

# Absolute frequency measurement in the 28 THz spectral region with a femtosecond laser comb and a long-distance optical link to a primary standard

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## ABSTRACT

A new frequency chain was demonstrated to measure an optical frequency standard based on a rovibrational molecular transition in the 28 THz spectral region accessible to a CO<sub>2</sub> laser. It uses a femtosecond-laser frequency comb generator and two laser diodes at 852 nm and 788 nm as intermediate oscillators, with their frequency difference phase-locked to the CO<sub>2</sub> laser. The RF repetition rate of the femtosecond laser was compared with a 100-MHz signal from a Hydrogen Maser, located at BNM-SYRTE. The 100 MHz signal is transmitted by amplitude modulation of a 1.55 μm laser diode through a 43-km telecommunication optical fibre. As a first example, the absolute measurement of a saturation line of OsO<sub>4</sub> in the vicinity of the P(16) laser line of CO<sub>2</sub> is reported with a relative uncertainty of 10<sup>-12</sup>, limited by the CO<sub>2</sub>/OsO<sub>4</sub> frequency day-to-day reproducibility. The current limit on the stability of the frequency measurement is 4×10<sup>-13</sup> at 1 s.

PACS : 06.20.-f, 42.62.Eh, 06.30.Ft

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## 1. Introduction

Until recently, the measurement of the absolute frequency of an optical frequency standard required a complex and expensive set-up, as developed in only a few specialised metrological laboratories. In 1999 for the first time, a femtosecond laser comb generator was used for an absolute frequency measurement [1]. This much simpler technique is very attractive and saw a large development in many laboratories all around the world (see for instance the review article [2] and [3, 4]). A primary frequency standard is still needed for these measurements : a Cs- clock, a Rb- clock or a H-maser, the frequency of which is compared to TAI (Temps Atomique International or International Atomic Time).

Here we describe the first absolute frequency measurement performed with a femtosecond laser and using a reference frequency signal transmitted from the BNM-SYRTE laboratory at the Paris Observatory. This signal is transmitted from SYRTE to LPL through a standard telecommunication optical fibre of length 43 km. It is controlled with the H-maser and the Cs fountain from SYRTE. Also for the first time, a 28 THz optical frequency is measured with the direct 25-THz-wide (FWHM) comb of a Ti:Sa laser. This requires non linear optical generation to transfer this frequency to the near infrared region. This new measurement method gives us the possibility to reach a relative accuracy much better than can be obtained with a GPS receiver or a Rb clock for averaging time as short as a few s.

The first application consisted in measuring the CO<sub>2</sub> laser at 10 μm locked to a saturated absorption line of the molecule OsO<sub>4</sub>. Since 1997, a set of CO<sub>2</sub>/OsO<sub>4</sub> secondary standards are recommended for the "mise en pratique du mètre" [5]. They had a preferential role in frequency measurement, because such infrared frequencies are intermediate between the Cs-clock primary standard and the visible optical clocks. They were used as references for many optical measurements, which could now be usefully verified with this new method. CO<sub>2</sub>/OsO<sub>4</sub> frequency standards can also be used to calibrate a large variety of molecular spectra : for example the widely-used frequency grid of CO<sub>2</sub> laser lines has been partly obtained from frequency measurement with respect to the OsO<sub>4</sub> lines [6].

## 2. The CO<sub>2</sub>/OsO<sub>4</sub> frequency standard

The CO<sub>2</sub>/OsO<sub>4</sub> stabilisation set-up was developed a few years ago for high-resolution molecular spectroscopy, high-precision measurement and fundamental physics. Fig. 1 displays the OsO<sub>4</sub> frequency stabilization system, which was already described in detail in [7]. It consists in a CO<sub>2</sub> laser stabilized onto a saturation signal detected in transmission of a Fabry-Perot cavity filled with OsO<sub>4</sub> [8]. A crucial component of the stabilization scheme is a broadband CdTe electrooptic modulator (EOM), which generates sidebands, one of which is tuned into resonance with the molecular line. Efficient and pure frequency modulations can then be applied to the sidebands simply by modulating the frequency of the synthesizer, which drives the EOM. The cavity resonance is locked to the sideband frequency and a second servo-loop then locks the EOM sideband to the molecular line. Standard conditions for the detection of the OsO<sub>4</sub> saturated absorption signal are a pressure of 0.04 Pa and a laser power of 50 μW inside the cavity. In this regime, the third harmonic of the molecular signal has a peak-to-peak linewidth of about 20 kHz and the signal-to-noise ratio is 500 in a bandwidth of 1 kHz. Two equivalent and independent systems has been developed to characterise the frequency stability. By stabilizing both lasers onto the same strong P(46) A<sub>1</sub><sup>2</sup>(-) OsO<sub>4</sub> line in coincidence with the P(14) CO<sub>2</sub> laser line we obtained an Allan deviation of  $3.5 \times 10^{-14} \tau^{-1/2}$  for integration time between 1 and 100 s, and a reproducibility of a few tens of Hz.

### 3. The optical link between LPL and BNM-SYRTE

The LPL is located in the surroundings of Paris, whereas in the centre of Paris the SYRTE develops cold atoms frequency standards in the microwave domain. Their Cs atomic fountain has a demonstrated accuracy of  $10^{-15}$ , and a frequency stability of  $5 \times 10^{-14} \tau^{-1/2}$  [9]. They have developed an internal reference signal at 100 MHz, which is distributed around their whole laboratory for various metrological experiments. Its stability is given by an H-Maser, Allan deviation in Fig. 3, and the accuracy is controlled either with the Cs fountains or with a comparison to the local time scale and with GPS. Typical accuracy of the 100-MHz signal is below  $10^{-14}$ .

It was very attractive to take advantage of this excellent reference for our measurements. Following what was done on the 3 km between SYRTE and Laboratoire Kastler-Brossel (LKB)[10], two optical links has been developed to transmit the 100-MHz signal from SYRTE to LPL. Interconnecting many different sections of existing standard single mode optical fibres results in two close optical links of 43 km, which is much larger than the physical distance between the two laboratories of 13 km. At the few tens of network interconnection points the fibres were fused to insure the continuity of the link. Optical time domain reflectometry measurement of one of the two link fibres has been performed, giving a one-way attenuation of about 10 dB at 1.55  $\mu\text{m}$  wavelength.

The complete optical link includes a distributed feedback (DFB) semiconductor laser, emitting at 1.55  $\mu\text{m}$ , and a p-i-n photodiode. The current of the laser is modulated at 100 MHz directly controlled by the H-Maser. The amplitude modulation is then converted back into the electrical domain by the photodetector at the output of the link [11].

This optical link must not degrade the spectral quality of the metrological signal, while the background noise could impact on the 100-MHz stability along the 43 km propagation. The fibres are sensitive to both the mechanical stress and the temperature variations. The first affects the phase noise performances of the link and the short-term frequency stability, the second is a slowly change of the optical length of the fibre, and impacts on the long-term stability. The metrological features of this link were measured by a complete round trip (86-km) of the 100 MHz signal as shown on fig. 2. The phase difference between the input and return signal was detected and its variation was analysed in terms of frequency fluctuations. The Allan deviation (square root of the Allan variance) is reported in fig. 3. This gives an upper limit for the Allan deviation for a one-way pass<sup>1</sup>. On the same graph is depicted the Allan deviation of the H-maser, which exhibits greater deviation. This demonstrates that the optical link does not degrade the performance characteristics of the H-Maser reference frequency for measurement times below 10000 s. A complete characterisation of the phase noise introduced by the optical link is under progress.

This signal is then used at LPL to phase-lock a low phase noise quartz oscillator at 5 MHz with an Allan deviation of  $5 \times 10^{-13}$  from 1 to 100 s. The servo bandwidth is a few Hz, in order to filter out the high frequency phase noise of the optical link. The quartz second harmonic at 10-MHz is then distributed through the laboratory and is used as an external reference clock for all the RF synthesisers and the counter involved in the frequency measurements. This 10-MHz signal reproduces the frequency stability of the 100-MHz received signal for integration time longer than 1 s. Its frequency stability is estimated a few  $10^{-13}$  for 1 s.

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<sup>1</sup> The time scale of Fig. 3 is larger than the round trip propagation time of 0.5 ms, and the corresponding phase fluctuations for the two-ways are thus strongly correlated. If phase noise is dominant against amplitude noise, the one-way Allan deviation can be deduced as half of the two-way pass.

## 4. Comparison of the 28 THz CO<sub>2</sub>/OsO<sub>4</sub> standard and the primary standard with a fs laser comb

### 4.1 Principle of the frequency measurement

The principle of frequency measurements with a femtosecond laser comb has already been widely described [3, 4]. The emission spectrum of a mode-locked femtosecond laser is composed of modes which are spaced by the laser repetition rate  $f_r$  (around 1 GHz in our case). It has been demonstrated that this space  $f_r$  is constant from one side to the other side of the spectrum at a level better than  $3 \times 10^{-17}$  [3]. This fundamental property permits us to use the femtosecond comb as a "frequency ruler". The comb width is proportional to the inverse of the pulse duration. The frequency of one mode is of the form :  $f_n = n \times f_r + f_0$ , where  $n$  is an integer and  $f_0$  the offset of the frequency comb. A first method of measurement consists in recording the beatnote between this comb and the optical frequency  $f$  to be measured :  $b = f - f_n = f - (n \times f_r + f_0)$ . Accurate measurements of  $f_r$  and  $f_0$ , and the determination of  $n$ , are needed to complete the frequency measurement.

Here we proceed in a quite different way because no direct beatnote can be observed between the CO<sub>2</sub> laser emitting at 10  $\mu\text{m}$ , that is 28 THz, and the frequency comb centered at 800 nm. We take advantage of the fact that the comb frequency width is larger than the CO<sub>2</sub> frequency. Then the CO<sub>2</sub> frequency can be compared to the frequency difference  $(n-m)f_r$  between two modes. This method avoids any measurement of  $f_0$ , which simplifies the experimental arrangement [4]. The CO<sub>2</sub> laser frequency must just be transferred to the spectral range of the comb. A similar way is used in [12] to build up a scheme to measure the frequency of He-Ne/CH<sub>4</sub> standard.

The measurement setup is depicted on Fig. 4. Two laser diodes emit at 852 nm and 788 nm and their frequency difference is phase-locked to the CO<sub>2</sub> laser with an offset  $\Delta_0$  :  $f(\text{CO}_2 / \text{OsO}_4) + \Delta_0 = f(788\text{nm}) - f(852\text{nm})$ . Then, the beatnotes  $\Delta_1$  and  $\Delta_2$  of these intermediate oscillators with the lower and upper part of the femtosecond laser comb are detected and controlled. In fact a very-high harmonic of the repetition rate (about the 28000<sup>th</sup>) is phase-locked to the CO<sub>2</sub> laser frequency:  $f(\text{CO}_2 / \text{OsO}_4) + \Delta_0 - \Delta_1 - \Delta_2 = p \times f_r$ . Finally, the repetition rate is measured with respect to the H-maser reference signal and the CO<sub>2</sub>/OsO<sub>4</sub> absolute frequency is deduced.

### 4.2 Experimental set-up

A scheme of the experimental set-up is displayed on Fig. 5.

The femtosecond Ti:Sa laser (GigaJet from GigaOptics company) has a repetition rate of  $\sim 1$  GHz and emits 550 mW around 800 nm for 5 W of pumping power. Its emission spectrum spans 25 THz (FWHM) from 775 nm to 825 nm.

The laser diodes LD1 and LD2 emit respectively at 852 nm and 788 nm, and are in an extended cavity configuration. LD1 is tuned to the Cs D2 resonance transition, for a simple and easy check of its frequency. An AgGaS<sub>2</sub> crystal is used to generate the sum-frequency (SF) of the CO<sub>2</sub> laser and LD1, in a type I angular phase-matching scheme [13]. By coupling 20 mW at 852 nm and 80 mW at 10  $\mu\text{m}$ , about 1.5  $\mu\text{W}$  of sum-frequency radiation (SF) at 788 nm is obtained (efficiency  $\approx 1 \text{ mW/W}^2$ ). Since this power is very small, LD2 is used as an optical tracking oscillator: the SF is separated from the incident beams with a polarization beamsplitter and mixed with the output of LD2. The frequency of LD2 is set to the sum-frequency with the help of a lambdameter.

Finally the two beatnotes  $\Delta_1$  and  $\Delta_2$  of LD1 and LD2 with the femtosecond laser comb are detected after spectral filtering with a 1200-line/mm grating. The signal-to-noise ratios are respectively 35 dB (LD1) and 45 dB (LD2) in a bandwidth of 100 kHz.

### 4.3 Measurement procedure

The beat note between LD1 and the  $n^{\text{th}}$  mode of the comb (frequency  $f_n$ ) is phase-locked to a RF synthesiser (phase-lock loop #1 in Fig. 5):  $f_n - f(\text{LD1}) = \pm\Delta_1$ . Corrections are applied both to the laser diode current and to a PZT controlling the length of the extended cavity. Then the beat note between the SF and DL2 is phase-locked to the same RF synthesiser and corrections are applied to LD2 (phase-lock loop #2) :  $f(\text{LD2}) - f(\text{SF}) = \pm\Delta_0 \equiv \mp\Delta_1$ . Finally the beat note between LD2 and the comb (the  $m^{\text{th}}$  mode) is phase-locked to a second RF synthesizer (phase-lock loop #3) and corrections are applied to the femtosecond laser cavity length via a PZT:  $f(\text{LD2}) - f_m = \pm\Delta_2$ .

To increase the dynamic range of the different phase-lock loops, the beatnotes are divided by 4 for loops #1 and #2, and by 64 for the loop #3. The synthesisers' frequencies are then respectively  $\Delta_1 / 4 = \delta_1$  et  $\Delta_2 / 64 = \delta_2$ . For the loops of the laser diodes bandwidths of 1 MHz are obtained and error signals do not exceed  $\pi/50$ , but the bandwidth of the servo-loop for the repetition rate is limited to a few kHz.

The combination of these frequency relations with  $f(\text{CO}_2/\text{OsO}_4) + f(\text{LD1}) = f(\text{SF})$  gives :  $f(\text{CO}_2/\text{OsO}_4) = f_m - f_n \pm 4\delta_1 \pm 4\delta_1 \pm 64\delta_2 = pf_r \pm 4\delta_1 \pm 4\delta_1 \pm 64\delta_2$  (a)<sup>2</sup>. The integer  $p$  is around 28378 ( $\approx 28$  THz/1 GHz), and is determined unambiguously from the existing OsO<sub>4</sub> frequency grid. Finally  $f(\text{CO}_2/\text{OsO}_4)$  is obtained by measuring  $f_r$ . It is detected with a fast photodiode, mixed with the output of a RF synthesizer  $\Delta_3$  to translate the difference frequency  $\delta$  typically below 100 kHz :  $f_r = \Delta_3 \pm \delta$ , and finally  $\delta$  is measured with a high-resolution reciprocal counter (Agilent 53132a) at a rate of one sample per s. The Allan deviation for longer time is then evaluated by averaging these 1 s measurements.

The uncertainty in  $f(\text{CO}_2/\text{OsO}_4)$  measurements depends mainly on the  $f_r$  measurement because it is multiplied by  $p \approx 28378$ . Thus the key point is to reduce the uncertainty in  $f_r$  measurement as much as possible. This is less than  $10^{-14}$ , using the BNM-SYRTE 100-MHz reference signal.

The measurement could be performed in a different way. The repetition rate could be first controlled with the SYRTE reference signal and then the beatnote between LD2 and the comb measured to deduce the CO<sub>2</sub>/OsO<sub>4</sub> frequency. But, for technical reasons, this method is less favourable in terms of noise performance. Moreover, with the present technique, an optical clock is actually demonstrated in the sense that the RF repetition rate frequency is controlled with the CO<sub>2</sub>/OsO<sub>4</sub> frequency standard. The stability of the infrared standard is transferred to the femtosecond laser, and  $f_r$  may ideally reach the optimal stability of  $3 \times 10^{-14}$  at 1 s of the CO<sub>2</sub>/OsO<sub>4</sub> frequency standard [7, 8], even if there is no possibility to measure this performance yet. This is better than the current short-term stability of the Hydrogen maser, and is a motivation to develop an infrared optical clock.

## 5. Results and discussion

Various OsO<sub>4</sub> frequencies, in the vicinity of the different CO<sub>2</sub> laser lines, have been measured in the past with classical frequency chains with an uncertainty in the range 20-60

<sup>2</sup> Changing the signs of the phase-lock loops increases the capture range of the whole system.

Hz [14, 15]. For the demonstration of the method, we focus our attention on a P(55) line of  $^{190}\text{OsO}_4$  near the P(16)  $\text{CO}_2$  laser line, the frequency of which is already well-known. It is located at 59.070 MHz from the  $\text{CO}_2$  line centre and was recently used as a reference for the measurement of two-photon absorption line in  $\text{SF}_6$  [16]. Its absolute frequency  $\nu_{\text{ref}}=28412648819596$  (45) Hz is calculated from two independent measurements. First the absolute frequency of the line located at 33.379 MHz from the  $\text{CO}_2$  line centre, which is an unidentified line belonging to the  $\text{OsO}_4$  frequency grid, is well known [5, 17]. Its frequency was recently re-measured by conventional frequency chain techniques with an uncertainty of 40 Hz [15]. Second, the frequency difference between the P(55) line and the grid line was measured at the LPL with an uncertainty of 20 Hz [14].

Fig. 6a displays the repetition rate measurement and the associated  $\text{OsO}_4$  frequency, deduced from the relation (a) above. Fig 6b gives the corresponding relative Allan deviation, which is  $4 \times 10^{-13}$  for a time of 1 s. This is due to the stability of the RF synthesiser's frequency  $\Delta_3$ , involved in the  $f_r$  measurement. The noise comes both from the imperfect 10-MHz to  $\Delta_3 \approx 1$  GHz frequency synthesis, and from the 10-MHz itself (see § 3). We measured the Allan variance of the beatnote of two synthesizers referenced to the 10-MHz signal. It is  $3 \times 10^{-13}$  for 1 s integration time and this gives a limit for the  $\text{CO}_2/\text{OsO}_4$  Allan variance. A direct 100-MHz to 1 GHz synthesis is being currently developed in order to improve this short-term stability.

Measurements were performed over a period of 9 months and are displayed on Fig. 7. Each point gives the statistical mean over 3 to 9 measurements performed the same day, and the error bars are the  $1-\sigma$  deviation of these measurements. The day-to-day change in the error bars is related to the variation in the experimental conditions: the alignment of the Fabry-Perot cavity, the EOM's frequency, or the adjustment of the baseline of the  $\text{OsO}_4$  error signal. The main limitation for the total uncertainty is the day-to-day repeatability.

Finally the mean value of the 9 measurements is calculated :  $\nu - \nu_{\text{ref}} = -16.5 \pm 58$  Hz , where the uncertainty is the  $1-\sigma$  deviation of the data of fig. 7. This result compares within less than  $1-\sigma$  with the previous measurements [14, 15], which was performed however with conventional frequency chain, and with a slightly different  $\text{CO}_2/\text{OsO}_4$  stabilisation set-up [18]. This reproducibility is probably limited by systematic shifts connected with the use of a Fabry-Perot cavity for the reference saturated absorption line. The influence of the absorption and collisions on the central frequency, and the diaphragm effects across the cavity mirrors [19] are indeed not well known yet. Previous measurements on a more favourable line and with a very-well optimised optical set-up gave a repeatability a few times better [7].

## 6. Conclusion

In this paper, the absolute frequency measurement of the  $\text{CO}_2/\text{OsO}_4$  frequency standard using a femtosecond laser frequency comb is reported. Although the HeNe/ $\text{CH}_4$  infrared standard has already been measured using a frequency chain involving a femtosecond laser [20] as part of a more complicated scheme [21], this is the first frequency measurement of an infrared standard where the femtosecond laser with two flywheel oscillators forms the entire frequency chain. It requires that the frequency to be measured is less than the width of the femtosecond laser comb and with the femtosecond laser this method could be extended to frequencies up to 60 THz. To reach higher frequency, the spectrum has to be broadened in a photonic crystal fibre [12, 22, 23]. It is also the first time that the primary standard reference frequency is transmitted from another laboratory through a 43-km optical fibre.

The measurement agrees within less than  $10^{-12}$  with the previous value deduced from a measurement [15] performed with a conventional frequency chain. The  $2 \times 10^{-12}$  uncertainty of this measurement is limited by the reproducibility of the frequency standard [19], and does not correspond, by far, to the present performance of this new frequency chain. With a connection to the SYRTE Cs fountains an accuracy of about  $10^{-15}$  after 1000 s should be reached.

The short-term stability performance of this new "frequency chain" is now limited by the absolute reference frequency. So far, the transmission along the optical fibre under quiet conditions does not degrade the stability performance of the 100-MHz signal coming from SYRTE. But this 100-MHz does not exhibit yet the best frequency stability performance. A cryogenic oscillator will be used soon for short-term stabilisation [24], which will set the relative Allan deviation to less than  $10^{-14}$  between 1 and 1000 s. With the present set-up, this stability will be degraded through the optical link. Thus a correction system for the phase noise is currently under development, to improve its performance. The other crucial point is the direct synthesis from the 100-MHz signal of the 1 GHz frequency ( $\Delta_3$ ) which is necessary for the  $f_r$  measurement. Preliminary phase noise evaluation indicates that a stability of a few  $10^{-14}$  for 1 s could be reached.

This new frequency chain is currently being more severely tested through the measurement of the metrological performance of a potential new frequency standard at 28 THz, studied for several years at LPL [25]. It consists of a two-photon resonance of the SF<sub>6</sub> molecule, detected in a Ramsey set-up with 2 zones separated by 1 m. It leads to a peak-to-peak width of 100 Hz, which is two orders of magnitude narrower than the width of the CO<sub>2</sub>/OsO<sub>4</sub> reference line, and a S/N of 15 in a 1 Hz bandwidth. Thus one can expect to gain two orders of magnitude for the long-term stability and accuracy of this CO<sub>2</sub>/SF<sub>6</sub> system compared to CO<sub>2</sub>/OsO<sub>4</sub>. With the femtosecond laser comb, we shall be able to measure the expected repeatability better than 1 Hz, by a direct comparison with a primary standard of SYRTE.

Finally, this frequency chain permits ultra-precise frequency comparison between a hyperfine transition in a Cs atom and a rovibrational frequency of a molecule. Repeated measurements then offer the possibility to test for variations of fundamental constants over time [26]. These fundamental constants are not involved in the same way in these different quantum systems and this will provide an alternative test to the ones performed only with atoms. This is one of the most exciting field of investigation offered by this new facility.

#### ACKNOWLEDGEMENTS

The authors would like to thank a few members of the SYRTE for technical support and useful advices : O. Acef, S. Bize, A. Clairon, M. Lours, F. Narbonneau and D. Rovera.

The authors would like to thank the Ministère de la Recherche and the CNRS for specific funds. A.G. would like to thank University Paris 13 for its hospitality and financial support.

**Note added in proofs** : After this paper was submitted, a paper concerning an optical fiber link was published : J. Ye et al: J. Opt. Soc. Am. B 20, 1459 (2003).

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## FIGURES CAPTION

FIGURE 1 : experimental arrangement for the  $\text{CO}_2/\text{OsO}_4$  stabilization, OI : optical isolator, P : polariser, FPC : Fabry-Perot cavity, PZT : piezoelectric transducer, AOM : acoustooptic modulator.

FIGURE 2 : experimental set-up for the characterisation of the phase noise of the optical link. LPL and BNM-SYRTE are the two laboratories involved in this experiment, AM : amplitude modulation; DFB : distributed feedback.

FIGURE 3 : Allan deviation for the maser (circles) and the round-trip optical link (squares), as recorded along a period of 5 days under quiet conditions.

FIGURE 4 : Principle of the frequency measurement.

FIGURE 5 : Experimental set-up. Optical beams are symbolised with solid lines, electronic connection with dotted lines. LD : laser diodes; RF synt. : radiofrequency synthesiser.

FIGURE 6: a) Repetition rate of the femtosecond laser versus time when its 28378 harmonic is locked to the  $\text{CO}_2/\text{OsO}_4$  frequency standard. Right-hand scale is the corresponding scale for the  $\text{CO}_2/\text{OsO}_4$  frequency, obtained with  $p=28378$ ,  $\delta_1= 51$  MHz,  $\delta_2= 2.49$  MHz and  $\Delta_3=1.00123$  GHz leading to a mean value of  $\delta= 20.13392039$  kHz. b) Corresponding Allan deviation

FIGURE 7 : Frequency shift to  $\nu_{\text{ref}}$  of the  $\text{CO}_2/\text{OsO}_4$  standard

Fig. 1

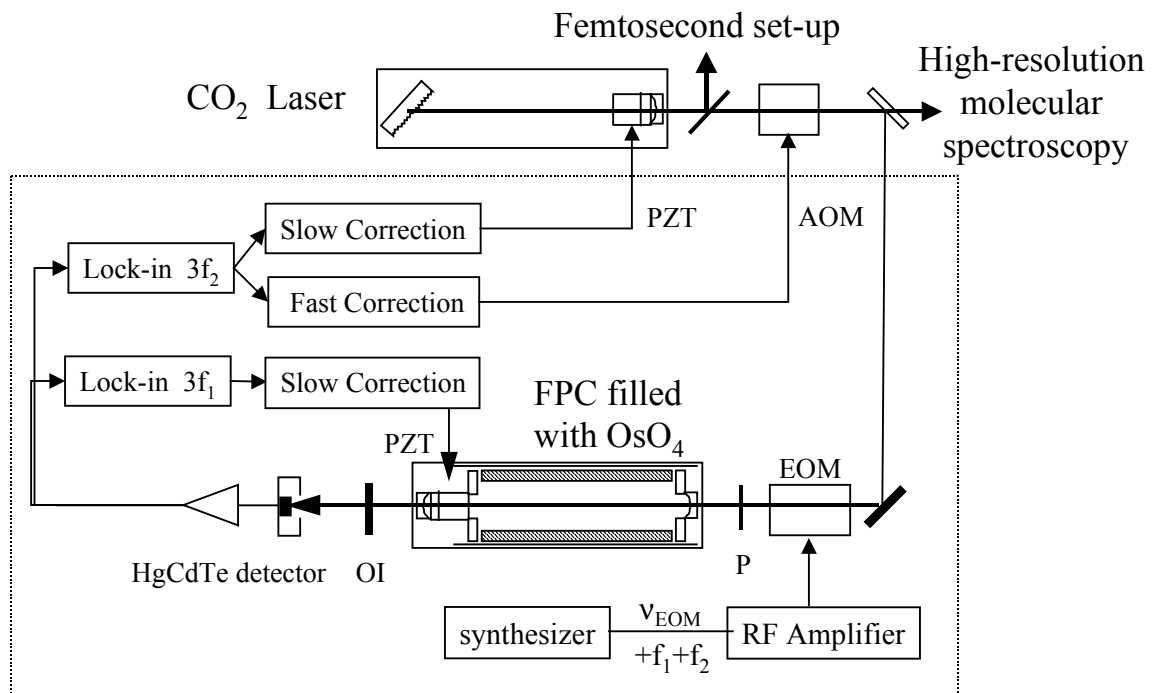


Fig 2

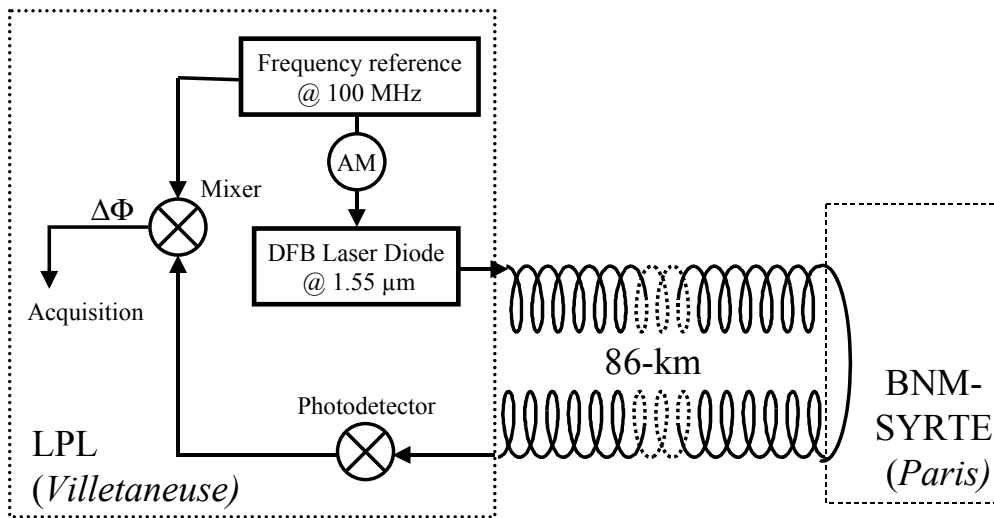


Fig 3

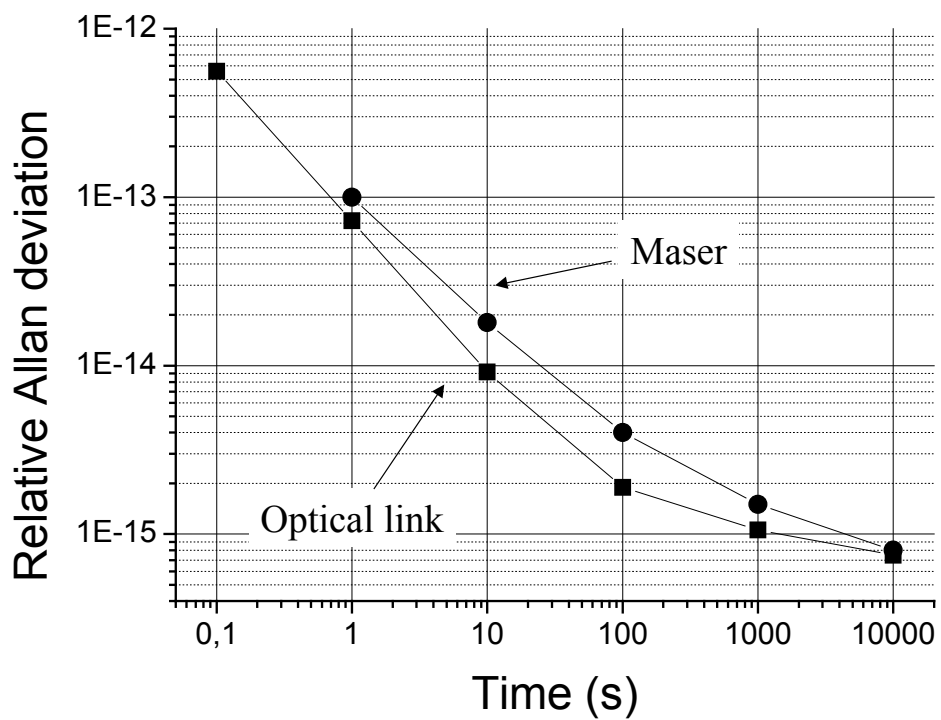


Fig 4

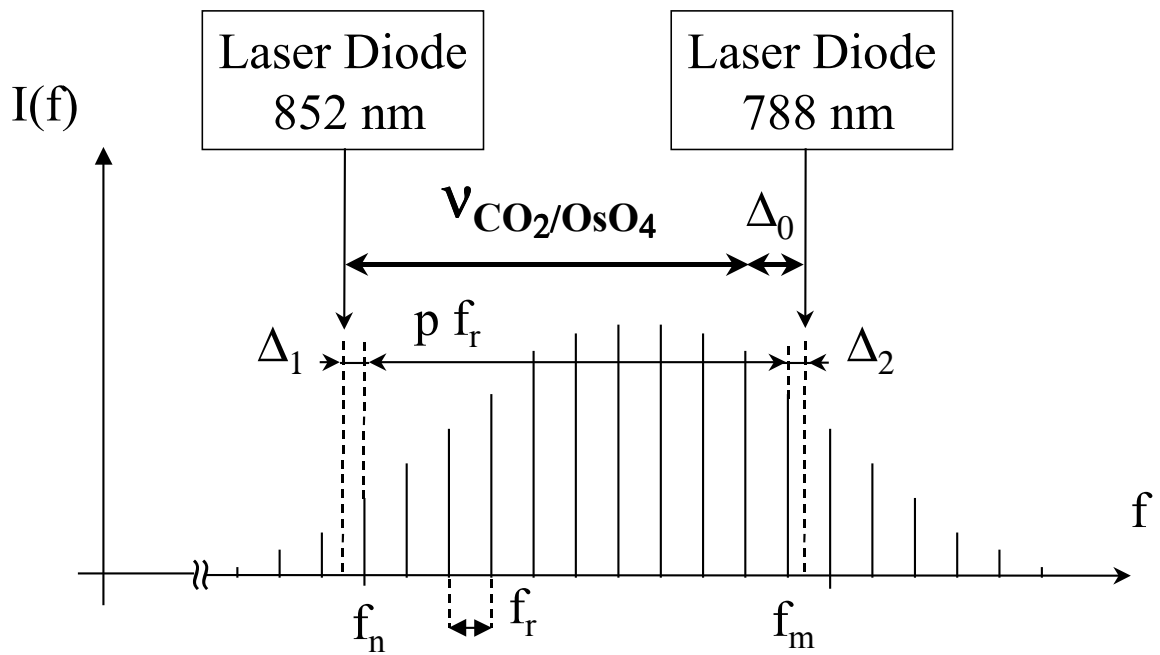


Fig 5

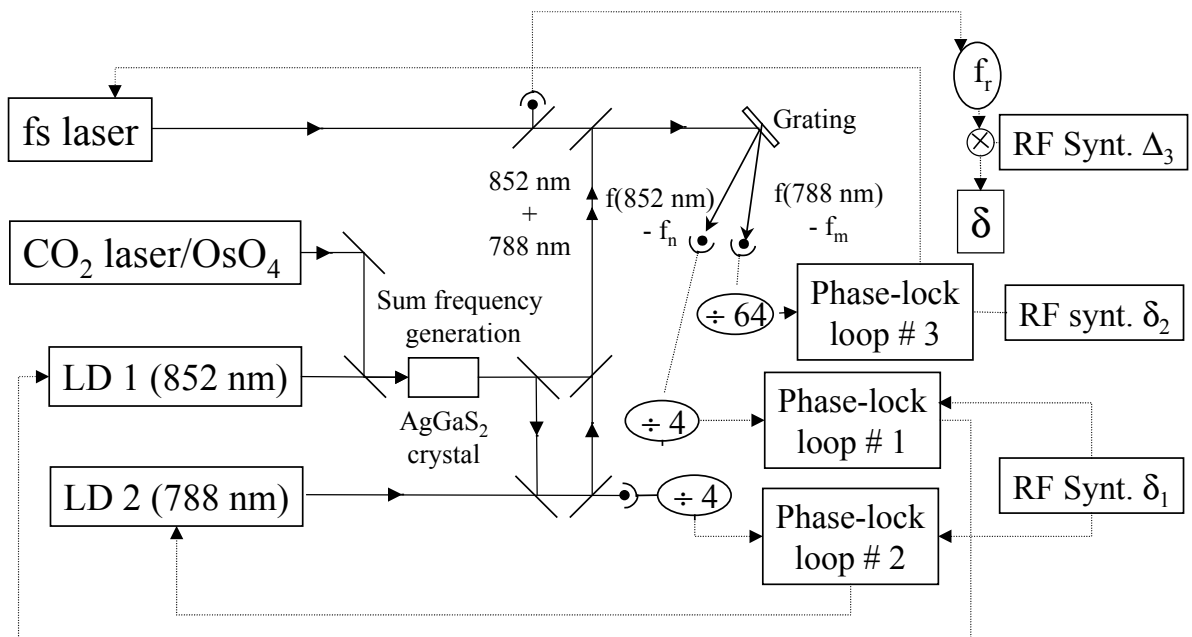


Fig 6a

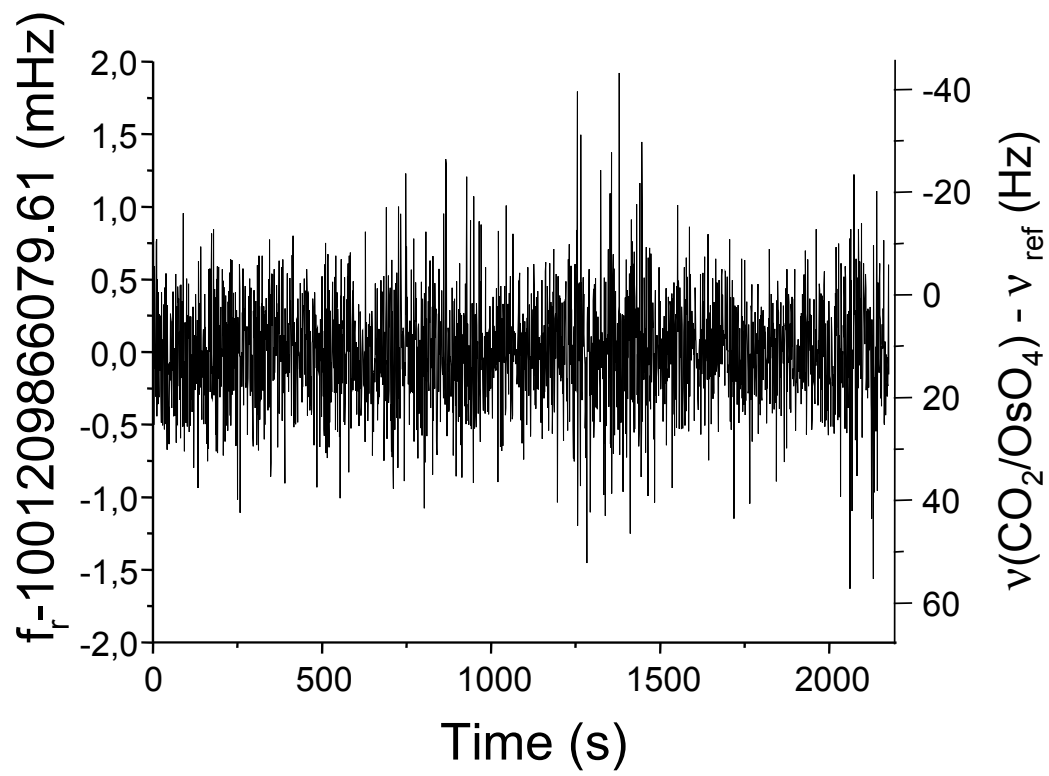


Fig 6b

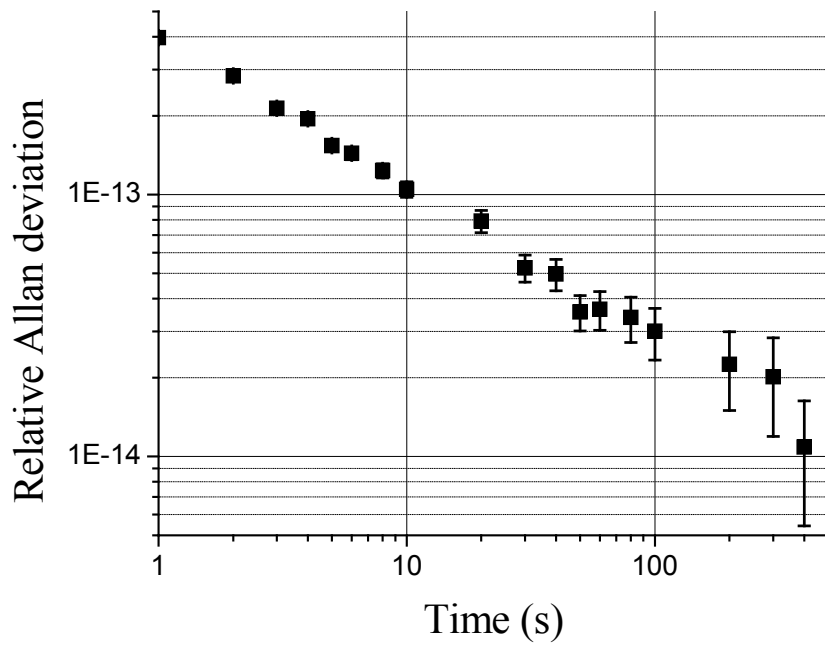


Fig. 7

