification to generate supercontinuum (SC) for self-referencing. Delfyett's group [8, 9] built a 10 GHz harmonically mode-locked fiber laser with a Fabry-Perot etalon integrated in the laser cavity, and measured its carrier-envelope offset frequency,  $f_0$ . But the harmonic mode-locking mechanism may limit the instability, and the laser has insufficient power for  $f_0$  generation. Injection locking of a mode-locked fiber laser has allowed creation of a fiber laser-based frequency comb with up to 1 GHz repetition rate with ~5.5 nm spectral bandwidth [12]. And recent advances in microresonator-based combs make them a promising option for the future [13].

An alternative way to increase the repetition rate is to use an external filtering cavity. High repetition rate, broad bandwidth fiber combs are generated in this way for astronomical

calibration purposes [14, 15]. In such configurations, one of the major challenges is to optically amplify the comb without degrading the comb signal-to-noise ratio (SNR).

Our approach to single-tooth amplification builds on earlier work. Cruz *et al.* have shown an optical amplification of supercontinuum by a factor of 17 dB using a home-made semiconductor amplifier [16]. Moon *et al.* performed spectroscopy in Rb vapor by injection locking a cw laser to a phase stabilized single comb tooth [17]. In the early 1990s, the telecom industry spurred amplification of small cw light power with high gain and low noise figure [18, 19] by employing multiple small gain amplification stages to achieve high gain while preserving the SNR.

Here, as discussed in Section 2, we optically amplify a single comb tooth from 20 nW to 40 mW (by a factor of  $2 \times 10^6$ ) while preserving the comb's SNR using multiple filtering and amplification stages. During this process, an external filtering cavity is built to increase the pulse repetition rate from 89 MHz to 9.4 GHz. A distributed-feedback (DFB) laser is injection-locked to a 50  $\mu$ W single tooth out from the filtering cavity. No shift of the comb frequency is detected at the 10 kHz level during amplification. In Section 3, the amplified single tooth is then frequency-stabilized to the P (23)  $v_1 + v_3$  overtone transition of  $^{12}\text{C}_2\text{H}_2$  at  $\sim 1539.43$  nm through sub-Doppler spectroscopy inside a gas-filled hollow-core photonic crystal fiber [20, 21]. With  $f_0$  stabilized to an RF reference simultaneously, a fully phase stabilized, optically-referenced erbium fiber laser frequency comb is demonstrated. In Section 4, the comb stability is characterized through comparison with a cw laser locked to a gas-filled hollow fiber reference [21]. Section 5 outlines various ways the comb could be stabilized to the optical reference, and the resulting stability expected across the comb optical spectrum.

We believe this to be the first demonstration of an optically referenced comb based on a fiber laser using direct-comb saturated absorption spectroscopy. Furthermore, this work represents the first isolation and amplification of a single tooth directly from a sub-100 MHz fiber laser for saturated absorption spectroscopy. This comb amplification technique opens many possibilities for direct comb spectroscopy using low power, low repetition rate ( $< 100$  MHz) fiber laser frequency combs. In addition, the fiber comb optically referenced to a gas-filled fiber reference represents a large step towards an all-fiber portable frequency metrology system with low short-term instability, independent of the global positioning system (GPS).

## 2. Comb tooth amplification for sub-Doppler spectroscopy

The overview setup for this work is shown in Fig. 1. Section 2.1 presents the 89 MHz fiber ring laser built for sub-Doppler spectroscopy. The majority (90%) of power from the fiber ring laser output goes for  $f_0$  detection and stabilization at  $\sim 30$  MHz via diode laser pump current, based on a collinear  $f$ - $2f$  interferometer [22]. The rest of the output is used for single comb tooth amplification, including multiple filtering and amplification stages (middle dashed box in Fig. 1), which are discussed in Section 2.2-2.4. A small tap from the ring oscillator is used for monitoring the comb's repetition rate.

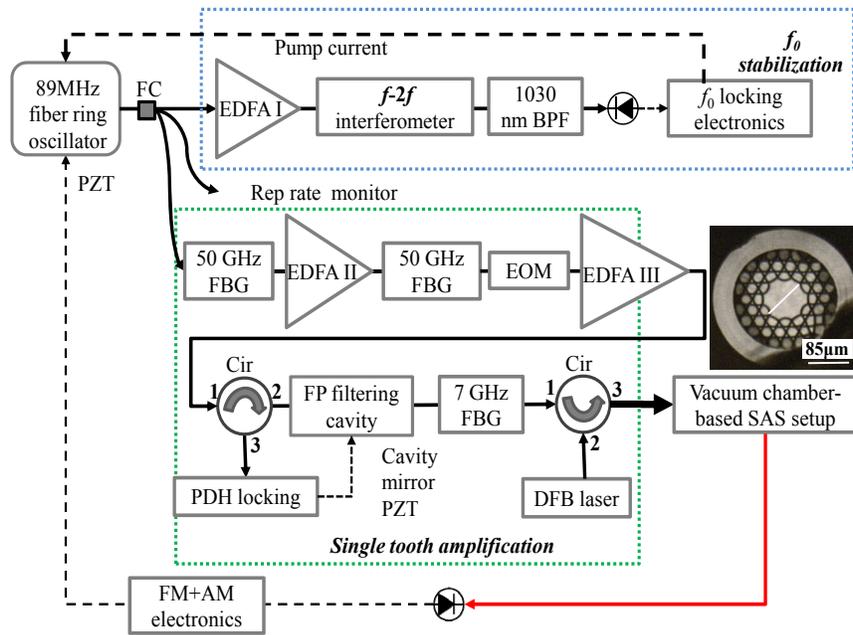


Fig. 1. Overview schematic of single tooth saturated absorption spectroscopy. FC: fiber coupler, EDFA: Er-doped fiber amplifier, BPF: band pass filter, FBG: fiber Bragg grating, EOM: electro-optic modulator, Cir: circulator, PDH: Pound-Drever-Hall, FP: Fabry-Perot, AOM: acousto-optic modulator, and AM: amplitude modulator. Solid lines indicate optical paths, and dashed lines indicate electrical paths.

### 2.1 The fiber ring laser

The schematic setup of the all-fiber ring laser is shown in Fig. 2 (left). The 89 MHz erbium-doped fiber ring oscillator is mode-locked using the nonlinear polarization rotation technique [23]. The gain fiber is a 64 cm erbium-doped fiber (Liekki Er110), with an absorption coefficient of 110 dB/nm at 1530 nm, pumped by a 300 mW diode laser at 980 nm. All other fibers in the cavity are standard single-mode fiber (SMF). The laser is mode-locked in the stretched pulse regime. The output spectrum has a 3-dB bandwidth of 30 nm (Fig. 2 right), with a total output power of 3.8 mW.

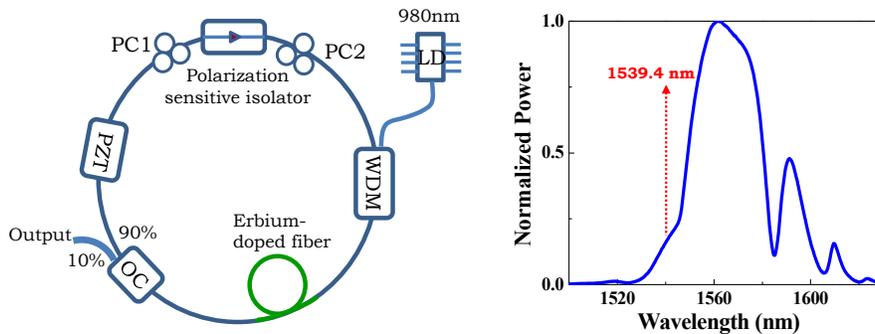


Fig. 2. Schematic setup and optical spectrum of an 89 MHz erbium-doped fiber ring laser for single tooth saturation spectroscopy. The red arrow in the spectrum indicates the wavelength of the single comb tooth to be amplified.

## 2.2 Comb amplification

Our initial study of comb amplification is done at 1532.83 nm. The output from the oscillator is first filtered by a 50 GHz fiber Bragg grating (FBG) and amplified by a home-made erbium fiber amplifier (EDFA II in Fig. 1), then filtered again at the same wavelength before entering a commercial polarization maintaining cw erbium-doped fiber amplifier (EDFA III in Fig. 1). The purpose of EDFA II is to generate enough light at the target wavelength to seed the following EDFA III. This EDFA II proves to be the key to preserving the comb SNR.

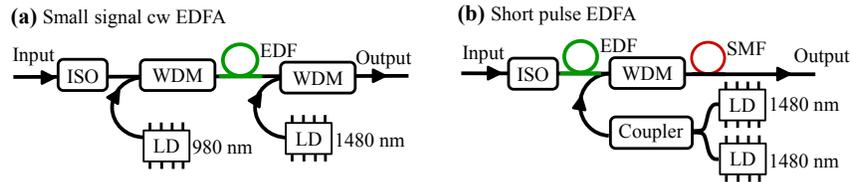


Fig. 3. Schematic setup up of (a) small signal cw EDFA, and (b) short pulse EDFA.

Figure 3(a) shows the schematic of EDFA II, which is a small signal cw EDFA [18, 19]. A 1 m length of EDF (EDF 80) with an absorption coefficient of 80 dB at 1530 nm is used as the gain fiber. The key point to this design is that a narrow bandwidth comb (~50 GHz) around our wavelength of interest is filtered from the oscillator output and seeded into this amplifier. The amplifier is mainly forward-pumped by a 200 mW pump diode at 980 nm, and slightly backward pumped by a 40 mW laser diode at 1480 nm. The small signal EDFA has 38 dB gain at 1532.83 nm when seeded with 1  $\mu$ W cw light, and 14 dB gain when seeded with 1 mW cw light.

We initially built a short pulse EDFA, shown in Fig. 3(b), which was not successful. The main reason is that it introduced devastating amplified spontaneous emission (ASE) noise that destroyed the comb's SNR. It was designed to be seeded directly by the oscillator comb, and to broaden the comb spectrum to have enough light at the target wavelength. The gain fiber is highly backward pumped with a total pump power of 1 W. The amplified pulses are further temporally compressed by the following SMF fiber via solitonic effects.

To characterize the comb's SNR before and after amplification, a heterodyne beat between the comb and a cw fiber laser with a much narrower linewidth (< 1 kHz) is analyzed by an electrical spectrum analyzer (HP 8561B), shown in Fig. 4. Before amplification, the beatnote has a SNR of 20 dB (mostly limited by comb power) at 1 MHz resolution bandwidth (RBW). Using the same amount of optical power for both the comb and cw laser, this SNR is well preserved after amplification by the small signal cw EDFA. However, when the short pulse EDFA is used instead, the SNR is dramatically reduced to 8 dB.

To further study how the two EDFAs behave in terms of noise and signal amplification, we measured the electronic noise floor and the SNR of the cw/comb beatnote in three scenarios, shown in Fig. 4(d). The cw fiber laser power was kept the same for all beat measurements, while the comb power was controlled by variable attenuation. The noise floor behaves like technical noise on the amplified comb. Figure 4 indicates that the small signal EDFA produces an amplified comb with a lower noise floor and higher SNR as compared to the short pulse EDFA. The degradation in SNR after the short pulse amplifier may be due to fiber nonlinearities.

Considering that there are multiple power amplification stages based on EDFAs in our setup, and also the fact that the amplified wavelength is at the edge of the erbium-doped fiber gain spectrum, we investigated the possible frequency shift introduced by all the EDFAs when seeded with the low power comb. Specifically, we measured the heterodyne beat between the pre and post amplified cw light or comb tooth. Our results show that for a cw seeding power of 100 nW and above, no obvious frequency shift is observed within 100 Hz. For comb seeds, the upper limit on the shift is 10 kHz due to low beatnote SNR.

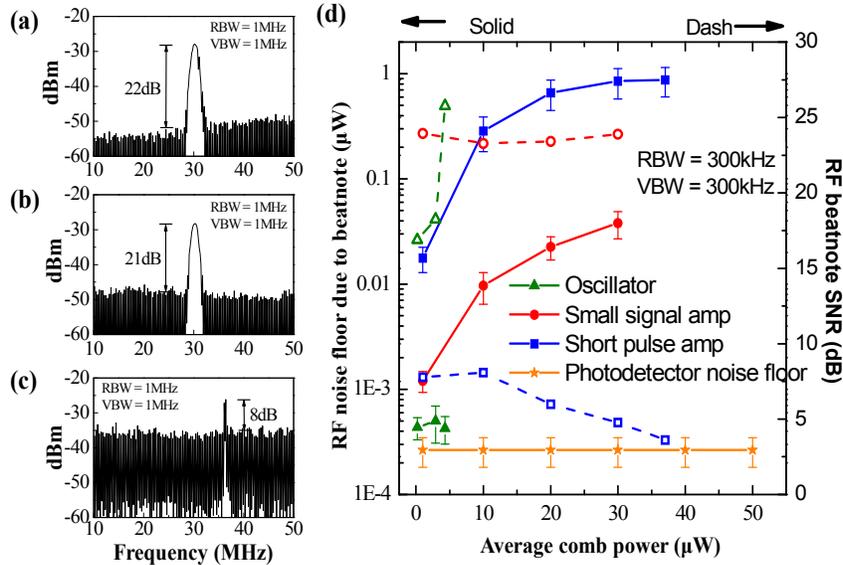


Fig. 4. RF beatnote between the cw fiber laser and (a) the oscillator comb, (b) the comb after amplification by the small signal cw EDFA, and (c) the comb after amplification by the short pulse EDFA. (d) RF noise floor for the above three beatnotes (left solid), and RF beatnote SNR (right dashed) as a function of average comb power. All measurements are taken under 300 kHz RBW.

### 2.3 Selection of a single tooth (filtering cavity)

A single comb tooth is selected using a free space Fabry-Perot filtering cavity followed by a narrow bandwidth fiber Bragg grating (FBG). The filtering cavity length must be adjusted such that each cavity resonance coincides with a comb tooth frequency. Therefore, the cavity length is stabilized to multiple comb teeth simultaneously, such that the cavity free spectral range (FSR) is an integer multiple of the comb repetition rate. In our case, one comb tooth is selected out of  $\sim 105$  teeth. Before passing through the cavity, a 50 GHz narrow bandwidth comb is amplified by a commercial cw polarization-maintaining EDFA (by Manlight, EDFA III in Fig. 1) to an average power of 200 mW.

The cavity consists of two nearly plano dielectric mirrors (with 50 cm radius of curvature) separated by 1.6 cm. One of the mirrors is attached to a ring PZT (Nolia) for cavity length stabilization. A collimated beam with 0.5 mm  $1/e^2$  diameter is focused by a 200 mm lens to ensure required mode matching with the cavity mirror curvature. The cavity has a finesse of about 250, and a suppression ratio of at least 21 dB as measured by a heterodyne beatnote between the filtered comb and a cw fiber laser. When the cavity is not resonant with the laser, light reflected from the first cavity mirror will be rejected back to the third port of the fiber circulator before the cavity, used for Pound-Drever-Hall locking, which will be discussed in more detail in Section 3. The filtered 9.4 GHz comb from the cavity output is coupled back into the fiber and further spectrally filtered by a customized FBG, with 3-dB bandwidth of 7 GHz, to obtain a single comb tooth. The FBG is under precise temperature control to have a stable central transmission wavelength at  $\sim 1539.4$  nm. The single comb tooth after the FBG has an optical average power of  $\sim 50$   $\mu$ W, which is sufficient for injection locking the DFB laser in the last amplification stage (Section 2.4).

### 2.4 Injection locking DFB laser

The last comb amplification step is accomplished by seeding the single comb tooth into a distributed-feedback (DFB) laser diode. This allows stable injection locking of the DFB laser

output to the seeding comb tooth. The DFB laser is a customized 14-pin butterfly pump laser diode (FITEL) without built-in isolator. The key to a stable injection lock is to seed the DFB laser with only one comb tooth. This was verified by seeding the DFB laser with two identical cw fiber lasers at 1532.8 nm with equal optical power to mimic the case when two comb teeth are injected into the DFB laser. Our results show that even when the two cw lasers are a few GHz apart, the DFB laser injection locks to each of the two seeds at different times, indicating mode competition between the seeds. This determines that the FSR of the filtering cavity has to be larger than the bandwidth of the following 7 GHz FBG.

For stable injection locking, the DFB laser diode is temperature-controlled at 32.92 °C, pumped by a current of 170 mA, and amplifies the injected single tooth to be 40 mW at 1539.43 nm. We obtained a stable RF beatnote between the amplified single tooth using a similar DFB laser at 1532.83 nm and cw fiber laser. The beatnote SNR is enhanced to be ~40 dB at 1 MHz RBW.

### 2.5 Single tooth saturated absorption spectroscopy

A 40 mW comb tooth is then directed to the saturated absorption spectroscopy (SAS) setup to generate a sub-Doppler error signal, as shown in the lower (green) dashed box in Fig. 1. Details of the setup can be found in [21]. Acetylene gas is loaded inside a 7.9 m length of hypocycloid shaped core-contour kagome-structured hollow-core photonic crystal fiber, with a core size of 85  $\mu\text{m}$  [20, 24] and its pressure was set at 140 mtorr (18.7 Pa).

## 3. Frequency comb stabilization

To fully stabilize the comb, there are three servo loops, shown in Fig. 1, that lock the carrier offset frequency  $f_0$  to an RF synthesizer referenced to a GPS-disciplined Rb oscillator (GPS-Rb), stabilize the filtering cavity to the comb for proper filtering of every one out of ~105 comb teeth, and frequency stabilize the amplified single comb tooth to the P (23) overtone transition of  $^{12}\text{C}_2\text{H}_2$  at 1539.43 nm. The technique employed for  $f_0$  locking is described in [22]. Here,  $f_0$  can be locked within a few Hz for over ten hours. This section focuses on the last two servo loops. A GPS-Rb oscillator serves as the external reference for all synthesizers and frequency counters.

The filtering cavity is stabilized to a particular comb tooth using the Pound-Drever-Hall (PDH) technique [25]. Due to limited tunability of the fiber laser-based comb, this particular tooth of the 89 MHz comb has to be chosen as close to the P (23) line as possible. To select that tooth, a cw diode laser (Santec TSL-210) is stabilized to the P (23) transition as a reference laser and beat against the filtering cavity output. The cavity length is scanned to be resonant with a number of different comb teeth, until the optical heterodyne beat is close to zero frequency. For PDH locking, the comb is modulated at 30 MHz by using an electro-optic modulator. The PDH error signal generated by the comb has a SNR of about 20, a factor of 6 lower than that generated by a cw fiber laser as the source because most detected comb teeth do not contribute to the error signal. However, using a homemade servo box, we are still able to lock the cavity for hours.

The error signal for locking the single tooth to the P (23) overtone transition is similar to that of a cw fiber laser, which can be found in [21]. Both sub-Doppler error signals have SNR's above 100 within 60 kHz bandwidth, shown in Fig. 5. This sub-Doppler error signal is fed back to the fiber ring laser PZT for precise control of  $f_{\text{rep}}$ .

The amplified comb tooth would be more wavelength-tunable if the injection-locked DFB laser were replaced with an EDFA. We investigated this approach using a commercial cw EDFA and observed an error signal but with much worse SNR of only 4. This SNR degradation is likely due to non-uniform power amplification of the suppressed teeth from the cavity as revealed on an RF spectrum analyzer by increased power at the 89 MHz repetition rate. Comparing with this EDFA technique, the amplification technique using injection locking is superior because the small gain bandwidth of the DFB laser and the minimum

power threshold for stable locking ensure that only a single comb tooth can be amplified. In the end, the error signal shown in Fig. 5(a), generated from injection locking the DFB laser, is used to stabilize a comb tooth at 1539.43 nm to the P(23) overtone transition of acetylene. With the offset frequency  $f_0$  locked to an RF reference and one comb tooth locked to an acetylene transition, both degrees of freedom of the comb are stabilized. The system can stay locked for tens of hours.

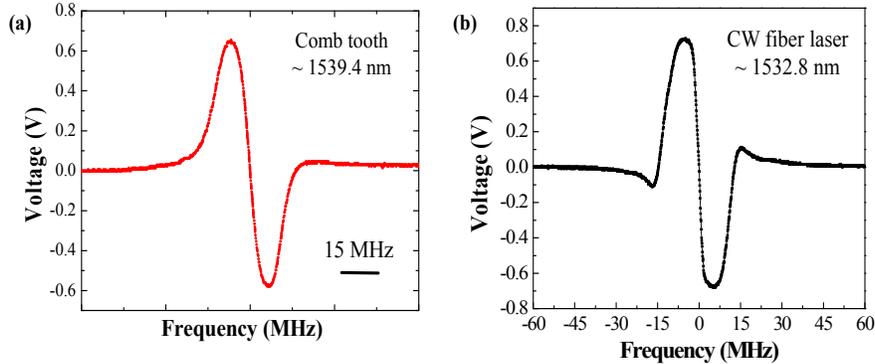


Fig. 5. The sub-Doppler error signal generated from (a) an amplified single tooth, and (b) a cw fiber laser.

#### 4. Optical instability measurement

Once the comb is stabilized as described in Section 3, the comb stability is characterized by a comparison with a cw reference at 1532.83 nm, which is  $\sim 7$  nm away from the stabilized single tooth, shown in Fig. 6(a). This cw reference is a cw fiber laser locked to the P(13) line of  $^{12}\text{C}_2\text{H}_2$  inside gas-filled hollow-core fiber [21] using the same SAS technique as the one described for the single tooth locking. The error signal for locking is plotted in Fig. 5(b). The stabilized comb is optically combined with the cw reference, resulting in an RF beatnote.

The fractional instability (Allan deviation) of the optical tooth at 1532.83 nm is calculated from the RF beat, as show in Fig. 6(b) (red dots). Since the fluctuation of the measured RF signal is divided by the optical frequency in THz, this result is not limited by the GPS-Rb-referenced counter (black stars). It has short time stability of  $6 \times 10^{-12}$  at 100ms gate time, which is over an order of magnitude better than the GPS-Rb oscillator (black stars). The stability of the cw reference used for comparison was extrapolated from two previous measurements. One is the heterodyne beating of two identical cw fiber references in our lab [26], shown in blue squares, which is independent of the GPS limit. The other is the beat between one cw fiber reference and a carbon nanotube fiber laser-based frequency comb [22] referenced to the GPS-Rb oscillator, originally published in [21]; the original data set has been reanalyzed to correct an error and shows improved stability at 1000 s (green diamonds). This improvement is important because it indicates the long-term stability of the cw fiber reference, which can also be expected from this fiber-referenced comb system.

Because the stability of the optically referenced comb is comparable to that of the cw reference at short time scales, the stability of our optical reference at 1539.43 nm has been transferred to the measured tooth at 1532.83 nm. The beatnote shows a slow drift at a time scale of a few hours, which correlates with drift in the repetition rate. This slow drift is independent of leakage of vacuum chambers, temperature or humidity, and is likely due to offsets in the locking electronics. We believe with improved servo electronics, the system is expected to have comparable long-term stability as the single cw reference data (green diamonds) in Fig. 6(b).

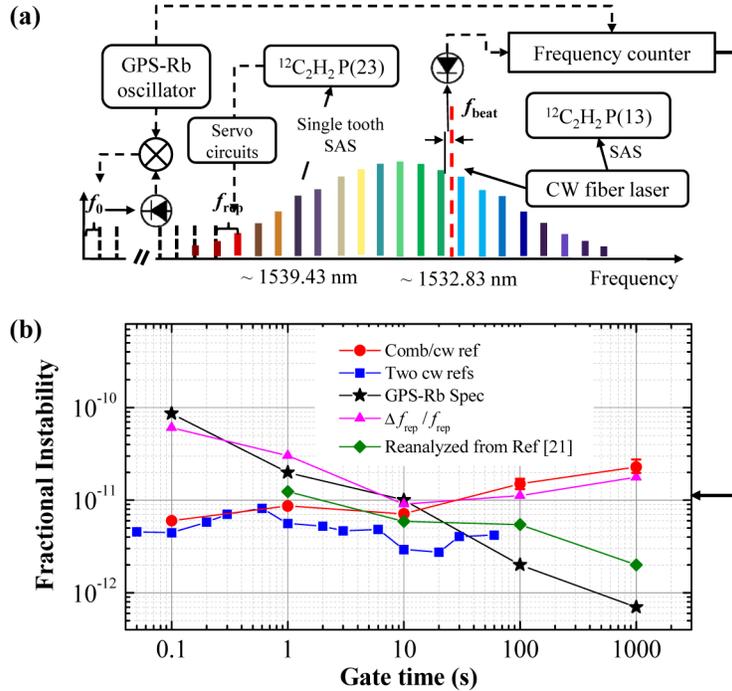


Fig. 6. (a) Schematic setup for the optically referenced comb stability measurement. (b) Fractional instabilities (Allan deviations) of: (red dots) the comb at 1532.8 nm compared to a cw fiber laser reference; (blue squares) the cw reference at 1532.8 nm extrapolated from two identical reference heterodyne beat measurement; (black stars) GPS-disciplined Rb oscillator from spec sheet; (pink triangles) comb's repetition rate frequency recorded by a GPS/Rb-referenced frequency counter; (green diamonds) reanalyzed data for Fig. 3(c) in [21]. Error bar on the red-dot line represents  $1\sigma$  confidence intervals.

The instability of the RF repetition rate from the same measurement, shown in pink triangles in Fig. 6(b), is expected to follow the comb instability shown in red dots. However, the measurement is limited by the performance of the GPS-Rb oscillator at short time scales. This is because all the frequency counters used in the experiment are referenced to GPS-Rb. For a real measurement of  $f_{rep}$  stability below the GPS-Rb limit, we would have to use a better RF reference, for example, a hydrogen maser. The accuracy of this comb reference is expected to be comparable to the cw reference in [21] of  $\pm 10$  kHz.

For comparison, the frequency comb of Hu *et al.* [4] is referenced to a Rb vapor cell by stabilizing a single tooth to the two-photon transition at 778 nm and  $f_{rep}$  to an electromagnetically induced transparency resonance at 3 GHz; the microwave reference limits the optical fractional instability to be about  $10^{-10}$  at 1s, and  $10^{-11}$  at 1000 s. Stability similar to that of Fig. 6(b) is achieved in the direct comb spectroscopy demonstrated by Heinecke *et al.* [5] based on an optically-referenced 10 GHz Ti:sapphire comb. There, a single tooth is stabilized to a Rb transition using sub-Doppler spectroscopy while the comb repetition rate is locked to a hydrogen maser. The stability of a comb tooth 20 nm away from the Rb resonance is measured to be  $7 \times 10^{-12}$  at 1 s gate time.

## 5. Comb frequency instability calculation for different locking schemes

There are many ways to stabilize a frequency comb using RF and optical references. Reference [5] describes the relative instabilities using various reference schemes. Here we specially calculate the comb stability using our gas-filled hollow-core fiber reference and RF references of various stabilities, thereby showing that good comb performance can be

achieved using either the gas-filled hollow-core fiber reference with modest RF (scheme 2 below), two optical references (scheme 3), or no RF references (scheme 4). We estimate the comb stability for four different locking schemes: scheme 1,  $f_0$  and  $f_{\text{rep}}$  locked to an RF reference; 2,  $f_0$  locked to RF while a single comb tooth was locked to an optical reference; 3,  $f_{\text{rep}}$  locked to RF and a single comb tooth to optical reference; and 4, two comb teeth locked to two optical references. The comb instability calculations for these four locking schemes are summarized in Table 1. The degradation of the comb stability is investigated when different RF references are used, as we look 100 nm and 500 nm away from 1539.4 nm, shown in the last two columns of Table 1. In this calculation, we used the following fiber comb parameters: repetition rate is 90 MHz,  $f_0$  is 30 MHz, and one stabilized comb tooth is at the reference wavelength  $\lambda_{r1} = c/f_{r1}^{(\text{opt})} = 1539.4$  nm (case 2-4). For the optical reference, we assume the fractional instability at 100 ms is  $10^{-12}$  as measured in our experiment.

**Table 1. Calculation of Comb Optical Instability at 100 ms within 100 and 500 nm of 1539 nm When Various Locking Schemes and RF and Optical References Are Employed**

	Locking parameters	$\delta f_{\text{RF}}/f_{\text{RF}}$		$\delta f_{\text{opt}}/f_{\text{opt}}$	$\lambda_{r1}$ (nm)	$\Delta\lambda_r^a$ (nm)	Comb tooth instability about 1539.4 nm	
							$\pm 100$ nm	$\pm 500$ nm
(1)	$f_0^{(\text{RF})} + f_{\text{rep}}^{(\text{RF})}$	Quartz	$1 \times 10^{-9}$	—	—	—	$1 \times 10^{-9}$	$1 \times 10^{-9}$
		GPS-Rb	$2 \times 10^{-11}$				$2 \times 10^{-11}$	$2 \times 10^{-11}$
		H maser	$1 \times 10^{-13}$				$1 \times 10^{-13}$	$1 \times 10^{-13}$
(2)	$f_0^{(\text{RF})} + f_{r1}^{(\text{opt})}$	Quartz	$1 \times 10^{-9}$	$1 \times 10^{-12}$	1539.4	—	$1 \times 10^{-12}$	$1 \times 10^{-12}$
		GPS-Rb	$2 \times 10^{-11}$				$1 \times 10^{-12}$	$1 \times 10^{-12}$
		H maser	$1 \times 10^{-13}$				$1 \times 10^{-12}$	$1 \times 10^{-12}$
(3)	$f_{\text{rep}}^{(\text{RF})} + f_{r1}^{(\text{opt})}$	Quartz	$1 \times 10^{-9}$	$1 \times 10^{-12}$	1539.4	—	$7 \times 10^{-11}$	$3 \times 10^{-10}$
		GPS-Rb	$2 \times 10^{-11}$				$2 \times 10^{-12}$	$7 \times 10^{-12}$
		H maser	$1 \times 10^{-13}$				$9 \times 10^{-13}$	$7 \times 10^{-13}$
(4)	$f_{r1}^{(\text{opt})} + f_{r2}^{(\text{opt})}$	—	—	$1 \times 10^{-12}$	1539.4	3 nm	$7 \times 10^{-11}$	$3 \times 10^{-10}$
						30 nm	$5 \times 10^{-12}$	$3 \times 10^{-11}$
						300 nm	$2 \times 10^{-12}$	$3 \times 10^{-12}$

<sup>a</sup> wavelength separation between two stabilized comb teeth  $\Delta\lambda_r = \lambda_{r1} - \lambda_{r2}$

In scheme 1, the instability of the  $m^{\text{th}}$  comb tooth can be expressed as Eq. (11) of [5] in terms of the instability of  $f_0$  and  $f_{\text{rep}}$ . The microwave instability of  $f_{\text{rep}}$  is transferred to the optical regime ( $\nu_m$ ) due to the large  $m$  number  $\sim 10^6$  for a sub-100 MHz fiber laser. In this case, the  $f_0$  fractional instability is essentially irrelevant. Within  $\pm 500$  nm around the stabilized comb tooth, the comb instability is dominated by the RF instability of the repetition rate.

In scheme 2, if  $f_0$  is locked to an RF reference while one comb tooth is locked to an optical reference, the stability of the  $m^{\text{th}}$  comb tooth can be expressed as:

$$\sigma_{\nu_m} = \frac{\delta\nu_m}{\nu_m} = \left(1 - \frac{m}{n_r}\right) \frac{\delta f_0}{\nu_m} + \frac{m}{n_r} \frac{\delta\nu_r}{\nu_m} \quad (1)$$

where  $\nu_r$  is the optical frequency of the stabilized comb tooth and  $n_r$  is the mode number of that tooth. In this equation, the first term is the uncertainty induced by the  $f_0$  instability. As

shown in Table 1, the  $f_0$  instability ( $\delta f_0$ ) is irrelevant for the resulting comb instability, and the comb instability is dominated by the second term, which depends on two parameters: the instability of the comb tooth locked to the optical reference ( $\delta v_r$ ), and how far the comb tooth of interest is away from the stabilized tooth. Based to our calculation, within  $\pm 500$  nm from the stabilized comb tooth at 1539.43 nm, the uncertainty of the stabilized optical tooth is still the dominant factor.

Scheme 3 involves  $f_{\text{rep}}$  locked to an RF reference while one comb tooth is locked to an optical reference. An advantage of this locking scheme is that the generation of carrier-envelope offset frequency  $f_0$  can be avoided, which reduces the system complexity. However, a high performance RF reference is required such that the multiplied RF instability does not dominate. The instability of the comb tooth of interest can be expressed by Eq. (5) of [5] in terms of the fractional instability of the repetition rate and the locked single tooth. This calculation shows that the optical instability depends critically on the employed RF reference.

In scheme 4, two comb teeth are stabilized to two separate optical references at wavelengths  $\lambda_{r1}$  and  $\lambda_{r2}$ , which can be two transitions from the same or different types of atoms/molecules, or high finesse optical cavities. By using gas-filled photonic microcells [26] as the optical references, the system can likely be made simpler and more portable without the integration of RF references. For simplicity, we assume that both feedback loops are identical and independent from each other. Therefore, the stability of the  $m^{\text{th}}$  comb tooth can be expressed as:

$$\sigma_{v_m} = \frac{\delta v_m}{v_m} = \frac{m - n_{r2}}{n_{r1} - n_{r2}} \times \frac{v_{r1}}{v_m} \times \frac{\delta v_{r1}}{v_{r1}} + \frac{n_{r1} - m}{n_{r1} - n_{r2}} \times \frac{v_{r2}}{v_m} \times \frac{\delta v_{r2}}{v_{r2}} \quad (2)$$

In this case, besides the instability of the two stabilized comb teeth ( $\delta v_{r1}/v_{r1}$  and  $\delta v_{r2}/v_{r2}$ , respectively), another key parameter that determines the overall comb instability is the wavelength separation between the two stabilized comb teeth. As an example, Table 1 shows the comb instability when two stabilized comb teeth are separated by 3 nm, 30 nm and 300 nm. If the two locked teeth are separated by only 3 nm, the comb stability degrades by almost two orders of magnitude 500 nm away from the locked tooth at 1539.4 nm. In contrast, if the separation is 300 nm, the degradation is only a factor of three even for comb teeth almost 500 nm away from one of the locked teeth. Therefore, the two stabilized comb teeth need to be at least hundreds of nanometers apart to avoid fast degradation in comb stability, which may be achievable with 2 separate gas-filled fiber references based on different gasses. This result is consistent with the conclusions obtained in [5].

## 6. Summary

We have demonstrated the first direct sub-Doppler spectroscopy with a single tooth from an optically-referenced fiber comb. To do this, a single comb tooth was amplified from 20 nW to 40 mW with high fidelity, sufficient to perform saturated absorption spectroscopy on an overtone transition in acetylene near 1540 nm directly with the amplified comb tooth. No intermediate cw laser was required, nor did a cw laser need to be phase-locked to the comb.

The resulting optical frequency comb exhibits high short-term stability ( $6 \times 10^{-12}$  at 100 ms) exceeding that of the GPS-disciplined Rb oscillator by an order of magnitude; thus the stability of the comb is equal to that of a cw fiber laser locked to the reference. Long-term drift is attributed to technical noise and should be readily reduced to the level of a cw laser locked to the fiber reference (shown in green diamonds in Fig. 6(b), the corrected data from [21]). Calculations indicate that  $f_{\text{rep}}$  can be read out as a source of stable RF, a factor 10 better than that of a quartz oscillator at 100 ms, when  $f_0$  is stabilized to a modest RF reference (quartz oscillator) or a second optical reference at least 300 nm away from the first. Thus this work is a significant advance towards an all-fiber metrology system for moderate accuracy and good short-term instability in the near-IR and RF regimes without reliance on GPS.

This result demonstrates the viability of direct stabilization of a sub-100 MHz repetition rate fiber comb to a gas-filled hollow-core fiber toward an all-fiber metrology system. With some modifications, this system can be made more portable. The gas-filled hollow fiber, here mounted between two vacuum chambers, can be replaced with a sealed photonic microcell [26]. Furthermore, the 89 MHz rep rate comb with 9 GHz single stage filtering cavity may in the future be replaced with a GHz repetition rate comb based on one of many technologies [6–10, 12].

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