Widely tunable THz synthesizer

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Abstract The generation of cw-THz radiation by pho-
tomixing is particularly suited to the high resolution spec-
troscopy of gases; nevertheless, until recently, it has suffered
from a lack of frequency metrology. Frequency combs are
a powerful tool that can transfer microwave frequency stan-
dards to optical frequencies and a single comb has permitted
accurate (10^{-8}) THz frequency synthesis with a limited tun-
ing range. A THz synthesizer composed of three extended
cavity laser diodes phase locked to a frequency comb has
been constructed and its utility for high resolution gas phase
spectroscopy demonstrated. The third laser diode allows a
larger tuning range of up to 300 MHz to be achieved with-
out the need for large frequency excursions, while the fre-
quency comb provides a versatile link to be established from
any traceable microwave frequency standard. The use of a
single frequency comb as a reference for all of the cw-lasers
eliminates the dependency of synthesized frequency on the
carrier envelope offset frequency. This greatly simplifies the
frequency comb stabilization requirements and leads to a
reduced instrument complexity.

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1 Introduction

The opto-electronic generation of cw-THz radiation is at-
tractive as a single device is able to access all frequencies
from 300 GHz to 3 THz [1–3]. In competition with this
solution are purely electronic sources now commercially
available up to 1 THz, with a specific multiplier chain typ-
ically covering a bandwidth of around 150 GHz. One of
the shortcomings of opto-electronic solutions has been the
limited metrology of the THz frequency generally based on
wavelength measurements of the source lasers which are
able to provide a relative accuracy of $\Delta \nu_{\text{THz}}/\nu_{\text{THz}} = 10^{-5}$.
A Pound–Drever–Hall scheme can be used to lock two lasers
to a single high finesse Pérot Fabry (PF) etalon [4]. This
yields a solution that is sensitive to thermal, acoustic, and
mechanical interference and does not provide a direct link
to a frequency standard. The construction of a PF with a
suitable stability and the determination of its free spectral
range (FSR) are the principal weaknesses of this approach.
A calibration step is generally undertaken using a well-
known molecular transition to measure the FSR, this pro-
cedure is closer to a wavelength measurement rather than a
frequency synthesis. Replacing the etalon by an optical fre-
quency comb (FC) allows a direct link to a traceable fre-
quency standard to be established. A configuration using
two cw-lasers locked to a FC has a limited potential for
frequency scanning due to the large frequency excursion
requirement, as described in Sect. 3.1, however, an accu-
racy of 10^{-8} can be realized [5, 6]. Alternatively, two CW
lasers can be locked to two independent FC, although large
direct tuning can be achieved the system requires two FC

Fig. 1 Frequency diagram of the three CW lasers phase locked to the FC
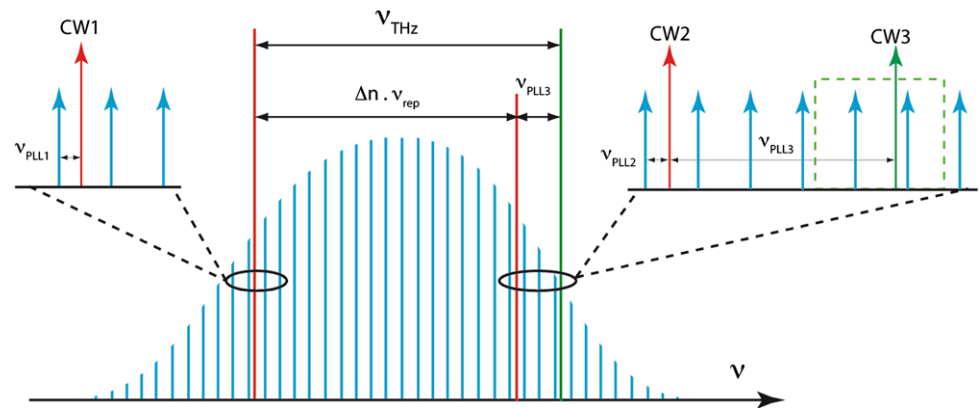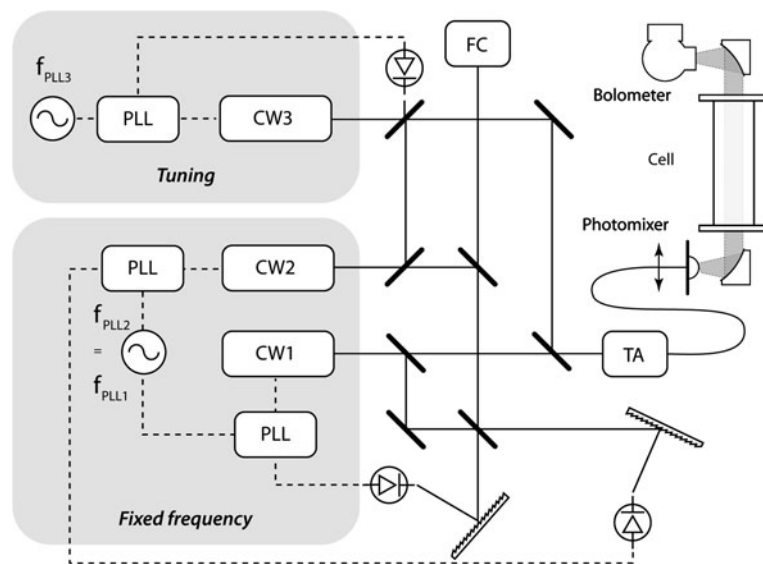

Fig. 2 THz frequency synthesizer and high resolution spectrometer. The fixed base frequency is generated by locking CW1 and CW2 to a FC. The phase locked tuning of the synthesized frequency is performed by PLL3 locking CW3 to CW2. The optical beatnote between CW1 and CW3 is converted to THz radiation by a standard LTG-GaAs spiral photomixer

each with a carrier envelope offset frequency (f_{ceo}) stabilization scheme [7]. Each FC must be spectrally extended and frequency doubled in order to generate a beatnote between the second harmonic and fundamental FC; this yields a particularly complex and expensive experimental configuration. The need to measure and stabilize f_{ceo} has significantly hampered application of FC for numerous frequency measurement tasks. We propose a simple configuration with three cw-lasers to give access to a larger tuning range without the need for large frequency excursions or a second FC. Two cw-lasers (CW1, CW2) are phase locked onto the FC whereas the third (CW3) is phase locked onto CW2. The THz beatnote between CW1 and CW3 is synthesized and can be continuously tuned over 300 MHz; see Fig. 1.

2 Experimental configuration

An erbium doped femtosecond fiber laser (MenloSystem C-Fiber) operating with a pulse duration of 150 fs was used to generate a FC extending from 1540 nm to 1600 nm in

the infrared. The laser repetition rate (f_{rep}) was stabilized to a direct digital synthesizer (DDS), the nominal repetition rate is 100 MHz with a tuning range of ± 200 kHz. All of the radio frequency synthesizers are referenced to a calibrated temperature controlled low phase noise crystal (Wenzel, BTULN) with a relative accuracy of 10^{-8}, and a stability of around 10^{-10}/day. A power amplifier and second harmonic generation module was used to transfer the FC into the near-infrared providing an emission power of 80 mW over the range from 774 nm to 780 nm, giving access to in excess of 2.8×10^4 FC modes. Three extended cavity diode lasers (ECLD) operating around 780 nm with typical powers of 50 mW are used for the cw-lasers. Each cw laser is equipped with a field effect transistor (FET) for rapid modulation of the diode current. CW1, CW2, and the FC are mixed together and a ruled grating employed to allow a small area photodiode to isolate a single cw-laser with its corresponding FC mode, limiting the incident power on the detector; see Fig. 2. The resulting beatnote is compared to a synthesized frequency using a digital phase comparator (MenloSystems), a proportional-integral-

derivative (PID) controller is used to apply an error signal to the ECLD current via the FET, in this way two hybrid opto-electronic phase locked loops (PLL1 & PLL2) are formed with CW1 and CW2 being coherently locked at 20 MHz to two different FC modes. CW2 and CW3 are mixed and their optical beatnote detected with a rapid photodetector forming third hybrid opto-electronic PLL3 using a MCH12140 phase detector. The output of PLL3 is filtered into fast and slow signals that are used to apply the correction signals to the current and piezo of control of CW3, respectively. The bandwidth ($\approx$1 MHz) of the servo loop permits the spectral purity of the beatnote to be narrowed, and its operating frequency range gives access to continuous tuning from 50 MHz to 375 MHz. A tapered amplifier (TA) (Toptica BoosTA) was simultaneously seeded by CW1 and CW3 to increase the available power of the beatnote, a maximum power of 500 mW can be obtained at the output of the TA. Optimal spatial mode matching between the two frequencies is ensured by coupling the output of the TA to a polarization maintaining fiber optic. A LT-GaAs photomixer device consisting of a interdigitated electrode array and a spiral antenna is used to convert the THz beatnote into free space THz radiation [8, 9]. A total laser power of 30 mW is used to illuminate the photomixer resulting in a photocurrent of 0.5 mA. The THz beam is precollimated by a Si hemisphere and subsequently manipulated by a pair of off-axis parabolic mirrors, which propagate it through a gas cell and focus it onto a He cooled InSb detector. The maximum power available from the photomixer is 1 μW at frequencies of 500 GHz or less. The device response decreases as the frequency increases due to the electrode/antenna structure and LT-GaAs charge carrier lifetime, at 1 THz 100 nW can be obtained which falls to around 80 pW at 3 THz.

2.1 Frequency tuning—configuration with two CW lasers

The frequency of any FC mode is the product of its mode number and repetition rate plus the carrier envelope offset frequency (f_{ceo}). In the case of cw-THz generation by optical heterodyning, the locking of two lasers to a single FC eliminates the dependency on f_{ceo}

$$\upsilon_{THz} = \Delta n f_{rep} \pm f_{PLL1} \pm f_{PLL2} \tag{1}$$

This greatly simplifies the optical configuration and the stabilization needs of the FC. The beatnote between the second harmonic and fundamental FC is not required, simply a servo loop acting on the FC cavity length to control f_{rep} is sufficient. A standard wavelength-meter with offering an accuracy of 10^{-7} is used to determine the number of FC modes ($\Delta n = n_2 - n_1$) separating the CW1 and CW2 lasers. This difference frequency is said to be synthesized as the FC mode spacing is defined by the repetition rate DDS and the cw-lasers are phase locked to the FC. A direct link is

therefore established between the synthesized THz domain frequency and the master frequency reference, in our case an accuracy of 10 kHz is achieved at 1 THz. The polarity of the PLL defines if the CW laser is locked on the high or low frequency side of the FC mode. If the system is constrained by using a single oscillator for $f_{PLL1} = f_{PLL2}$, configuring both PLL1 and PLL2 allows either $\upsilon_{THz} = \Delta n f_{rep}$ or $\upsilon_{THz} = \Delta n f_{rep} \pm 2 f_{PLL}$ to be selected. Tuning the synthesized frequency can be realized by scanning either f_{rep}, or $f_{PLL1} = f_{PLL2}$ in the latter case, however, only a limited range can be obtained [5]. In the case of f_{rep}, the absolute frequency of the mode is dependent on its mode number whereas the difference frequency varies according to the mode separation. A penalty factor (P) corresponding to the ratio between the laser frequency and the synthesized frequency is therefore incurred for the required laser frequency excursion, a pair of ECDL with continuous tuning over 10 GHz would yield a variation of approximately 25 MHz in the synthesized frequency (2).

$$P = \frac{\Delta \nu_{CW1}}{\Delta \nu_{THz}} = \frac{n_1}{\Delta n} \approx \frac{\nu_{CW1}}{\nu_{THz}} \tag{2}$$

Alternatively scanning $f_{PLL1} = f_{PLL2}$ benefits from direct variation of the synthesized frequency but is limited by the presence the neighboring FC mode and the electronic filtering required to isolate desired beatnote, a maximum scanning of 10 MHz can be realized with this FC [5].

2.2 Frequency tuning—configuration with three CW lasers

The accuracy of the FC and the versatility of the hybrid opto-electronic PLL are harnessed by adding CW3. CW1, and CW2 are used to contribute a fixed frequency component to the synthesized frequency. The frequency reference to the FC is conserved by the phase locking of CW3 to CW2 while the entire operating range of PLL3 can be scanned due to the absence of the FC in the detected signal. Hence, direct tuning of the THz frequency synthesized between CW1 and CW3 by up to 300 MHz is achieved (3).

$$\begin{aligned} \upsilon_{THz} &= \Delta n f_{rep} + f_{PLL3} \quad \text{or} \\ \upsilon_{THz} &= \Delta n f_{rep} + f_{PLL3} \pm 2 f_{PLL} \end{aligned} \tag{3}$$

In this configuration, the maximum tuning range is only limited by the operating range of the PLL. The FWHM of the three beatnotes used in the PLLs are typically 10 kHz or less. The spectral purity of the synthesized THz frequency is the convolution of these three beatnotes; we therefore estimate it to be in the order of 50 kHz.

3 Measured spectra and discussion

The extended tuning range and high resolution offered by this configuration can be demonstrated by measuring the

Fig. 3 Measured spectrum of
CH_3Cl for the transitions
$J = 38 \leftarrow 37$ and $0 \leq K \leq 4$,
using a standard 65 cm sample
cell, $P = 7.8$ Pa and $T = 293$ K
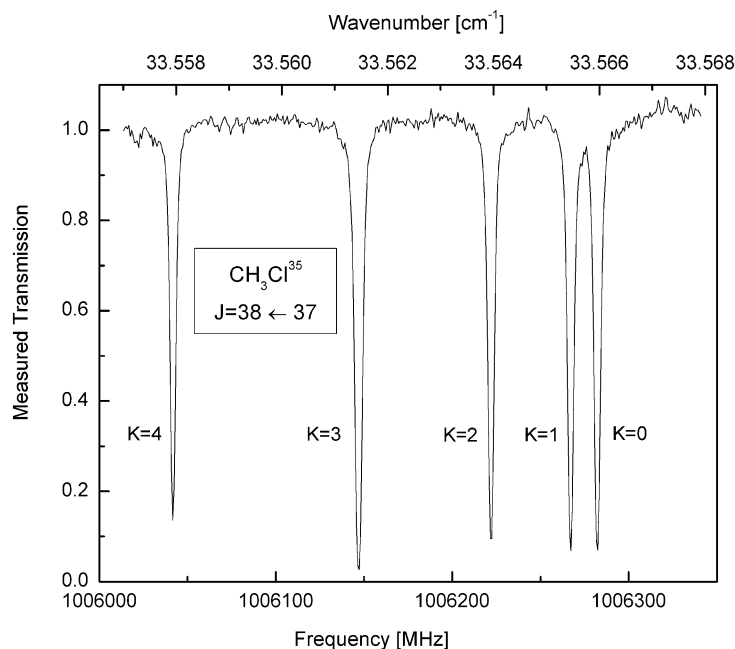
various molecular transitions, for example, chloromethane (CH_3Cl) is the most abundant atmospheric haloalcane. It is considered to be the only natural source of chlorine present in the stratosphere and has a strong impact on the destruction of the ozone layer and contributes to the greenhouse effect. It displays a reasonably intense signature in the frequency range 50 GHz to 1600 GHz, with the strongest lines be located around 715 GHz. Its transitions are described by the principal quantum number J along with K its projection on the principal axis. A repetition rate $f_{rep} = 99.8675280$ MHz and a mode separation $\Delta n = 10{,}073$ allowed the synthesized frequency to be scanned from 1,006,013.610 MHz to 1,006,340.810 MHz. All the lines for $J = 38 \leftarrow 37$ and $0 \leq K \leq 4$ were measured in a single scan (Fig. 3) using a measurement cell with a path length of 65 cm. The frequency scale is sampled with a resolution of 800 kHz (2.7×10^{-5} cm^{-1}) and the entire spectrum was obtained in around 5 min. The high resolution of this instrument is underlined by the detail contained in this spectrum, a standard commercially available Fourier Transform spectrometer can typically provide a resolution of 300 MHz (10^{-2} cm^{-1}). Whereas the large continuous tuning range maybe compared with previous FC based solutions which until now have been limited to 10 MHz for a fully phase locked system [5]. The temperature (300 K) and pressure (78 Pa) used for this experiment ensure the line broadening is dominated by the Doppler contribution, hence the narrow lines allow the dependency on K to be fully resolved. This molecule also exhibits a hyperfine structure caused by the presence of chlorine but this effects is significantly finer than the Doppler broadening so can not be resolved under these experimental conditions. Taking into account the hyperfine structure, the

measured lines centers are within 100 kHz of the tabulated lines in the Jet Propulsion Laboratory (JPL) database [10].

Both ethanol and methanol also display particularly congested spectra in the THz region, their THz signatures are more complex than CH_3Cl because they are asymmetric tops with coupling between their torsional and rotational states [11, 12]. The intensity of the molecular lines required the use of a White configuration multiple pass gas cell with a path length of 4.24 m. The direct absorption signal (amplitude modulation) of a mixture of these gases at 1.197 THz illustrates the ability of THz radiation to discriminate between two molecular species; see Fig. 4. Two isolated ground state transitions of ethanol (E) ($J' = 29$, $K'_a = 12$, $K'_c = 17$–18)$\leftarrow$ (28, 11, 17–18) and methanol (M1) ($J' = 17$, $K' = 5, -$) $\leftarrow$ (17, 4, +) are observed together with three overlapping transitions of methanol. One ground state (M2) (17, 5, +) $\leftarrow$ (17, 4, −), and two first excited torsional state transitions (M_e3)(25, 3) $\leftarrow$ (24, 3), and (M_e4)(24, 9) $\leftarrow$ (23, 8). The complexity of the methanol spectrum is illustrated by the transitions M1 and M2 which have the same rotation state but two different torsional transitions. The sensitivity of the measurement can be increased by implementing a frequency modulation scheme, however, this requires a careful control of the modulation depth. A standard laboratory radio frequency synthesizer can generate a frequency modulated signal with a precisely defined modulation depth and speed. Provided that the loop bandwidth of PLL3 is not exceeded a frequency ramp with FM can be easily applied, via CW3, to the synthesized THz frequency while remaining phase locked at all times. A modulation of depth 2.4 MHz and at a rate of 7 kHz was used for the same spectral region with a synchronous detection

Fig. 4 *Upper pane* direct absorption of an ethanol (10 Pa) and methanol (5 Pa) mixture. 2nd pane FM signal of pure ethanol (5 Pa) *black line*, pure methanol (5 Pa) *blue line* and an ethanol (7 Pa) methanol (5 Pa) mixture *red line*. 3rd pane line intensities for ethanol (*red squares*) and methanol (*blue triangles*) as tabulated [10]. All measurements performed with a path length of 4.24 m and at 293 K
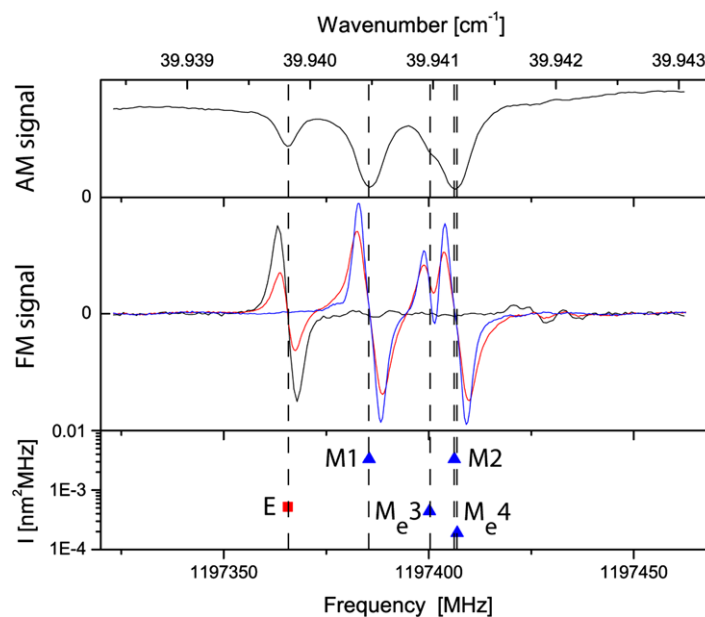
referenced to the modulation signal giving the first derivative of the absorption profile; see 2nd pane, Fig. 4. The isolated lines show strong signals and the zero crossing frequencies are within 30 kHz of the values from other work [11, 12]. The previously overlapped M_e3 can now be clearly distinguished and the entire measured profile is in excellent agreement with the predicted center frequencies and the line intensities as tabulated by JPL [10]; see 3rd pane, Fig. 4. According to the tabulated values M2 and M_e4 are separated by only 730 kHz while the Doppler broadening here at 20°C is 2.6 MHz FWHM preventing these lines from being resolved. A weak untabulated transition of ethanol can also be observed at 1197426.29 MHz. The sensitivity offered by the multiple pass cell and the FM allowed an ethanol transition with an intensity of 1.6×10^{-6} nm^2 MHz to be observed at 607 GHz with an SNR = 1 using a room temperature Schottky diode detector. The spectra of pure ethanol, pure methanol, and a mixture show that the synthesized frequency scale has an excellent reproducibility. Here a calibrated crystal was used as the master frequency reference however the flexibility of the FC allows this to be easily replaced by a signal derived from a traceable standard such as a GPS locked Rubidium source. With the addition of CW3 not only can larger spectral features containing many lines now be scanned but an entire frequency range can be covered without the need of changing the FC repetition rate. This feature is particularly attractive for molecules that have complex signatures or for which the predicted transition frequencies contain significant uncertainties. As this frequency, tuning mechanism does not incur any penalty, $\delta\upsilon_{IR} = \delta\upsilon_{THz}$ then further extension of the scanning range to values in excess of 1 GHz are possible by improving the performance of PLL3. The use of PLLs in the instrument architecture allows the straightforward manipulation of the synthesized THz frequency. As the instrument is phase locked accurate sweeping and modulation patterns are easily programmed, however, more complex frequency hopping series aimed at rapidly probing the multiple molecular species may be envisaged. The overall system complexity with a single FC, three CW lasers and the associated PLLs is considerably simpler than using two independent FCs as proposed elsewhere [7]. When not only two femtosecond lasers are required but the emission from each must be spectrally extended and frequency doubled in order to allow the carrier envelope frequency to be stabilized.

There are two different possibilities which may be envisaged to further simplify our instrument while maintaining the spectral resolution required for molecular spectroscopy of gases and increasing the tuning range. Firstly, rather than phase locking CW3 to CW2, it may be operated free-running close to CW2. The unlocked beatnote between CW2 and CW3 should be measured by a frequency counter and synchronously recorded with the corresponding FM or AM detected signal as CW3 is swept across the required frequency range. The success of such a system is dependent on the natural stability of CW3, and the rapidity of the frequency counter. However, a relatively large tuning range may be easily achieved as it is only limited by the detector and frequency counter. The second solution would be to use a FC with a larger separation, either a higher repetition rate femtosecond laser or even an electro-optic modulator based optical frequency comb generator (EOM-OFCG). The use of a more widely spaced comb structure permits direct PLL tuning to be used over a reasonable frequency range. A EOM-OFCG operating at 1.5 μm with an EOM frequency of 6.25 GHz has already been demonstrated [13]. The avail-

ability of this kind of comb at 800 nm, or a photomixer device for use at 1.5 μm, will this allow this proposition to be realized.

4 Conclusion

A relatively simple THz synthesizer configuration has been implemented using three cw-lasers and a singled FC operating at 100 MHz. No f_{ceo} stabilization scheme is required as all the lasers are referenced to the same FC. Any microwave frequency standard can be directly introduced to the instrument via the f_{rep} synthesizer. A continuous frequency tuning range of over 300 MHz has been realized thanks to the addition of CW3, some 30 times greater compared with a two cw-laser configuration [5]. The ability to continuous tune over a frequency range larger than the f_{rep} provides additional flexibility as all frequencies can be accessed without the need to modify f_{rep}. This source is a powerful tool for the measurement of the molecular line positions of gases, its integration into a laser spectrometer has clearly shown its utility to measure complex THz signatures whose line positions are known only within tens of MHz. The synthesized frequency can be readily modulated by programming the desired pattern on the PLL radio frequency oscillator. A standard frequency modulation scheme associated with a multipass cell was used to measure the weak molecular transitions of ethanol and methanol, demonstrating the excellent discrimination offered by this technique at low pressure. The implementation of a free-running CW3 version of this instrument may easily allow a larger tuning range to be obtained without the need for PLL3. The future development and availability of higher frequency FCs will allow a two cw-laser system to provide sufficient tunability to fulfill the needs for high-resolution spectroscopy.

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