

# Frequency measurement of acetylene-stabilized lasers using a femtosecond optical comb without carrier-envelope offset frequency control

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**Abstract:** We have established a frequency measurement system for frequency-stabilized lasers operating in telecommunication wavelength bands, by using a femtosecond optical comb without the need for carrier envelope offset frequency control. This system has been used to measure the frequency of an acetylene-stabilized laser operating at 1542 nm for a period of over 10 hours. The frequency stability of the acetylene-stabilized laser is estimated to be  $3 \times 10^{-12}$  for a 10-s averaging time, improving toward  $1 \times 10^{-13}$  after 10000 s. We have measured three acetylene-stabilized lasers, including one commercially available laser, and confirmed that the frequency values are in good agreement (a frequency scatter of 2.1 kHz) with previously measured results reported by different institutes. In addition to the P(16) line of acetylene at 1542 nm, we measured the absolute frequencies of the P(24) line at 1547 nm, the P(1) line at 1534 nm, and the R(5) line at 1530 nm with a view to improving the accuracy of the acetylene frequency atlas. The acetylene-stabilized laser serves as an important optical frequency standard for telecommunication applications.

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## 1. Introduction

Recent advances in optical communication systems that include high-density wavelength-division multiplexing (DWDM) have motivated research on optical frequency standards in the 1.5- $\mu\text{m}$  wavelength region. Doppler-free high-resolution spectroscopy of acetylene was demonstrated and used to stabilize the frequency of a laser diode in the 1.5- $\mu\text{m}$  wavelength region [1, 2]. Acetylene provides several overtone and combination bands, of which a relatively strong band corresponds to the combination band of a C-H stretching vibrational mode (assigned as  $\nu_1+\nu_3$ ). This  $\nu_1+\nu_3$  band of acetylene is coincident with the International Telecommunication Union's C band (1530-1560 nm) and has attracted considerable attention as a frequency standard for telecommunication applications [2, 3].

The frequency of acetylene-stabilized lasers has been measured with different methods and improving accuracy. In the early days, the frequency of acetylene-stabilized lasers was measured by using a secondary frequency standard based on the Rb two-photon transition at 778 nm, where the measurement uncertainty was limited by the Rb-stabilized transfer laser [2, 4]. On the basis of this research, the International Committee for Weights and Measures (CIPM) recommended a value for the optical frequency of the P(16) transition line in the  $\nu_1+\nu_3$  band of acetylene with an uncertainty of 100 kHz (fractional uncertainty of  $5.2\times 10^{-10}$ ) [5].

Nowadays, optical frequency combs based on femtosecond lasers have revolutionized the field of optical frequency metrology, making possible the absolute measurement of optical frequencies with unprecedented accuracy [6, 7]. In 2003, for the first time, we measured the absolute frequency of a diode laser that was locked on the P(16) line of acetylene by using a femtosecond optical comb based on a Kerr-lens mode-locked (KLM) Ti:sapphire (Ti:s) laser [8]. Although the P(16) line does not lie within the bandwidth of the comb based on the Ti:s laser ( $\sim 500$ -1000 nm), we frequency-doubled the acetylene-stabilized laser to 771 nm. In this experiment, the measurement accuracy was limited not by the uncertainty of the frequency measurement but by the reproducibility of the acetylene stabilized laser. The frequency of the P(16) line based on the Ti:s comb has also been measured by other groups in different institutes [9, 10].

During frequency measurements based on the optical comb, it is usually necessary to stabilize two degrees of freedom of the comb, namely the repetition rate ( $f_{\text{rep}}$ ) and the carrier-envelope offset (CEO) frequency ( $f_{\text{CEO}}$ ). The  $f_{\text{CEO}}$  of a femtosecond comb spanning more than one octave was directly measured with an  $f$ -to- $2f$  interferometer [7]. An octave-spanning comb is often obtained by injecting the output of a KLM Ti:s laser into a photonic crystal fiber (PCF) [11]. Due to the small core diameter of the PCF ( $\sim 1 \mu\text{m}$ ), the efficiency with which the

laser beam couples into the PCF varies during the operation. This affects the stability of the signal-to-noise ratio (S/N) of  $f_{\text{CEO}}$ , and hence limits the long-term operation of the frequency measurement system. There are several methods for improving the long-term operation of the frequency measurement system.

- The core area of the PCF input side has been increased by splicing a short piece of tapered single-mode fiber onto the PCF. Meanwhile, active control of the coupling stage has been introduced to optimize the coupling efficiency [12].
- With a mode-locked fiber laser, a highly nonlinear fiber has been spliced onto the laser fiber to remove the coupling problem [13].
- With a broadband mode-locked laser,  $f_{\text{CEO}}$  has been observed without a PCF [14].

Alternatively, the sum-frequency generation (SFG) of an optical comb is considered an excellent method for measuring optical frequencies without the need for  $f_{\text{CEO}}$  control. SFG between an infrared optical frequency standard and the low-frequency wing of an optical comb yields an SFG comb. Assuming the SFG comb can be tuned to realize a spectral overlap with the high-frequency wing of the original comb, the beat note between the SFG comb and the high-frequency wing (both combs have the same origin) allows us to make an  $f_{\text{CEO}}$ -independent frequency measurement. This scheme was employed for measuring the absolute optical frequency of an  $\text{OsO}_4$ -stabilized  $\text{CO}_2$  laser [15] and implementing optical clockwork [16]. An SFG comb was evaluated and found to have excellent accuracy and stability [17].

In this paper, we demonstrate the absolute frequency measurement of acetylene-stabilized lasers by using an SFG scheme without  $f_{\text{CEO}}$  control. By using the measured frequency, the Allan variation of an acetylene-stabilized laser is calculated up to an averaging time of 10000 s without showing its flicker floor. The measured frequency values of the acetylene-stabilized lasers, together with previously measured values from different institutes throughout the world, show a frequency scatter of 2.1 kHz that is about 50 times smaller than the uncertainty of the CIPM recommended frequency. The absolute frequency of four acetylene lines in the 1.5- $\mu\text{m}$  wavelength region is measured for the purpose of improving the accuracy of the acetylene frequency atlas. The measured frequencies are important as regards updating the CIPM recommended frequency list [5].

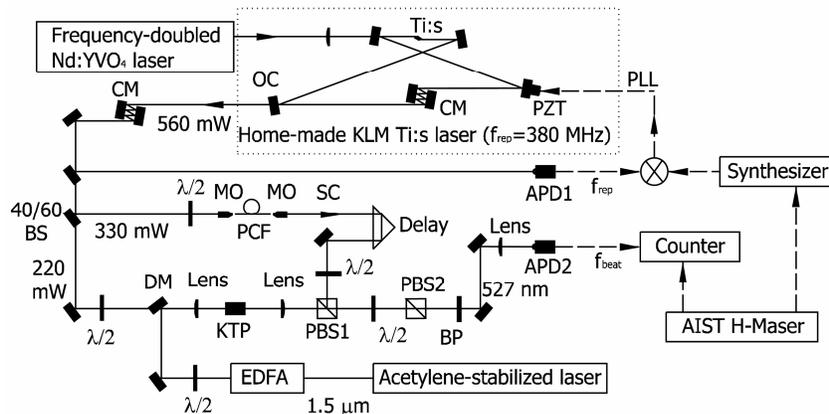


Fig. 1. Schematic diagram of the experimental setup. Ti:s, Ti:sapphire; CM, chirped mirror; OC, output coupler; PZT, piezoelectric transducer; BS, beam splitter; MO, micro-objective lens; PCF, photonic crystal fiber; SC, supercontinuum; DM, dichroic mirror; PBS, polarization beam splitter; BP, band-pass filter; APD, avalanche photo detector; EDFA, erbium-doped fiber amplifier; PLL, phase-lock loop.

## 2. Experimental setup

Figure 1 shows a schematic diagram of our experimental setup. The light source is a home-made KLM ring-cavity Ti:s laser with an  $f_{\text{rep}}$  of 380 MHz. A 4-mm-long Ti:s crystal with an absorption coefficient of  $\sim 5 \text{ cm}^{-1}$  at 532 nm was mounted at the Brewster angle. All the cavity

mirrors except the output coupler were chirped mirrors. As a pump light, the output of a frequency-doubled Nd:YVO<sub>4</sub> laser (Coherent, Verdi V10) was focused into the crystal by using a lens with a 40-mm focal length. With a pump power of 5 W, we obtained an average output power of 560 mW from the Ti:s laser with a pulse duration of 40 fs. The central wavelength and the spectrum bandwidth were 800 and 25 nm (FWHM), respectively.

Part of the output of the mode-locked laser was detected with an avalanche photo detector (APD1) to allow us to measure  $f_{\text{rep}}$ . The repetition rate,  $f_{\text{rep}}$  was beat down to approximately 20 MHz by using a synthesizer with a fixed frequency of 360 MHz. The beat-down frequency was fed to a phase-locked loop to control the length of the piezoelectric transducer attached to the mode-locked laser. The synthesizer was phase locked to an H-maser.

The output pulses were again chirp-compensated and split into two parts by a 40/60 beam splitter. One part, with an average power of 330 mW, was launched into a PCF for supercontinuum (SC) generation. A cw acetylene-stabilized laser at 1542 nm was amplified by an erbium-doped fiber amplifier (EDFA) and overlapped with the other part of the laser pulses from the Ti:s laser by a dichroic mirror. The overlapped beams with dual wavelengths were focused into a KTiOPO<sub>4</sub> (KTP) crystal to generate an SFG comb.

The SFG comb was arranged so that it overlapped with the SC comb at a polarization beam splitter (PBS1). A delay line was introduced in the path of the SC comb to match the timing of the two combs (which were actually fs pulses in the time domain). A half-wave plate and another polarization beam splitter (PBS2) were introduced to adjust the power ratio of the two combs in the beat measurement. The beat frequency of the two combs was detected after a band-pass filter by a second avalanche photo detector (APD2) and recorded by a counter (Agilent 53132A Universal Counter). The counter used a time base obtained from the H-maser.

### 3. Experimental results

#### 3.1 Sum-frequency comb generation and beat frequency measurement

In Fig. 2(a), the spectra of the original Ti:s comb are indicated as the dashed curve. This comb is used to generate the sum-frequency comb with a cw acetylene-stabilized laser at 1542 nm. The SFG was realized by using a 5-mm-long Type-II phase matching ( $\theta=59.6^\circ$ ,  $\Phi=0^\circ$ ) KTP crystal. In Fig. 2(a), the solid curve indicates the spectra of the SFG comb, with a central wavelength and bandwidth of 527 and 7 nm, respectively. The spectrum bandwidth of the SFG comb was limited by the phase matching condition of the KTP crystal. The average power of the SFG comb was about 100 nW. The power level of each comb is relatively adjusted in Fig. 2(a) for better presentation. The cw laser power after the EDFA was about 160 mW.

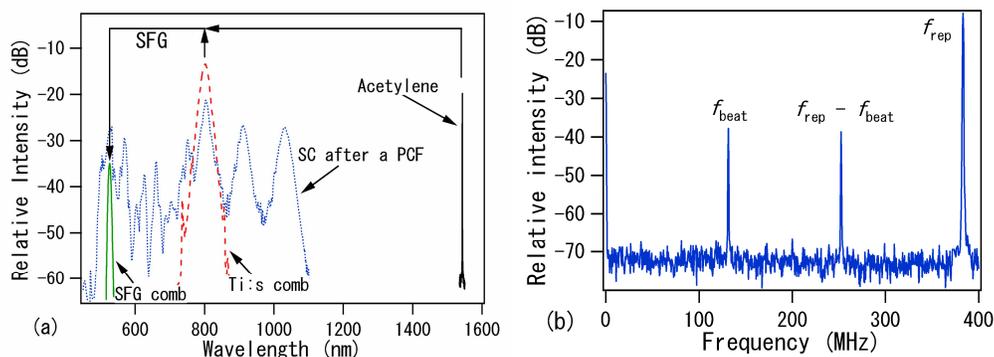


Fig. 2. (a) Spectra of the original Ti:s comb (dashed curve), the SC comb after the PCF (dotted curve), the SFG comb (solid curve), and the acetylene-stabilized laser (straight line). (b) Beat frequency observed between the SFG and SC combs at 527 nm. The resolution bandwidth was 300 kHz.

The spectrum of the SC after the PCF ranged from 480 to 1120 nm (-20 dB) and is shown by the dotted curve in Fig. 2. The PCF used in this experiment had a core diameter of 1.8  $\mu\text{m}$  and was 30 cm long. The power of the generated SC after the PCF was 160 mW. The SFG comb and part of the SC comb in the same wavelength region (527 nm) were combined spatially and temporally to generate a heterodyne beat note ( $f_{\text{beat}}$ ) [shown in Fig. 2(b)]. The S/N of  $f_{\text{beat}}$  exceeds 30 dB at a resolution bandwidth of 300 kHz. The average power of the SC comb used for the beat measurement around 527 nm was about 10 mW.

With the measured  $f_{\text{beat}}$ , the frequency of the cw laser at 1.5  $\mu\text{m}$  ( $f_{\text{laser}}$ ) can be expressed as  $f_{\text{laser}} = n \times f_{\text{rep}} \pm f_{\text{beat}}$ . We notice that  $f_{\text{CEO}}$  does not appear here in the formula because both the SFG and SC combs have the same origin.

Table 1. Selected operating parameters and characteristics of the laser systems

Parameter	A1 and A2	A4
C <sub>2</sub> H <sub>2</sub> pressure (Pa)	4	4
Laser power before the cavity (mW)	4.0	3.5
Coupling efficiency	60%	65%
Finesse calculated from the reflectivity	208	208
Cavity waist $2\omega$ (mm)	0.92	0.92
Modulation frequency $f$ (Hz)	333	1000
Modulation width (MHz)	1	1

### 3.2 Long-term stability of acetylene-stabilized laser

In the present experiment, three different acetylene-stabilized lasers are involved in the frequency measurement. Two of the lasers were developed in our institute [3] (denoted as A1 and A2) and the third is a commercially available laser (Neoark, model C2H2LDS-1540; denoted as A4). The operating parameters of the laser systems are similar and listed in Table 1. Figure 3 shows the square root of the Allan variance of the measured frequencies as a function of the averaging time,  $\tau$ . The solid line with the filled circles indicates the Allan variance of A4 calculated from the measured  $f_{\text{laser}}$  over ten hours. A4 was locked to the P(16) line of <sup>13</sup>C<sub>2</sub>H<sub>2</sub>. Since the stability of the H-maser-based comb was more than one order of magnitude better than the acetylene-stabilized laser, the observed instability of the measured  $f_{\text{laser}}$  was mainly contributed from the frequency instability of the acetylene-stabilized laser.  $f_{\text{rep}}$  was confirmed to be successfully phase locked to the H-maser by using an iodine-stabilized Nd:YAG laser in our previous experiments [8, 18].

The stability of A4 was  $3 \times 10^{-12}$  for a 10-s averaging time, improving to  $1 \times 10^{-13}$  after 10000 s, without showing a flicker floor. Stable long-term operation of the comb measurement system was achieved by removing one critical servo port ( $f_{\text{CEO}}$ ) using the SFG scheme. For comparison, the measured stability of A1 [8] is also indicated in Fig. 3 as a straight line. The stability of A4 is slightly better than that of A1 in the 10 – 1000 s averaging time range.

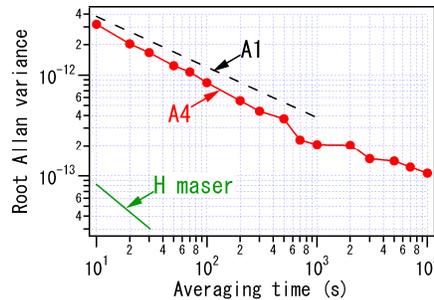


Fig. 3. Measured root Allan variance of A4 (solid line with filled circles). The root Allan variance of A1 and H-maser are shown as a dashed line and a solid line, respectively, for comparison.

### 3.3 Frequency measurement of acetylene-stabilized lasers

Absolute frequency measurements for A1, A2 and A4 were performed with the SFG scheme. The measured frequency and uncertainty are listed in the second column of Table 2. The lasers were locked on the P(16) line of  $^{13}\text{C}_2\text{H}_2$ . Each measured frequency in the table is an average of 50 to 400 pieces of beat frequency data (depending on the measurement run), where each piece of beat frequency data was measured with a 10-s gate time by the frequency counter. The standard deviation of the beat frequency data for each measurement run was about 0.8 kHz (fractional uncertainty  $4 \times 10^{-12}$ ). This uncertainty corresponds to the root Allan variance of the laser frequency measured with a gate time of 10 s. During the measurement, all the lasers were kept locked all the time. Therefore, the measurement uncertainty does not include the lock-to-lock reproducibility of the laser. The lock-to-lock reproducibility of acetylene-stabilized lasers was investigated in detail during our previous measurements [8, 13], and determined to be 2 ~ 3 kHz.

As shown in Table 2, the present frequency value of A1 is about 2.2 kHz lower than our previously measured result [8]. This frequency difference is within the reproducibility of A1. A similar discussion is also applied to the frequency difference of A2. The absolute frequency of A4 is measured for the first time. The accuracy of the H-maser was calibrated to UTC and confirmed to be better than  $1 \times 10^{-14}$  during the measurements.

Table 2. Absolute frequency of three different lasers locked on the P(16) line of  $^{13}\text{C}_2\text{H}_2$ .

Laser	$f_{\text{Present}}$ (kHz)	$f_{\text{Previous}}$ (kHz) <sup>a</sup>	$f_{\text{Present}} - f_{\text{Previous}}$ (kHz)
A1	194 369 569 381.4 (1.1)	194 369 569 383.6 (1.3)	- 2.2
A2	194 369 569 383.0 (0.5)	194 369 569 386.5 (1.6)	- 3.5
A4	194 369 569 386.8 (0.7)		

<sup>a</sup>From Ref. [8].

In Fig. 4, together with the present measurement results for A1, A2 and A4, we also show our previous results [8] and the results from the National Physical Laboratory (NPL), United Kingdom [9] and the National Research Council (NRC), Canada [10]. The uncertainty of each measurement depends on the measurement conditions. For example, when we measured A1 and A2 in 2003 we included the reproducibility of the acetylene-stabilized lasers, whereas this is not included in the present measurement. The frequency scatter of the eight measurements, indicated in Fig. 4, is 2.1 kHz. This frequency scatter represents the frequency reproducibility of both a single laser and an ensemble of acetylene-stabilized lasers. This is about 50 times smaller than the uncertainty of the 2001 CIPM recommended frequency [5]. The average frequency of these eight measurements is 194369569384.6(2.1) kHz. These results are important for updating the CIPM recommended frequency of the acetylene-stabilized laser.

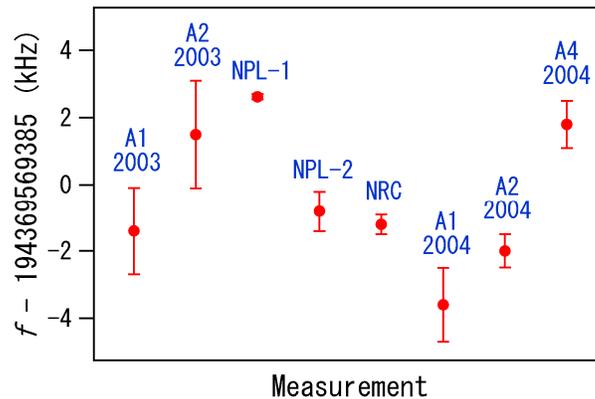


Fig. 4. Absolute frequency of acetylene-stabilized lasers obtained in different institutes. NPL-1 and NPL-2 are the lasers from NPL [9]. NRC is the laser from NRC [10].

Table 3. Absolute frequency values of acetylene lines

Wavelength (nm)	Line	Absolute frequency (kHz)
1547.5	P(24)	193 728 847 417.0 (0.5)
1542.4	P(16)	194 369 569 383.0 (0.5)
1533.8	P(1)	195 455 036 457.5 (0.9)
1530.2	R(5)	195 911 420 899.6 (0.3)

### 3.4 Absolute frequency measurement of multiple acetylene lines in 1.5- $\mu\text{m}$ band

The acetylene lines in the 1515 to 1550 nm wavelength range [2] provide an attractive frequency atlas for telecommunication applications. The absolute frequency of the P(24), P(1) and R(5) lines are measured, for the first time, using the SFG comb. The measured frequencies are listed in Table 3. The frequency measurement was performed with A2. The absolute frequency of each line was measured by taking an average of 10 to 50 beat frequency data (depending on the measurement run), where each beat frequency datum was measured with a 10-s gate time by the counter. The P(24) line is at the low frequency end of the operation range of our laser due to the S/N limitation of the absorption signal. In contrast, the lines with frequencies higher than the R(5) line are outside the amplification range of our EDFA. We notice that the frequency measurement range with the present SFG scheme is much wider than the phase-matching frequency range of the periodically poled LiNbO<sub>3</sub> (PPLN) crystal, which was used to double the frequency of the acetylene-stabilized laser from 1542 to 771 nm for frequency measurements with a Ti:s based comb [8].

The frequency of the line spacing was measured by measuring the heterodyne beat between two acetylene-stabilized lasers bridged with an optical frequency comb localized at 1.5  $\mu\text{m}$  [19]. From the metrological point of view, it is important to confirm this line spacing measurement with a different method. By using the absolute frequency values listed in Table 3, we calculated the frequency spacing between the lines and listed them in Table 4. These results are compared with the measured line spacing in [19] (also listed in Table 4). Good agreement is obtained between the two measurements. These results should also be included in the CIPM recommendation.

Table 4. Line spacing of the acetylene frequency atlas

Line spacing	$f_{\text{Present}}$ (kHz)	$f_{\text{Previous}}$ (kHz) <sup>a</sup>	$f_{\text{Present}} - f_{\text{Previous}}$ (kHz)
P(24)-P(16)	-640 721 966.0(0.7)	-640 721 966.5(1.0)	0.5
P(1)-P(16)	1 085 467 074.5(1.0)	1 085 467 073.9(1.9)	0.6
R(5)-P(16)	1 541 851 516.6(0.6)	1 541 851 518.2(1.4)	-1.6

<sup>a</sup>From Ref. [19].

## 4. Conclusion

We have established a frequency measurement system without  $f_{\text{CEO}}$  control for frequency-stabilized lasers operating in the 1.5- $\mu\text{m}$  telecommunication band by using an SFG comb. We demonstrated continuous frequency measurement without interruption for over 10 hours. The measurement scheme can be easily extended to any IR wavelength region (longer than 1.5  $\mu\text{m}$ ) by employing a suitable SFG crystal.

The results of frequency measurements of three different lasers show that the acetylene-stabilized laser is a reliable optical frequency standard for wavelength-division multiplexing (WDM) applications. We suggest adjusting the value and the uncertainty of the CIPM recommendation based on these new measurement results.

The optical frequency standard at 1.5  $\mu\text{m}$  is easily delivered through fiber networks. With the help of a high efficiency nonlinear crystal, such as waveguide PPLN, it should also be possible to convert the frequency standard at 1.5  $\mu\text{m}$  to frequency standards at red (771 nm) or green (514 nm) wavelengths using second-harmonic generation or third-harmonic generation, respectively. The acetylene-stabilized laser will open the way to many applications related to frequency metrology.

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