Investigation of carrier to envelope phase and repetition rate — fingerprints of mode-locked laser cavities

Ladan Arissian\textsuperscript{1,2}, Jean Claude Diels\textsuperscript{3}

\textsuperscript{1} Joint Laboratory for Attosecond Science, University of Ottawa and National Research Council, 100 Sussex Drive, Ottawa, Canada
\textsuperscript{2} Department of Physics, Texas A & M University, College Station, USA
\textsuperscript{3} Center for High Technology Materials, Albuquerque, NM, USA

Contents

1 Introduction 2

2 Defining the carrier frequency 3
  2.1 Defining the pulse 3
  2.2 Single pulses 3
  2.3 Carrier to envelope phase 4
    2.3.1 Defining CEP for ultrashort pulses 5
  2.4 Pulse train 5
    2.4.1 Pulse train in time 5
    2.4.2 Fourier Transform of the pulse train 6
    2.4.3 Time-frequency pictures 7

3 How to create a pulse/pulse train 9
  3.1 Evolution of a single pulse 10
  3.2 The laser as an orthodontist: correcting the unequal spacing of a mode comb 12

4 Mapping of mechanical perturbations on the frequency comb 14
  4.1 Constant velocity 14
    4.1.1 Doppler Picture 14
    4.1.2 Resonator picture 15
  4.2 Constant acceleration 16
  4.3 Fast changes 17

5 Measurement and control of CEP and CEO 17
  5.1 Measurement and stabilization of CEP $\varphi_e$ 17
    5.1.1 Control through group and phase delay 17
    5.1.2 Propagation of carrier-less pulses 18
    5.1.3 Measurement of CEP 19
  5.2 Measurement of the offset frequency $f_0$ 20
    5.2.1 f-2f interferometer 20
    5.2.2 Measuring CEO with a reference cavity 20
    5.2.3 Beating two pulse trains of the same repetition rate 24
### Unconventional application of CEP and CEO in mode-locked laser sensors

1. **Intracavity phase interferometry (IPI)**
   - Repetition rate control by saturable absorption
   - Cavities producing two synchronized pulse trains
   - Repetition rate control in synchronized parametric oscillators

2. **Selected examples of IPI**
   - Measurement of electro-optic coefficient
   - Measurements of small displacements

### Repetition rate spectroscopy

1. **Introduction**
2. **Coherent Population Trapping**
3. **Theory of Coherent Population Trapping by repetition rate spectroscopy**
   - Bandwidth and frequency considerations
   - Spatial effects

4. **Experimental demonstration of repetition rate spectroscopy**
5. **Implication of Dark line spectroscopy**

### Conclusion
Abstract

We use mode locked lasers in a non conventional way, as a sensor to perform intracavity measurements. To understand this new technique of Intracavity Phase interferometry (IPI), one should have a detailed look at the characteristics of the frequency comb and its sensitivity to its parent cavity. The laser cavity provides a means to perform phase interferometry, while outside the cavity one can only observe amplitude interference. Many physical quantities such as nonlinear index, earth rotation, magnetic field, Fresnel drag etc is converted to phase. IPI is performed by designing laser cavities in which two pulses circulate independently, generating two pulse trains that can have a phase difference that will be converted to frequency. We also explore repetition rate spectroscopy in $^{87}\text{Rb}$ by tailoring a laser wavelength, power and bandwidth. Coherent population trapping is observed when the laser repetition rate matches submultiples of the hyperfine splitting.

1 Introduction

Mode locked lasers are invaluable sources in biology, chemistry, and physics and had found their position in medical and industrial applications. From the early days [1] up to very recently, it was accepted that the stability in the laser repetition rate was determined by that of the radio-frequency oscillator driving a modulator to “pulse” the laser. Measurements of mutual coherence between successive pulses of a train are difficult because of the huge delay lines required, and often wrongly concluded that the coherence was lost after a few pulses. One can reasonably argue that a fs pulse circulating in a long cavity has no more coupling with the modes, hence the notion of “mode-locking” should be a misnomer. It has now been experimentally proven that a stabilized mode-locked laser produce a comb of rigorously equally spaced frequency teeth. It is now mathematically trivial to characterize the output of a mode-locked laser – assumed to have infinite coherence —by two parameters: the “Carrier to Envelope Offset” which is the frequency of the first tooth of the comb, and the “repetition rate”, which is the spacing between successive teeth of the comb. Since there is an increasing tendency to use these sources as plug and play tools, we found it instructive to look at what is imprinted on the frequency combs, in another word how a frequency comb is related to the physical parameters inside a laser cavity.

A simple model is presented for the evolution of a pulse inside a cavity, and the generation of a pulse train. In particular, the impact of mechanical fluctuations inside the cavity is investigated. Optical and electrical stabilization expanded the range of applications of mode locked sources to metrology [2]. Opto-electronic manipulation led to the same control of a waveform at an optical frequency, as is commonly realized with electronics signals. Expansion of the spectrum to a broader comb, in the time domain leads to shorter pulse, down to a few optical cycles [3, 4]. The subcycle control that has been achieved with a few cycle pulse of high peak field led to reversibility in ionization. The photoelectron ejected near the peak of an optical cycle is pulled back into the atom/molecule at the next half cycle, resulting in “ High Harmonic Generation” and attosecond science [5]. For these processes, exact waveform manipulation is essential, in particular the position of the peak field in the pulse envelope has to be accurately controlled. Measurement and control of “Carrier to Envelope Phase” ignited research in laser design [6] as well as strong field physics [7, 8].

In this paper our goal is not to discuss the stabilization [9] techniques, but to turn our attention towards the sensitivity of a mode-locked pulse train to its cavity. We believe that a laser cavity is a unique environment for making measurements. Any measurement made on the output beam of a laser, is like throwing an object in rapids, whereas in the resonator cavity which is a stable reservoir of field, ripples of a small stone can be sensed. Of course one has to walk on a tight rope between sensitivity and stability.

The paper is organized by defining first a pulse and pulse train, with pictures in time and frequency domain. The mechanism of soliton formation is shown to be responsible for making the modes equally spaced (in mode locking) in a cavity with dispersion. Then we follow the generated frequency modes in the cavity and see the imprints of the cavity fluctuations on the modes. That is what is usually suppressed [10] in stabilization.
techniques. In this paper we try to measure this fluctuations and introduce a new type of sensor class of “Intracavity Phase Interferometry” (IPI). Three optical techniques for defining the offset of the frequency comb are presented. Selected implications of (IPI) are introduced, where the laser cavity is used as a synthesizer of interwoven combs. We present another unconventional use of mode locked lasers, which is repetition rate spectroscopy. For spectroscopic applications it is always desired to have a well defined tunable frequency. Using short pulses means expanding the pulse spectrum resulting in a lower spectrum amplitude for each individual mode, and making it more difficult to isolate transitions. Even with the broadest spectrum it is impossible to extend from THz to MHz to cover both atomic and vibrational transitions. To find a middle ground we had tailored a ps pulse laser with tunable repetition rate to have high amplitude in spectrum and at the same time be a tunable RF source, which is naturally coherent with the optical frequency. Coherent Population Trapping is observed in Rb$^{87}$ when the repetition rate is a submultiple of the hyperfine splitting.

We hope that this paper offers you the opportunity to lift the cover of your laser to peak at its internal mechanism.

2 Defining the carrier frequency

2.1 Defining the pulse

The pulse is characterized by measurable quantities which can be directly related to the electric field. A complex representation of the field amplitude is particularly convenient in dealing with propagation problems of electromagnetic pulses.

The complex spectrum of the pulse $\tilde{E}(\Omega)$ is defined by taking the complex Fourier transform $\mathcal{F}$ of the real electric field $E(t) = \mathcal{E}(t) \cos[\omega t + \varphi(t)]$:

$$\tilde{E}(\Omega) = \mathcal{F} \{ E(t) \} = \int_{-\infty}^{\infty} E(t) e^{-i\Omega t} dt = |\tilde{E}(\Omega)| e^{i\Phi(\Omega)} = \tilde{\mathcal{E}}(\Omega - \omega) = \tilde{\mathcal{E}}(\Delta \Omega).$$

In the definition (1), $|\tilde{E}(\Omega)|$ denotes the spectral amplitude and $\Phi(\Omega)$ is the spectral phase. Since $E(t)$ is a real function, its Fourier transform is symmetric, and its negative frequency part can be considered as redundant information. We will therefore choose to represent the light pulse by either the positive frequency function $\tilde{E}(\Omega) = E(\Omega) e^{i\Phi(\Omega)}$ (defined as being equal to zero for $\Omega_0 = 0$) or its complex inverse Fourier transform in the time domain

$$\tilde{E}(t) = \frac{1}{2\pi} \int_{0}^{\infty} E(\Omega) e^{i\Omega t} d\Omega = \frac{1}{2} \tilde{\mathcal{E}}(t) e^{i\omega t} = \frac{1}{2} \mathcal{E}(t) e^{i[\omega t + \varphi(t)]}.$$

The relation with the real physical measurable field $E(t)$ is:

$$E(t) = \tilde{E}(t) + c.c. = \mathcal{E}(t) \cos[\omega t + \varphi(t)].$$

2.2 Single pulses

When we are dealing with a single pulse, a traditional definition of the “average frequency” $\omega$ is the center of gravity of the pulse spectrum [11]:

$$\omega = \langle \Omega \rangle = \frac{\int_{0}^{\infty} \Omega |E(\Omega)|^2 d\Omega}{\int_{0}^{\infty} |E(\Omega)|^2 d\Omega}.$$  

The time domain equivalent of this expression is:

$$\langle \Omega \rangle = \omega + \langle \varphi \rangle = \omega + \frac{\int_{-\infty}^{\infty} [\varphi(t)]^2 dt}{\int_{-\infty}^{\infty} |\mathcal{E}(t)|^2 dt}.$$  

$^1$Complex quantities related to the field will be represented with a tilde.
From this last expression one can extract a preferred decomposition of the phase factor in “frequency” $\omega$ and “slowly varying phase” $\varphi(t)$, by imposing that:

$$\langle \varphi \rangle = \frac{\int_{-\infty}^{\infty} \varphi |\tilde{E}(t)|^2 dt}{\int_{-\infty}^{\infty} |E(t)|^2 dt} = 0. \quad (6)$$

This traditional definition has been used appropriately for long, near bandwidth limited pulses. It starts posing problems for pulses of a few optical cycles. Should the “average frequency” or “carrier frequency” be defined as the frequency of the peak spectral intensity, or the center of gravity of the spectrum, or its median value? It is not useful, for instance, to define the carrier frequency of a 800 nm 6.5 fs pulse, with ten digits of accuracy, when the pulse bandwidth is $1.5 \cdot 10^{14}$ Hz (while $\omega = 2.354 \cdot 10^{15}$ s$^{-1}$). On the other hand the notion of “carrier frequency” is widely used to define a “carrier to envelope phase”, an essential characteristic in experiments of interaction of ultrashort pulses with matter.

2.3 Carrier to envelope phase

For pulses of a few optical cycles, the variation of the phase factor (chirp) can often be neglected across the pulse, and $\varphi(t) = \varphi_e$ is constant. Even for a single pulse, the phase factor $\varphi_e$ is of practical significance, when a nonlinear phenomenon traces the electric field under the envelope of a pulse of only a few cycle duration. There are numerous experiments of high field interaction which are based on the concept and control of the “carrier to envelope phase”, despite the difficulties of uniquely defining a carrier and an envelope for the ultrashort electromagnetic waves being used. Therefore, we will try to clarify the conventional definition of CEP, and attempt to define a CEP independently of the factorization of a field in harmonic wave and envelope.

If the phase $\varphi_e$ is zero, the time dependence of the electric field is symmetric, with a peak in the center at $t = 0$, larger than the two opposite maxima at $t = \pm T/2$, where $T = 2\pi/\omega$ is the light period. If the phase $\varphi_e = \pi/2$, the time dependence of the electric field is antisymmetric, with equal opposite extrema at $t = \pm T/4$. These two pulses can give a different response in highly nonlinear phenomena. Let us consider for instance a short pulse that can be generated at 800 nm, which has a Full Width at Half Maximum (FWHM) of the intensity of 2.5 light periods $T$ or $2.5 \times 2.668$ fs $\approx 6.7$ fs at a wavelength near 800 nm ($\omega = 2.354 \cdot 10^{15}$ s$^{-1}$). Its complex electric field envelope can be written as

$$E(t) = \mathcal{E}_0 \exp[-(t/2T)^2] \frac{1}{2} \left[ e^{i(\omega t + \varphi_e)} + e^{-i(\omega t + \varphi_e)} \right] = \mathcal{E}_0 \exp[-(t/2T)^2] \cos[2\pi(t/T) + \varphi_e] \quad (7)$$

which corresponds to the real electric field, and is plotted as a solid line in Fig. 1 for $\varphi_e = 0$ (left) and $\varphi_e = \pi/2$ (right). If we consider that this pulse is used to excite a 7-photon process (for instance a 7 photon ionization), the driving function for that process is the $7^{th}$ power of the field, which is plotted as a dotted line in Fig. 1. One can see that the different values of $\varphi_e$ makes a significant difference on how the process is driven. For $\varphi_e = 0$, the excitation is a single spike, as close approximation as practical to a $\delta$-function. In the case of $\varphi_e = \pi/2$ (right), the excitation consists in a succession of positive and negative spikes.

The concept of carrier to envelope phase is clouded in uncertainty for ultrashort as well as long pulses. A similar uncertainty principle applies as for the simultaneous measurement of position and momentum. For long pulses, the center of the envelope is not well defined. For ultrashort pulses, it is the average carrier frequency that cannot be easily pinpointed.

It should be noted that, in the definition (7) of the carrier to envelope phase, the pulse energy is a function of the phase $\varphi_e$. For Gaussian pulses of a few optical cycles, the pulse spectrum extends to infinity, hence to zero and negative frequencies. The combination of the positive and negative frequency components is phase dependent, hence can be constructive ($\varphi_e = 0$) or destructive ($\varphi_e = \pi/2$) interference. Such a situation will never occur in a traditional laser, because the low frequency components will scatter away. Some situation of
coherent propagation have been shown mathematically to lead to the propagation of pulses without carriers [12, 13], but such a situation has never been observed experimentally. In the time domain, the energy is

$$\int_{-\infty}^{\infty} \frac{1}{\eta} E^2(t) dt = \frac{1}{\eta} \int_{-\infty}^{\infty} \frac{1}{\eta} \mathcal{E}^2(t) \cos^2(\omega t + \varphi_e) dt = \frac{1}{\eta} \int_{-\infty}^{\infty} \frac{1}{2\eta} \mathcal{E}^2(t)[1 + \cos 2(\omega t + \varphi_e)] dt,$$

where $\eta = \sqrt{\mu_0/\epsilon}$ is the characteristic impedance of the medium. For ultrashort pulses, the pulse spectrum covers even the second harmonic frequency, and therefore, the last term of Eq. (8), which is phase dependent, may not always be neglected. In experimental situations, one starts usually with a pulse for which a carrier and an envelope can be defined. As the pulse is being amplified and compressed down to a few cycles, the “carrier frequency” is assumed to be unchanged, and applied to define a decomposition of the signal in an envelope and phase. However, taken out of context, the decomposition of the few cycle pulse in a carrier and envelope is not straightforward. This raises two basic questions:

- Can a CEP be defined, independently of a decomposition in “carrier” and “envelope”?
- Does the adjustment of CEP by introducing a group delay affect the pulse energy?

Both questions will be answered in Section 5.1.

2.3.1 Defining CEP for ultrashort pulses

In Fig. 1, the important practical implication of the zero CEP pulse is that it has the highest peak intensity. The CEP is traditionally controlled by delaying the envelope (group delay) with respect to the phase function (phase delay). For the family of pulses transformed by this dispersion to first order, we will use the phase of the highest peak field as reference $\text{CEP} = 0$. The CEP of pulses transformed by linear dispersion is defined as the phase at this peak, as it evolves through the transformation. This will be detailed in the Section 5.1 on control of the CEP.

2.4 Pulse train

2.4.1 Pulse train in time

When dealing with a pulse train — more specifically an infinite train of pulses — the notion of carrier frequency takes a different significance. This is because the extended pulse train brings with it a long time scale, hence a
tighter frequency resolution.

Let us construct a pulse train from a very short pulse defined by the electric field envelope of a single pulse $\tilde{E}(t) = E(t) \exp(i\varphi_e)$, by a carrier frequency $\omega$, and a delay $\tau_{RT}$ between successive pulses.

\[
E_{\text{train}} = \sum_{q=0}^{\infty} \tilde{E}(t - q\tau_{RT}) e^{i\omega(t - q\tau_{RT})}
\]

\[
= e^{i\omega t} \sum_{q=0}^{\infty} \tilde{E}(t - q\tau_{RT}) e^{-iq\omega \tau_{RT}}.
\]

(9)

In general, there will be a large number ($\geq 10^6$) of optical cycles (each with a period of approximately 2.5 fs near 800 nm) between successive pulses separated by $\tau_{RT}$, which leads us to write $\omega \tau_{RT} = 2N\pi - \varphi_p$, where $\varphi_p$ is the phase shift between successive pulses. Pulse $q = 0$ has thus the phase $\varphi_e$, pulse $q = 1$ the phase $\varphi_e - \varphi_p$, pulse $q = 2$ phase $\varphi_e - 2\varphi_p$ . . . Equation (9) becomes:

\[
E_{\text{train}} = e^{i\omega t} \sum_{q=0}^{\infty} \tilde{E}(t - q\tau_{RT}) e^{-iq(2N\pi - \varphi_p)}.
\]

(10)

We already see from this last expression that the value of $\omega$ needs to be established with considerably higher precision that can be measured with standard techniques in order to have a meaningful value for the pulse to pulse phase shift $\varphi_p$. Indeed, in order to determine $\varphi_p$ with a precision of $\Delta \varphi_p = 2\pi/10$, in the case of an optical pulse train at 100 MHz, the average carrier frequency $\omega$ needs to be measured with an accuracy of $\Delta \omega = \Delta \varphi_p/\tau_{rt} \approx 60$ MHz, or a relative accuracy of $\Delta \omega/\omega \approx 3 \cdot 10^{-8}$. While frequency measurement can be made with a precision of $10^{-17}$, the determination of an average carrier frequency involve finding the average of an intensity distribution, a measurement that cannot be performed with such a relative accuracy and dynamic range. Fortunately, the important quantity to define a pulse train is the change in CEP from one pulse to the next, a change that is independent of the value or definition of the frequency $\omega$. The ratio of the pulse to pulse CEP by the pulse repetition rate is the carrier to envelope offset, which is defined in the Section 2.4.2 that follows.

2.4.2 Fourier Transform of the pulse train

Let us construct the Fourier transform of the pulse train defined in Eq. (9). The Fourier Transform of the first pulse ($q = 0$) is:

\[
\tilde{E}(\Omega) = \int_{-\infty}^{\infty} \tilde{E}(t)e^{-i\Omega t}e^{i\omega t} dt = \tilde{E}(\Omega - \omega).
\]

(11)

For the $q^{th}$ pulse, delayed by $q\tau_{RT}$:

\[
\tilde{E}_q(\Omega) = \tilde{E}(\Omega - \omega)e^{-iq\tau_{RT}(\Omega - \omega)}.
\]

(12)

Summing over $q$, the infinite series of imaginary exponentials leads to $\delta$-functions:

\[
\tilde{E}(\Omega) = \tilde{E}(\Omega - \omega) \sum_{p=-\infty}^{\infty} \delta[\Omega - \omega - \frac{2\pi p}{\tau_{RT}}].
\]

(13)

Since $\omega \tau_{RT} = 2N\pi - \varphi_p$, we have a series of $\delta$-functions centered at:

\[
\Omega_m = \frac{1}{\tau_{RT}}(2N\pi - \varphi_p + 2p\pi)
\]

\[
= \frac{1}{\tau_{RT}}(2m\pi - \varphi_p).
\]

(14)
To the pulse train corresponds a frequency comb, which starts at a frequency \( f_0 = -\varphi_p/(2\pi\tau_{RT}) \), and has its teeth regularly spaced by the frequency \( 1/\tau_{RT} \). The frequency \( f_0 \) is called “Carrier to Envelope Offset” of CEO, not to be confused with the Carrier to Envelope Phase CEP (\( \varphi_e \)). The frequency comb extends to infinity. However, the amplitude of the various modes are set by the spectral envelope function \( \tilde{E}(\Omega - \omega) \), which exist only over a limited spectral range in the positive frequency space.

We note that the frequency “\( \omega \)” no longer appears in the definition of the pulse train Eq. (14). The exact value that is chosen for \( \omega \), when dealing with pulse trains, is therefore arbitrary A convenient definition for \( \omega \) is the angular frequency of the mode \( \Omega_m \) closest to the center of gravity [see Eq. (4)] of the pulse spectral envelope. Control of the CEO as treated in Section 5.2 is important when the ultimate goal is to generate single amplified pulses with controllable CEP. For these applications, a pulse train is generated such that \( f_0 = 0 \), which implies that all pulses of the train have the same CEP.

To stress the futility of trying to pinpoint the exact mode that is the “average frequency”, let us consider a train of fs pulses at 100 MHz. It requires measuring the center of gravity of a 100 THz wide spectrum, with MHz accuracy, which is beyond the dynamic range of spectral intensity measurements. It is easy to show that it really does not matter – the use of a frequency \( \omega \) is a theoretical numerical convenience, when studying the propagation of pulses longer than a few optical cycles, since it makes it possible to use a “slowly varying envelope approximation”, i.e. factor out the field evolution is a fast varying part (the optical carrier) and a slow varying part (the envelope). Such a decomposition has been widely used in linear and nonlinear optics, leading for instance in laws for the evolution of the carrier frequency and pulse envelope in the field of coherent propagation [11]. Let us consider the pulse train of Eq. (9), for which \( \omega \) was defined as the center of gravity of the pulse labelled \( q = 0 \), where the temporal pulse envelope is a Gaussian, similar to the one considered in Fig. 1. How different is the pulse if we substitute \( \omega + 2\pi/\tau_{RT} \) to \( \omega \)? The equation of the pulse at \( q = 0 \) becomes:

\[
E(t) = E_0 \exp[-(t/\tau_G)^2] \cos[\omega t + 2\pi t/\tau_{RT} + \varphi_e].
\]  

Since the range of \( t \), as defined by the Gaussian, is only \( \approx \pm \tau_G \), the effect of hopping to the next mode is a slight linear sweep in phase \( 2\pi t/\tau_{RT} \), with a maximum phase shift of \( 2\pi \tau_G/\tau_{RT} \approx 10^{-7} \) which is not a measurable phase shift. An error of \( \pm 1000 \times 1/\tau_{RT} \) would still not have any appreciable effect. Therefore, it seems that it does not matter whether we choose the carrier frequency to be anywhere between \( \pm 1000 \) “modes”. The frequency comb, as defined by Eq. (14), is completely defined by the parameters \( f_0 \) and \( \tau_{RT} \). The “average frequency” \( \omega \) is related to the pulse spectral envelope \( \tilde{E}(\Omega - \omega) \). It is however natural to choose for \( \omega \) a mode of the comb. The reason is that, in a mode-locked laser, the oscillation starts with the laser cavity mode with the highest net gain, at a frequency that will ultimately be near the peak of the pulse spectrum.

### 2.4.3 Time-frequency pictures

Let us consider an ideal frequency comb, consisting of a series of \( \delta \)-functions separated by \( 2\pi/\tau_{RT} \):

\[
\tilde{E}_{\text{comb}}(\Omega) = E_0 \sum_{p=0}^{\infty} \delta[\Omega - \frac{2\pi p}{\tau_{RT}}].
\]  

Since the comb extends to infinity, there is no particular tooth that can be called an average frequency. Each mode \( \nu_p = p/\tau_{RT} \) of index \( p \) carries the same weight, and corresponds in the time domain to an infinite cosine wave, which is a particular term of a Fourier series representation of a \( \delta \)-function. The inverse Fourier transform is also an infinite series of \( \delta \)-function, spaced by \( \tau_{RT} \). Let us now shift the frequency comb by an amount \( f_0 \): \( \tilde{E}_{\text{comb}}(\Omega) \rightarrow \tilde{E}_{\text{comb}}(\Omega - 2\pi f_0) \). The shift of the comb in the frequency domain corresponds in the time domain to a multiplication by \( \exp(i2\pi f_0 t) \). This product of the harmonic function by the comb of \( \delta \)-functions is shown in Fig. 2 (a). The carrier to envelope phase \( \varphi_e \) defined in the previous section can be
identified in the time domain, even with this train of $\delta$-functions. The phase $\varphi_e$ is identified as the phase at which each $\delta$-function crosses the harmonic field. In the sketch of Fig. 2 (a), $\varphi_e = 0$ for the first pulse, and $\varphi_p$ is then the carrier to envelope phase $\varphi_e$ of the second pulse as indicated in the figure. Note that it is only in the case of zero CEO ($f_0 = 0$) that each tooth of the time comb has the same energy. The frequency comb is shown in Fig. 2 (b). The first tooth at frequency $\nu_0 = f_0$ represents the carrier to envelope offset as defined above. Note that a zero offset frequency does not provide necessarily zero carrier to envelope phase $\varphi_e$ but the same carrier to envelope phase for each pulse of the train. The difference of group versus phase delay in a thin glass plate is generally used to adjust this phase (see Section 5.1).

Figure 2: Idealized infinite train of $\delta$-function pulses (a), and its Fourier transform (b). Since the first tooth of the Fourier comb is shifted by $f_0$, in the time domain the pulse train of identical $\delta$-functions is multiplied by a cosine function (thick lines). In (a), the carrier to envelope phase $\varphi_e$ of the first pulse is assumed to be zero.

One may at this point ponder at the physical meaning of an “infinite frequency comb” with $f_0 = 0$. Fourier transform of an infinite set of “$\delta$-functions. So far we have only envisioned pulses are being generated from a central frequency. Even the shortest pulses generated out of a Ti:sapphire had a “carrier” centered at the wavelength of 800 nm. The concept of a “soliton” pulse however does not require a carrier. Early soliton equations were derived in the context of pulse propagation in a two level system. The “area theorem” [14] stipulated that, above a certain threshold, a pulse propagating in a near resonant two-level system would evolve towards a pulse of area $\theta = \int_{-\infty}^{\infty} \kappa \mathcal{E}(t) dt = 2\pi$, where $\kappa \mathcal{E}(t)$ is the instantaneous Rabi frequency or transition rate between the two levels. If a “$3\pi$” pulse is launched into such a two-level system, it will compress, keeping its energy constant while decreasing its area to $2\pi$. Indefinite pulse compression should occur if a $3\pi$ pulse is focused with the right convergence into the medium; the pulse continuously compressing to evolve towards a $2\pi$ pulse, while the focusing maintains its area close to $3\pi$. To find the theoretical limit of such a compression, one has to solve the Maxwell–Bloch equations to second order, i.e. without making a slowly varying approximation. Eilbeck and Bullough [12, 13] have demonstrated the existence of the carrier-less solution, an electric field with a secant hyperbolic temporal shape. The experimental demonstration is a challenge that remains to be met. The approach of McCall and Hahn [14] requires propagation through “isolated” two-level systems, a physical impossibility in atomic system with a plurality of levels that enter into resonance as the pulse compresses.
The introduction of artificial two-level systems (arrays of coupled resonators [15, 16, 17]) may see a revival of attempts of pulse compression down to zero cycle.

Figure 3: (a) Train of pulses of finite duration $\tau$. The successive pulse envelope repeats every $\tau_{RT}$. Within the coherence time of the train $\tau_{coherence}$, it is the same carrier at the optical frequency that is modulated by the successive envelopes. (b) The Fourier transform of the pulse train shown in (a).

Let us consider next a train of identical pulses, not necessarily Gaussian, characterized by a Full Width at Half Maximum (FWHM) of the pulse intensity profile $\tau$. The time domain picture is sketched in Fig. 3 a. In the frequency domain [Fig. 3 b], the infinite pulse train is represented by a finite frequency comb. The envelope of the comb is the Fourier transform of the envelope of a single pulse of the train, thus of extension $\approx 1/\tau$. The teeth of the frequency comb are no longer $\delta$-functions, but sharp peaks of width $1/\tau_c$, where $\tau_c$ is the coherence time of the pulse train. The carrier to envelope phase $\varphi_p = \Delta \varphi_e$ is indicated for a pulse of the time sequence in Fig. 3 a. Note that this phase is changing from one pulse to the next. The rate of change $\varphi_p/(2\pi\tau_{RT})$ is the frequency $f_0$, which is indicated in the frequency picture by the lowest frequency tooth of the extension [dashed lines in Fig. 3 (b)].

Using the definition of $f_0 = -2\pi \varphi_p/\tau_{RT}$, the comb equation (14) can be re-written:

$$\omega_m = 2\pi f_0 + m \frac{2\pi}{\tau_{RT}}.$$  \hspace{1cm} (17)

where $\omega_m$ is the angular frequency of the $m$th mode of the comb. However, in the case of a train of pulses of finite duration, the frequency $f_0$ is no longer a real tooth of the comb, but the first mode of an extension of the frequency comb beyond the pulse bandwidth as shown by the dotted line in [Fig. 3 (b)].

3 How to create a pulse/pulse train

A mode-locked laser can be summarized as a laser cavity, in which a single pulse circulates. A pulse train results from the transmission of the intracavity pulse through an output mirror, at each round-trip. As a pulse
circulates inside the cavity, it will be shaped by various processes such as Kerr modulation, dispersion. If a train of identical pulses have to emerge through the output mirror, it is essential that the propagation of the intracavity pulse through an infinite medium representing the content of the laser cavity, leads to a steady state. In any laser, losses and gain compensate each other. The ultimate shaping mechanism in an ultrashort laser source comes from dispersive effects.

We consider first the evolution of a single pulse in an infinite medium, including self-phase modulation and dispersion. The conditions that lead to a bandwidth limited steady-state pulse will be determined. If a laser cavity contains a dispersive medium, the modes of the cavity cannot be equally spaced. In the section that follows, we will find the conditions that will lead to equally spaced modes, despite the cavity dispersion.

### 3.1 Evolution of a single pulse

When a laser is in continuous operation, the cavity gain and losses are in equilibrium. In the case of fs mode-locked lasers, the major pulse shaping mechanism is a combination of self-phase modulation and dispersion at each round-trip. The self phase modulation results from a nonlinear index of refraction $n_2$ ($I$ being the intensity in W/cm$^2$, $n_2$ being the nonlinear index in cm$^2$/W of a nonlinear element of length $\ell$ in the cavity). The dispersion $k''_\text{av}$ results from the frequency dependence of the average index of refraction $n_{\text{av}}$ defined previously, and is characterized by the second derivative of a cavity averaged $k_{\text{av}}$ vector with respect to frequency. In what follows, for simplicity, we will neglect higher order terms in Kerr effect and in dispersion. The evolution of a pulse in the mode-locked laser cavity can be considered as a propagation (of a non-diffracting beam) through an infinite lossless medium, with a positive Kerr nonlinearity ($n_2 > 0$) and a negative dispersion (as can be introduced with intracavity prisms [18] or chirped mirrors [19] in the cavity). The pulse evolution generally converges towards a steady state solution, designated as “solitons,” which can be explained as follows. The nonlinearity is responsible for spectral broadening and up-chirp. Because of the anomalous dispersion, $k''_0$, the high frequency components produced in the trailing part of the pulse, travel faster than the low frequency components of the pulse leading edge. Therefore, the tendency of pulse broadening owing to the exclusive action of group velocity dispersion can be counterbalanced. To determine the approximate parameters of that solution, let us assume a Gaussian pulse $E(t) = E_0 \exp\left(-t/\tau_G\right)^2$, and let us state that the chirp produced in the pulse center by the nonlinearity and the second order dispersion are of equal magnitude (but of opposite sign). Under this equilibrium condition the pulse circulates in the cavity without developing a frequency modulation and spectral broadening.

The effect of group velocity dispersion is to create a pulse broadening and a down-chirp (in a medium of negative dispersion). The medium of negative dispersion is characterized by a wave vector $k(\Omega)$ averaged over the cavity:

$$k(\Omega) = k_{\text{av}0} + k'_{\text{av}}(\Omega - \omega) + k''_{\text{av}} \frac{(\Omega - \omega)^2}{2} + \ldots$$  

The transmission of a pulse characterized by its spectral field, $\tilde{E}(\Omega - \omega)$, through a medium of length $P$ with the average $k$ vector of Eq. (18), is given by:

$$\tilde{E}(\Omega - \omega) e^{-ik(\Omega)P} \approx \tilde{E}(\Omega - \omega) e^{-ik''_{\text{av}} \frac{(\Omega - \omega)^2}{2} P}$$  

where we have omitted the first and second terms of the expansion, responsible respectively for a constant phase shift, and a group delay. In the case of a Gaussian pulse, initially free of phase modulation, the inverse Fourier transform of the product Eq. (19) is ([20], pages 34–35):

$$\tilde{E}(t)e^{i\omega t} = E_0 e^{-\frac{1+ic}{ct} \frac{1}{ct}} e^{i\omega t}$$  

10
where:

\[
\zeta = \frac{2k''_{av}P}{\tau G_0^2} \left( \frac{1}{1 + \left( \frac{2k''_{av}P}{\tau G_0} \right)^2} \right)
\]

\[
\tau_G = \tau G_0 \sqrt{1 + \left( \frac{2k''_{av}P}{\tau G_0} \right)^2}
\]

(21)

If a short pulse starts unmodulated through the cavity, after a round-trip (or perimeter) \(P\), it acquires the chirp (due to dispersion):

\[
\Delta \left( \frac{\partial^2 \varphi(t)}{\partial t^2} \right) \bigg|_{t=0} = \frac{4k''_{av}P}{\tau G_0^2} \left( \frac{1}{1 + \left( \frac{2k''_{av}P}{\tau G_0} \right)^2} \right)
\]

(22)

The second order dispersion \(k''_{av}\) averaged over the cavity of perimeter \(P\), can be positive or negative. We will consider here a cavity where it is negative.

Let us assume next that the cavity contains an element with a nonlinear index \(n = n_0 + n_2 I\) of length \(\ell_{Kerr}\), the phase induced by self-phase modulation, near the center of the Gaussian pulse, is:

\[
\Delta \varphi(t) = -k_{NL} \cdot \ell_{Kerr} \big|_{t=0} = -\frac{2\pi n_2}{\lambda} \ell_{Kerr} I \approx \frac{4\pi n_2 I_0 \ell_{Kerr}}{\lambda} \frac{t^2}{\tau G_0^2}
\]

(23)

where we have used a quadratic approximation for the Gaussian near \(t = 0\). Taking the second derivative yields the chirp induced by phase modulation at the pulse center:

\[
\Delta \left( \frac{\partial^2 \varphi(t)}{\partial t^2} \right) \approx \frac{8\pi n_2 I_0 \ell_{Kerr}}{\lambda \tau G_0^2}
\]

(24)

The peak intensity of the pulse (at \(t = 0\)) is \(I_0 = E_0^2/2\eta; \eta = \sqrt{\mu_0/\varepsilon}\) being the characteristic impedance of the medium. Expressing that the chirps induced by phase modulation [Eq. (24)] and dispersion [Eq. (22)] should cancel each other leads to:

\[
I_0^2 \tau G_0^2 = -\frac{\lambda k''_{av} P}{2\pi n_2 \ell_{Kerr}} \left( \frac{1}{1 + \left( \frac{2k''_{av}P}{\tau G_0} \right)^2} \right)
\]

(25)

Within the approximation that \((2k''_{av}P)/\tau G_0 \ll 1\), this expression leads to a value for the pulse duration as a function of the pulse intensity:

\[
\tau G_0^2 = \frac{\lambda |k''_{av}| P}{2\pi n_2 I_0 \ell_{Kerr}}
\]

(26)

In a Ti:sapphire laser, \(n_2 = 10.5 \cdot 10^{-16}\) cm²/W; the crystal length is typically \(\ell = 4\) mm, and a well designed laser can produce a train of 10 fs pulses with an intracavity average power of the order of 10 W. These numbers can be used in Eq. (26) to determine the negative cavity dispersion \(k''_{av}\) required for stable laser operation.

One might wonder about the stability of the soliton, which is a stationary solution of the nonlinear Schrödinger equation, derived by making a parabolic approximation for the Kerr modulation, and limiting dispersion considerations to second order. The stability of solitons have been demonstrated in various situations. In general, the effect of higher order terms that have been neglected is a perturbation that separates in time or space from the propagating soliton. For instance, spatial “Townes Soliton” get narrow in space, leaving the perturbations further away from their center [21]. Temporal solitons of self-induced transparency separate in time from any perturbation (chirp, amplitude or phase perturbation), thanks to the slower group velocity of the soliton [11, 22]. The group velocity \(|k''_{av}|\) is also an essential parameter in the present soliton representing the circulation of the laser pulse in the cavity. One would expect also higher order perturbations to separate temporally from the main soliton.
3.2 The laser as an orthodontist: correcting the unequal spacing of a mode comb

In general, the pulse train emitted by a mode-locked laser will have a non-zero carrier to envelope offset \( f_0 \). The physical reason for a non-zero \( f_0 \) is that the average group velocity \( v_g = P/\tau RT \) of the pulse in the laser cavity is not equal to the phase velocity \( c/n_{av} \) (\( n_{av} \) being the linear index of refraction averaged over the laser components). The two quantities are related by:

\[
\frac{1}{v_g} = \frac{n_{av}}{c} + \frac{\omega}{c} \left. \frac{dn_{av}}{d\Omega} \right|_\omega.
\]

Note that these quantities \( n_{av} \) and \( v_g \) are functions of the spectral frequency of the pulse. The fact that group and phase velocities are not equal implies that there is dispersion in the cavity: \( dn_{av}/d\Omega \neq 0 \). But dispersion implies also that the spacing between modes of the laser cavity, which is \( c/(2n_{av}P) \), is not equal across the spectrum. The “ideal mode-locked laser” considered in this section already poses a conceptual dilemma. Mode-locking is generally described as putting the modes of a laser cavity in phase. The unequal spacing of the cavity modes is contradictory to the fact that the frequency comb has rigourously equally spaced teeth. To resolve this apparent contradiction, we will look at the pulse train formation, and discuss how an initially irregular set of modes can lead to a perfect frequency comb.

As shown in the previous section, a minimum negative cavity dispersion \( k_{av}'' \) is required for stable mode-locked operation. Such a cavity dispersion implies that the index of refraction \( n_{av} \) is frequency (wavelength) dependent, hence the spacing of the cavity modes \( c/[n_{av}(\Omega)P] \) varies across the pulse spectrum.

The laser is in steady state, modeled by a circulating pulse, which enters a Kerr medium of thickness \( \ell \), resulting in phase modulation at each passage, and a medium that represents the linear dispersive properties of the cavity. We will assume that the balance of gain and losses maintains a constant Gaussian shape for the envelope of the circulating pulse. At each passage through the cavity, the phase of the pulse is modified in the time domain through the Kerr effect, and in the frequency domain through dispersion. We consider first the modulation in the time domain:

\[
\varphi(t) = -k_{NL} \ell_{Kerr} = -\frac{2\pi n_2 \ell_{Kerr} I_0 e^{-2(t/\tau_G)^2}}{\lambda}
\]

where \( \tau_G \) is the 1/e halfwidth of the pulse electric field envelope (the FWHM of the intensity is \( \tau_p = \tau_G \sqrt{2 \ln 2} \)). Ignoring at this point the influence of dispersion (which will be introduced after Fourier transformation into the frequency domain), the pulse train issued from the laser can be represented by:

\[
\sum_{q=0}^{\infty} \mathcal{E}(t - \tau_q) e^{-iq\varphi(t - \tau_q)} e^{i\omega t}
\]

where \( \tau_q \) is the time of arrival of the center of gravity of the successive pulses. At this point \( \tau_q \) is not set to any value. It is assumed here that at \( t = 0 \), the first pulse is unmodulated. Using a parabolic approximation for the Gaussian intensity profile, the time dependent phase is:

\[
\varphi(t - \tau_q) \approx \frac{4\pi n_2 I_0 \ell_{Kerr}}{\lambda} \left( \frac{t - \tau_q}{\tau_G} \right)^2 = \zeta \left( \frac{t - \tau_q}{\tau_G} \right)^2.
\]

The Fourier transform of the pulse train given by Eq. (29) is:

\[
\mathcal{E}(\Delta \Omega) \left[ \sum_{q=0}^{\infty} e^{i\Delta t \tau_q} e^{i\Delta \Omega^2 \tau_q^2} \right]
\]

\footnote{A fact that has been verified experimentally with mHz accuracy [23].}
\[
\Delta \Omega = \Omega - \omega
\]
\[
\mathcal{E}(\Delta \Omega) = \frac{\mathcal{E}_0 \sqrt{\pi \tau G}}{\sqrt{1 + \zeta^2}} \exp \left\{ - \frac{\Delta \Omega^2 \tau G^2}{4(1 + \zeta^2)} \right\}
\]
\[
\tau_k^2 = \frac{\zeta \tau G^2}{4(1 + \zeta^2)}.
\]

The width of the Gaussian pulse spectrum, broadened by the Kerr effect, is the inverse of the characteristic time \( \tau_k \). Let us now take dispersion into account. The operation representing the dispersion of the cavity is a product of the spectral field by \( \exp[-ik_{av}(\Delta \Omega)P] \), where \(-k_{av}(\Delta \Omega)P\) is the phase change per round-trip\(^3\).

The combined Kerr effect and dispersion, in the frequency domain, leads to the output spectral field:

\[
\mathcal{E}_{out}(\Delta \Omega) = \mathcal{E}(\Delta \Omega) \left[ \sum_{q=0}^{\infty} e^{i\Delta \Omega \tau_q} e^{iq\Delta \Omega^2 \tau_k^2} e^{-iqk_{av}(\Delta \Omega)P} \right]
\]

The Taylor expansion of the wave vector \( k_{av}(\Delta \Omega) \) to second order is:

\[
k_{av}(\Delta \Omega)P = k_{av}(\Delta \Omega = 0)P + \Delta \Omega k'_{av}P + \frac{\Delta \Omega^2}{2} k''_{av}P,
\]

where the derivatives \( k'_{av} \) and \( k''_{av} \) are calculated at the light frequency \( \omega \) (\( \Delta \Omega = 0 \)). Note that \( k'_{av} = 1/v_g = \tau_{RT}/P \) [cf. Eq. (27)] and \( k''_{av} \) are material properties independent of the index \( q \), as is the cavity perimeter \( P \). Since the laser cavity is at resonance, we must have \( k_{av}P = 2p\pi \) where \( p \) is an integer. In expressing in Eq. (29) that successive pulses are delayed by \( \tau_q \), we have already included the first order term of the series expansion of the wave vector \( k_{av}(\Delta \Omega) \), and we should have \( \tau_q = q\tau_{RT} \). The parameter \( k''_{av} \) characterizes the departure from equal spacing of the cavity modes. Substituting (34) in Eq. (33),

\[
\mathcal{E}_{out}(\Delta \Omega) = \mathcal{E}(\Delta \Omega) \left[ \sum_{q=0}^{\infty} e^{-iqk_{av}P} e^{-iq\Delta \Omega \tau_{RT}} e^{iq\Delta \Omega^2(\tau_k^2 - k''_{av}P/2)} \right].
\]

The condition:

\[
\tau_k^2 = \frac{k''_{av}P}{2}
\]

leads to modes that are exactly equally spaced, with Eq. 35 reducing to:

\[
\mathcal{E}_{out}(\Delta \Omega) = \mathcal{E}(\Delta \Omega) \left[ \sum_{q=0}^{\infty} e^{-iq\Delta \Omega \tau_{RT}} \right] = \mathcal{E}(\Omega - \omega) \sum_{p=-\infty}^{\infty} \delta[\Omega - \omega - \frac{2\pi p}{\tau_{RT}}].
\]

Equation (37) is identical to Eq. (13) derived in Section 2.4.2 for a perfect comb, with equally spaced pulses in time and frequency. In the case of small Kerr modulation, \( \zeta \ll 1 \), it can easily be verified that the condition (36) is identical to the soliton equation (26). Indeed, substituting:

\[
\tau_k^2 = \frac{\zeta \tau G^2}{4(1 + \zeta^2)} \approx \frac{4\pi n_2 P_0 \ell_K \tau_{RT}^2}{4\lambda} = \frac{k''_{av}P}{2}
\]

\(^3\)In the argument of \( k_{av} \), the light frequency \( \omega \) is taken as origin (\( \Delta \Omega = 0 \)) of the frequency scale.
which is indeed equivalent to Eq. (26). One can thus conclude that the mechanism that leads to an equal spacing for the teeth of the frequency comb emitted by the laser is the same combination of Kerr effect and dispersion responsible for creating an intracavity pulse soliton.

It should be noted that the same approximations that led to the description of a soliton propagating inside a laser cavity have been applied here. One might therefore wonder whether the approximate theory can be valid when the neglected terms are taken into account. As in the theory of the propagating soliton, the group velocity of the pulse is the essential element, which is responsible for the equal spacing of the modes of the comb. The same temporal filtering of the perturbation away from the main pulse that ensures stability of the soliton is expected to be responsible for the spectral stability of the solution presented here.

4 Mapping of mechanical perturbations on the frequency comb

So far we have been looking at a cavity with infinite coherence time and fixed perimeter $P$. The parameters $\tau_{RT}$ and $f_0$ in Eq. (17) that defines cavity modes can be influenced by thermal and mechanical changes. In a mode locked laser a packet of longitudinal dimension of 20 $\mu$m (an example of 60 fs pulse) travels back and forth in a resonator of the order of one or two meters. This light bullet likewise the continuous light would adjust its wavelength to stay a submultiple of the cavity length. Let us consider three cases of cavity fluctuations: constant velocity, constant acceleration and fast changes.

4.1 Constant velocity

For the case of general mechanical vibration, the frequency is of the order of kHz, which is five orders of magnitude smaller than the repetition rate (100 MHz) of a 1 m size cavity. For those motions the cavity drift can be considered to have a constant velocity. Here one should be aware that the round trip is the time it takes for the pulse to complete a trip in the cavity. Therefore it is related to the group velocity or speed of the envelope, while the mode frequency is related to the phase velocity. For the case of high dispersion intracavity material this distinction is inevitable.

4.1.1 Doppler Picture

When a pulse with group velocity $v_g$ hits a mirror moving at a constant mirror velocity $V$, it receives a Doppler shift. This shift will be experienced by each frequency tooth of the spectrum. The mode frequency after a single reflection $\nu'_m$ in non-relativistic Doppler is

$$\nu'_m = \nu_m \left(1 - \frac{2V}{v_p}\right),$$

(39)

where $\nu_m$ is the initial light frequency. The negative sign had been chosen for this equation, so that the frequency is downshifted for the expanding cavity and upshifted for the contracting one. We chose $m$ to be the mode closest to the pulse average frequency $\nu_{av}$. The change in mode frequency near the pulse center frequency is thus

$$\Delta\nu_m = -\nu_m \frac{\Delta P}{P},$$

(40)

where $\Delta P = 2V\tau_{RT}$ is the elongation of the cavity at each round-trip. It is interesting to compare the change in mode frequency with the change in cavity repetition rate due to the mirror motion:

$$\Delta \left(\frac{1}{\tau_{RT}}\right) = -\frac{v_g}{P^2}\Delta P = -\frac{1}{\tau_{RT}} \frac{\Delta P}{P}.$$  

(41)
The change in repetition rate frequency due to the mirror motion is thus \( m \) times smaller than the change in mode frequency (or smaller by the ratio of optical frequency to repetition rate). Finally, it remains to determine the change in carrier to envelope offset induced by the mirror motion. This can be done by writing the mode frequency after a round-trip \( \nu'_m \):

\[
\nu'_m = m \frac{v_p}{P} \left( 1 - \frac{2V \tau_{RT}}{P} \right) = f_0 + m \frac{v_g}{P} + \Delta f_0 - m \frac{v_g}{P} 2V \tau_{RT}.
\]

(42)

Subtracting \( \nu_m \) from all sides of the equalities:

\[
\Delta \nu_m = \nu'_m - \nu_m = m \frac{v_p}{P^2} 2V \tau_{RT} = \Delta f_0 - m \frac{v_g}{P^2} 2V \tau_{RT},
\]

(43)

from which we extract:

\[
\Delta f_0 = m 2V \tau_{RT} \left( \frac{v_g}{P^2} - \frac{v_p}{P^2} \right) = \nu_m \frac{\Delta P}{P} \left( 1 - \frac{v_g}{v_p} \right)
\]

(44)

The situation is similar to the one analyzed in a technique of “Cavity ring-down” for measurement of ultraslow velocities [24], with two notable differences. In the case of the passive cavity, the Doppler effect was seen as a modulation of the cavity “ring down” decay. Measurement of this Doppler effect is limited by the cavity lifetime, which is not the case in an active laser. A second difference is that the measurement of An et al is limited by cavity dispersion. We have seen that the comb corresponding to the mode-locked laser output consists of equally spaced teeth. The comb as a whole is continuously shifted by motion of a mirror, a shift that can be monitored by interfering successive pulses of the train. More complication will arise when non-successive pulses are chosen for cavity measurement. Lets assume that the mode frequency difference between the pulse associated with round-trip “i” and “i’ = i + j” is measured with a proper choice of a delay line. For the mirror of constant velocity the cavity changes by \( \Delta P = 2V \tau_{RT} \) at each round trip, so the mode shift at round trip “i’” is:

\[
\nu_m|_{i'} = \nu_m|_{i-1} \left( 1 - \frac{\Delta P}{P} \right) = \nu_m|_{i} \left( 1 - \frac{\Delta P}{P} \right)^j,
\]

(45)

for most cases of \( \Delta P \ll P \) the measured beat note is:

\[
\nu_m|_{i'} - \nu_m|_{i} \approx - j \frac{\Delta P}{P} = (i' - i) \frac{\Delta P}{P}.
\]

(46)

4.1.2 Resonator picture

Any mode in a resonator has a certain relationship with its home cavity, in order to have a constructive interference in its medium. In this picture each mode \( m \) adjusts its wavelength to be a submultiple of its cavity. The same result is realized by monitoring the change in a frequency mode \( \nu_m \) in equation (17). Note that as the cavity dimension changes both offset frequency \( f_0 \) and repetition rate \( \nu_{RT} = 1/\tau_{RT} = v_g/P \) will vary. The cavity length change is \( \Delta P = 2V \Delta t \). Rewriting the equation (17) in terms of phase velocity \( v_p \) of a frequency mode and group velocity of the pulse envelope \( v_g \)

\[
\nu_m = m \frac{v_p}{P}
\]

(47)

and the offset frequency is

\[
f_0 = m \left( \frac{v_p - v_g}{P} \right).
\]

(48)

The amplitude of mechanical vibrations is of the order of a 1 \( \mu \)m, which is 6 orders of magnitude smaller than a typical cavity length. So the changes in mode frequency and offset frequency, as the end mirror cavity
moves in a round trip to its new position \( P = P_0 + \Delta P \) with a velocity \( V \), are:

\[
\Delta \nu_m = -\frac{mv_p \Delta P}{P^2} = -\nu_m \frac{\Delta P}{P}, \\
\Delta f_0 = -m \left( \frac{v_p - v_g}{P^2} \right) \Delta P = -m \left( \frac{v_p - v_g}{P^2} \right) 2V \tau_{RT}. \tag{49}
\]

This result is based on the assumption that the phase and group velocity are the characteristics of the cavity medium, and will remain constant during the process. This assumption is valid for the case of slow and small mechanical variations.

Note that the cavity round trip \( \tau_{RT} = P/v_g \) will experience a shift of \( \Delta \tau_{RT} = \Delta P/v_g \). The relative change to the mode \( \nu_m \) is:

\[
\frac{\Delta \nu_m}{\nu_m} = \frac{\Delta P}{P} = \frac{\Delta \tau_{RT}}{\tau_{RT}}, \tag{50}
\]

implying that the relative changes in mode frequency and round trip are equal. The absolute magnitude changes in mode and repetition rate, however, are different:

\[
\Delta \nu_m = m \frac{v_p v_g}{P^2}, \\
\Delta \tau_{RT} = m \frac{v_p}{v_g} \Delta \nu_{RT}. \tag{51, 52}
\]

Constant velocity change in the cavity are not restricted to low frequency mechanical motions of the mirrors. Optical elements such as Electro Optic Modulators (EOM) can be used to modify the intracavity optical path at frequencies comparable to the repetition rate of the laser.

### 4.2 Constant acceleration

For the case of a constant acceleration with small amplitude, let us consider a cavity bounding element, such as an output coupler of a linear laser, moving with a constant acceleration \( a \). The velocity of the mirror is \( V = at + V_0 \). The shifts/round-trip in mode frequency \( \Delta \nu_m \) and carrier to envelope phase \( \Delta f_0 \) are:

\[
\frac{\Delta \nu_m}{\tau_{RT}} = -\frac{m}{P^2} (v_p)(a \tau_{RT} + V_0), \\
\frac{\Delta f_0}{\tau_{RT}} = -\frac{m}{P^2} (v_p - v_g)(a \tau_{RT} + V_0). \tag{53}
\]

There is a change in the operation frequency even for the case when the whole linear cavity experiences an acceleration. There is no need for physical contraction or expansion of the resonator, since there will be a difference between the velocity at the two end mirrors: \( |V_R - V_L| = a \tau_{RT}/2 \), where \( R \) and \( L \) are respectively the right and left mirrors, and the acceleration is from left to right. In this case

\[
\frac{\nu_{mR} - \nu_{mL}}{\tau_{RT}} = -\frac{m}{P^2} (v_p)(a \tau_{RT}/2) \tag{54}
\]

where “\( a \)” is the acceleration projected in the direction of the beam propagation, and \( \nu_{mR} - \nu_{mL} \) is the difference in frequency of the same mode for a linear cavity. We can conclude thus that a solid laser configuration is capable to identify the direction as well as the magnitude of the constant acceleration “\( a \)”.
4.3 Fast changes

Like any instrument, there is a response time for the laser mode to follow its cavity. If the cavity changes across the pulse, which is the case of a self phase modulation (SFM), or that of a resonance structure (such as atomic medium), the definition of mode frequency $\nu_m$ is not as simple as in the previous cases. The phase velocity will be different across the spectrum. In the time picture, the effective length of the cavity might vary across the pulse.

Note that there are a few time scales to be considered: the time scale of mechanical changes in the cavity, the time scale of cavity round trip, and the shortest time scale is the pulse length itself.

During its coherence time a frequency comb is well defined by two parameters of the repetition rate and the offset frequency. The laser adjust itself to its resonator, so that the absolute value of the mode varies due to mechanical vibrations and thermal drifts. Any stabilization method should be able to provide error signals and control for each of the two parameters.

5 Measurement and control of CEP and CEO

5.1 Measurement and stabilization of CEP $\varphi_e$

5.1.1 Control through group and phase delay

The carrier to envelope phase $\varphi_e$ defined in Section 2.3 becomes important in highly nonlinear experiments involving ultrashort pulses (a few optical cycles), such as high harmonic generation, attosecond pulse generation. Propagation through any material will modify this quantity. Therefore, any interaction of the pulse with matter, that is sensitive to this parameter, will be used as a measurement of the CEP at the location of the interaction.

In the absence of an “in situ” measurement, it is essential then to have a means to control the value of the CEP. The starting point is a train (of which the repetition rate may be as low as 10Hz) of ultrashort pulses with a zero CEO $f_0$, which implies $\varphi_p = 0$, and all pulses have the same carrier to envelope phase $\varphi_e$. Control of the value of $\varphi_e$ is simply achieved by transmitting the beam through a thin slab of glass [8] (thickness $d$), and varying the angle of incidence $\theta_i$. The group delay induced by propagation through a transparent medium of index of refraction $n$ for a length $\ell = d/c \cos \theta_i$ is:

$$\ell_{vg} = \frac{n}{c} \ell + \frac{\omega}{c} \frac{dn}{d\Omega} \ell = \frac{n}{c} \ell - \frac{\lambda}{c} \frac{dn}{d\lambda} \ell.$$

(55)

Since the first term in these equations is the phase delay, the second term is the difference between group and phase delay, responsible for the change in carrier to envelope phase. In order to have a range of carrier to envelope phase variation of $2\pi$, one should have a range of variation of (group minus phase) delay of one optical cycle $T = \lambda/c$. The range of optical path variation $\Delta \ell$ should thus be:

$$\Delta \ell = \frac{1}{dn/d\lambda}$$

(56)

Let us consider for instance a slab of BK7 glass ($n = 1.51078$ at 800 nm, $dn/d\Omega = 0.67 \cdot 10^{-17} \text{ s}$) of $d = 1$ mm thickness. The condition (56) for having a phase change of $2\pi$ of the CEP yields $\Delta \ell = 50 \mu \text{m}$. This corresponds for instance in varying the angle of incidence from $56.5^\circ$ (Brewster angle) to $48^\circ$. Let us consider the extreme case of a pulse with a square spectrum, centered at 800 nm ($\omega_0 = 0.75\pi \cdot 10^{15}$), extending from $\omega_{\text{min}} = 0$ to $\omega_{\text{max}} = 2\omega_0 = 1.5\pi \cdot 10^{15}$. The inverse Fourier transform of that pulse is:

$$\tilde{E}(t) = E_0 \sin(\omega_0 t) e^{i\omega_0 t}.$$

(57)
After propagation through $\ell = 13\mu m$ of glass, the phase delay is:

$$\frac{n_0\ell}{c} = 65.5 \text{fs}$$

(58)

end the group delay is

$$\frac{n_0\ell}{c} + \frac{\omega_0\ell}{c} \frac{dn}{d\Omega}\bigg|_{\omega_0} = 65.5 \text{fs} + 0.68 \text{fs} = 66.1 \text{fs}.$$  

(59)

The transmitted field is thus:

$$\tilde{E}_t(t) = E_0 \frac{\sin(\omega_0(t - 66.1))}{\omega_0(t - 66.1)} e^{i\omega_0(t-65.5)}.$$  

(60)

The difference between group and phase delay is thus 0.6 fs, or a quarter of light period, and the CEP has been changed from zero to $\pi/2$ through propagation.

5.1.2 Propagation of carrier-less pulses

As noted in Section 2.3, it becomes extremely difficult to pinpoint an “average carrier frequency” for pulses of a few optical cycles, hence the difficulty of determining a CEP. Techniques exist to measure time dependent electric fields such as the ones shown in Eq. (57), but such a shape does not lead in an unequivocal separation in amplitude and phase. It seems thus that for such ultrashort pulses, it makes sense to deal exclusively with the time dependent electric field $E(t)$, or its Fourier transform $\tilde{E}(\Omega)$, or the complex representation of the field $\tilde{E}(t)$ which is the inverse Fourier transform of the positive frequency part of $\tilde{E}(\Omega)$. If we do not define a carrier $\omega$ and envelope, the notion of phase delay $k/\omega$ and group delay $dk/d\omega$ equally disappear. We have to return to Maxwell’s propagation equation:

$$\left(\frac{\partial^2}{\partial z^2} - \frac{n^2}{c^2} \frac{\partial^2}{\partial t^2}\right) E(z, t) = 0,$$

(61)

or in Fourier space:

$$\left[ \frac{\partial^2}{\partial z^2} + \Omega^2 \epsilon(\Omega)\mu_0 \right] \tilde{E}(z, \Omega) = 0$$

(62)

or

$$\left[ \frac{\partial^2}{\partial z^2} + \frac{\Omega^2 n^2(\Omega)}{c^2} \right] \tilde{E}(z, \Omega) = 0$$

(63)

$$\left[ \frac{\partial}{\partial z} + \frac{i \Omega n(\Omega)}{c} \right] \left[ \frac{\partial}{\partial z} - \frac{i \Omega n(\Omega)}{c} \right] \tilde{E}(z, \Omega) = 0.$$

where the second order equation has been written as a product of a forward and backward propagator. The frequency dependence of the index of refraction implies that the polarization response of the medium is not instantaneous. The index of refraction increase from unity to the value of glass in the rising edge of the pulse may be the source of a self-induced reflection. Neglecting this loss, since we are only interested in the forward propagating wave, and expanding the index in series to first order:

$$n(\Omega) = n_0 + a\Omega$$

(64)

the solution of Eq. (63) is simply:

$$\tilde{E}(z, \Omega) = \tilde{E}(0, \Omega)e^{i(-\frac{\omega_0}{2}t - \frac{a}{2}n^2 t^2)}$$

(65)
Let us consider the same numerical example as in the previous section. For the Taylor expansion of the index of refraction we have now: \( n(\Omega) = 1.495 + \Omega \times 0.67 \times 10^{-17} \). The Fourier transform of the pulse propagated through 13 \( \mu \)m of BK7 glass is now:

\[
\tilde{E}(\ell, \Omega) = E(0, \Omega)e^{-i(64.8\Omega+0.29\Omega^2)}
\]  

(66)

The temporal electric field, found by a numerical inverse Fourier transform, is shown in Fig. 4 (solid blue line) and compared to the slowly varying envelope approximation of the previous section. As expected, the results are similar but not identical. Unlike the standard approach of Eq. (59) leading to the delayed pulse of Eq. (60), the propagator in Eq. (66) is completely independent of a description of the pulse in phase and amplitude. A better approximation would be to use the complete frequency dependence of the index approximation, rather than a linear fit, and to solve Maxwell’s propagation equation to second order. When dealing with few cycle pulses, the notion of CEP should be replaced by a full characterization of the pulse electric field. The pulse shape is modified in a deterministic fashion by the amount of glass traverse, which is the primary parameters in the experiment of Paulus et al [8]. A new definition of the CEP is the difference between the value of the phase of the complex electric field at the peak of its amplitude, and the value of the phase taken at the peak of the real field.

Figure 4: Real electric field after a propagation distance of 13 \( \mu \)m. The dashed red curve is the result of the decomposition of the field in envelope and carrier, hence the real part of Eq. (60). The solid blue curve is the real part of the inverse Fourier transform of Eq. (66), obtained from the direct application of Maxwell’s equation of the electric field of Eq. (57).

5.1.3 Measurement of CEP

As mentioned above, a determination of the CEP results from interpretation of a high order nonlinear interaction of the pulse with matter, of which the outcome can be correlated to the value of the CEP. An example of such an experiment is multiphoton ionization of xenon atoms [8] with pulse intensities of the order of \( 10^{14} \) W/cm\(^2\). The phase dependence of this process is the highest in the “above threshold ionization” (ATI — the number of photons absorbed being larger than 8 for Xe). The experiment consists in collecting the electrons created by photoionization with two time-of-flight spectrometers, facing each other along the polarization direction of the few cycle optical pulse. The electrons collected have energies ranging from a few eV to 55 eV. The CEP dependence for low energy electrons is as expected dominated by a high power of the field. The phase \( \varphi_e = 0 \) will produce more electron towards the positive direction (Fig. 1, left). A stronger, but shifted by \( \pi/2 \), phase dependence is observed for high energy (> 20 eV) electrons. The mechanism is acceleration of the electrons toward the ionized atom by the optical field that has reversed sign at the half cycle that follows.
the time of ejection. The ratio of high energy electrons collected by the opposing time-of-flight spectrometers
varies between 0.3 to 3. depending on the CEP phase. The largest number of high energy electrons is produced
in the direction/phase for which the field has two nearly equal and opposite intensity peaks (the case of Fig. 1,
right).

5.2 Measurement of the offset frequency $f_0$

We will consider here three different techniques leading to the measurement of the carrier to envelope offset $f_0$.
The first method, most traditional, is called f-2f interferometry. It is an absolute method, and does not require
any reference. However, it applies only to ultrashort pulses of which the bandwidth covers at least one octave.
The second method requires a reference cavity. It applies well to pulses of a few 100 fs, down to practical limits
of maintaining a dispersionless cavity (implying dispersionless mirror reflectivity) over the pulse bandwidth. A
third method is to beat the pulse train with a reference pulse train, of known $f_0$, with the same repetition rate as
the laser under measurement. It applies to any pulse duration.

5.2.1 f-2f interferometer

The most popular technique [25] is based on the beating of a group of short wavelength modes (frequency $\nu_{2m}$),
with the second harmonic of a group of long wavelength modes (frequency $\nu_m$) of the frequency comb. This
beat frequency is:

$$2\nu_m - \nu_{2m} = 2f_0 + 2m \frac{1}{\tau_{RT}} - \left[ f_0 + 2m \frac{1}{\tau_{RT}} \right] = f_0.$$  \hspace{1cm} (67)

This method requires thus to have enough power in these two groups of modes of the comb, separated by a
frequency span equal to $\nu_m$, to measure a beat note. It is therefore ideally suited for pulses of a few optical
cycles. Longer pulses (pulse duration less than 100 fs) can also be measured by this technique, provided the
frequency comb is extended by a photonic crystal fiber. The experimental set-up is sketched in Fig. 5.

There are some experimental challenges associated with such a measurement. The beat frequency to be
measured is generally weak. A phase-locked loop is often used to extract this signal from the noise.

5.2.2 Measuring CEO with a reference cavity

A reference Fabry-Perot cavity can provide a means to measure the carrier to frequency offset (CEO) $f_0$ [10].
Let us assume a perfect reference cavity with no dispersion. Such a cavity should be in vacuum (to avoid
the dispersion of air), with broadband dispersion-free reflectors. The transmission modes of such a cavity are
sketched in Fig. 6. Consistent with the approximation of no dispersion, the extension of the modes of this cavity
is zero, and the modes have all equal spacing $1/\tau_0$. Let us consider a fs laser, of which the end cavity mirror is
mounted on a piezoelectric. If $P$ is the perimeter (or round-trip $2L$) of the laser cavity, the mode equation can
be written as:

$$\nu_q = q \frac{v_p - v_g}{P} + q \frac{v_g}{P} = f_0 + q \frac{1}{\tau_{RT}}.$$  \hspace{1cm} (68)

where $v_p = c/n$ and $v_g$ are the group and phase velocities spatially averaged over the cavity. For the passive
cavity, since we have assumed a perfect dispersionless cavity:

$$\nu_q = q \frac{1}{\tau_0}.$$  \hspace{1cm} (69)

where the round-trip time $\tau_0$ of the reference cavity will be chosen as a reference. For $P = P_0$ (of the laser
cavity), the round-trip time of the laser cavity is equal to that of the reference cavity, i.e. $\tau_{RT} = \tau_0$. The two
Figure 5: The beam from the laser source enters a microstructure fiber, where the spectrum is expanded. For pulses shorter than 100 fs, this broadened spectrum conserves the mode-structure of the generating comb. The spectrum exiting the fiber is collimated by a lens $L_2$, passes above a mirror $M_1$, and is dispersed by a first prism to separate the long and short wavelengths parts of the spectrum. After passing through a second prism, each part of the spectrum is retro-reflected, and sent to mirror $M_1$. A pinhole $P_1$ selects the appropriate part of the short wavelength continuum. Mirror $M_0$ is on a translation stage, to adjust the temporal coincidence of the two interfering beams. A dichroic beam splitter sends the long wavelength part of the beam into a frequency doubling crystal type I. The second harmonic, cross polarized with the fundamental, is combined with the short wavelength beam by a polarizing beam splitter. A polarizer selects equal component of $\nu_{2m}$ and $2\nu_m$ to interfere in an avalanche photodetector $APD$.

Figure 6: Top: modes of a dispersionless Fabry-Perot cavity. Bottom: modes of the pulse train of the laser. Solid black lines: the repetition rate of the laser is at exact resonance with the Fabry-Perot cavity, but since $f_0 \neq 0$, there is no transmission. By shortening the laser cavity by an amount $\Delta L$, the mode spacing increases (red dashed lines), so that the central mode comes at resonance with a mode of the Fabry-Perot.

mode combs do not overlap, they have the same mode spacing but are shifted with respect to each other by the
frequency $f_0$. The electric field of the pulse train incident into the Fabry Perot was given in Eq. (13):

$$\tilde{E}_{\text{inc}}(\Omega) = \tilde{E}(\Omega) \sum_{q=-\infty}^{\infty} \delta[\Omega - 2\pi \nu_q]. \quad (70)$$

For each value of the perimeter $P$, we have a mode comb (68), which we insert in the definition (13), and calculate the transmission from:

$$\tilde{E}_{\text{transm}}(\Omega) = \tilde{E}_{\text{inc}}(\Omega) \frac{(1 - R)e^{-i\Omega \tau_0}/2}{1 - Re^{2\pi \nu_q \tau_0}} \quad (71)$$

where, consistently with the approximation of a perfect dispersionless cavity, we assume that the phase shift upon reflection is negligible. After substitution, we find the complete expression for the transmission of any mode $q$ of the comb by the Fabry-Perot:

$$\tilde{E}_{\text{transm}}(q) = \tilde{E}(2\pi \nu_q) \frac{(1 - R)e^{-i\pi \nu_q \tau_0}}{1 - Re^{2\pi \nu_q \tau_0}} \quad (72)$$

where

$$\nu_q = f_0(P) + q \frac{1}{\tau_{RT}(P)} \quad (73)$$

$$f_0(P) = q \frac{v_p - v_g}{P}$$

$$\frac{1}{\tau_{RT}(P)} = \frac{v_g}{P} \quad (74)$$

The measured signal is the total transmitted energy $W(P)$ for each value of the perimeter $P$:

$$W(P) = \sum_{q=0}^{\infty} \left| \tilde{E}_{\text{trans}}(q) \right|^2 \quad (75)$$

The calculation outlined above yields the transmission of the Fabry-Perot to the pulse train, as function of the cavity perimeter $P$. An experimental result and a computer simulation are plotted in Fig. 7.

It should be noted that there is no need to extend the “mode comb” to zero frequency to define CEO $f_0$, which exists for any pulse train. The CEO is simply the product of the difference between the group and phase delay, multiplied by the frequency $\omega_0$, divided by the round-trip time, and can be defined for the reference cavity at any frequency:

$$f_0 = \frac{\left( \frac{P}{v_g} - \frac{P}{v_p} \right) \omega_0}{2\pi \tau_{RT}} \quad (76)$$

Instead of the rigorous approach outlined above, it is possible to estimate the position of the peaks in Fig. 7 through linear approximations. Let us consider the mode $N$ in Fig. 6 of the laser, which is nearest to the peak of its spectrum. If the cavity perimeter is reduced by $\Delta P$, the frequency of this mode $N$ will upshift by an amount $\Delta \nu_N$ according to:

$$\Delta \nu_N = N \frac{v_p - v_g}{P_0^2} \Delta P + N v_g \frac{\Delta P}{P_0^2} = f_0 \frac{\Delta P}{P_0} + N \frac{1}{\tau_{RT}} \frac{\Delta P}{P_0}. \quad (77)$$

For the value $P = P_0$, the repetition rate of the laser is the same as that of the reference cavity $\tau_{RT} = \tau_0$. If $f_0 = 0$, there is perfect mode overlap across the whole spectrum, and the transmission is unity. If $f_0 \neq 0$, there
Figure 7: a)(experimental result) Transmission through a Fabry-Perot (60 cm) half of the length of the laser cavity, as a function of the elongation of the laser $\Delta P$ (one end mirror of the laser cavity is controlled by a piezo-element). The pulse duration is 50 fs. The axis of symmetry of the curve tracing the peak transmission of the modes corresponds to a perfect group velocity match between the laser and Fabry-Perot, and is taken as zero reference for the displacement $\Delta P$. The spacing between transmission peaks correspond to $1/\tau_0$, the round-trip rate of the reference cavity. If the reference cavity has no dispersion, the abscissa of the first transmission peak corresponds to $f_0$ (from [10]). b)(simulation) Transmission of 180 fs pulse train through a reference cavity half the size of the linear laser cavity (13.2 cm). The mode comb has offset frequency of $f_0 = 0.49\nu_{RT}$.

is no mode overlap between the pulse train and those of the reference cavity, hence the transmission is zero. Transmission will occur for a cavity change $\Delta P_1$ such that $\Delta \nu_N = f_0$ (dashed red lines in Fig. 6):

$$f_0 \left(1 - \frac{\Delta P_1}{P_0}\right) - \frac{N}{\tau_{RT}} \frac{\Delta P_1}{P_0} = 0$$

from which we extract

$$\Delta P_1 = -\frac{P_0 f_0}{\nu_N} = -\frac{P_0 f_0}{f_0 + \frac{N}{\tau_{RT}}}$$

(79)

For the next resonance, we need $\Delta \nu_m = f_0 + \frac{1}{\tau_{RT}} = f_0 + \frac{\nu_p}{P_0}$. For the resonance number $j (j = 0, 1, 2... - 1, -2, -3...)$:

$$\Delta P_j = -\frac{P_0 \left(f_0 + \frac{1}{\tau_{RT}}\right)}{f_0 + \frac{N}{\tau_{RT}}}$$

(80)

The transmission is less in higher order overlaps, because the chance of overlap decreases as the mode spacing between the reference cavity and the laser cavity is tuned off-resonance. The first order transmission itself is not unity since $\Delta P \neq 0$. The envelope of the Fabry-Perot transmission versus $\Delta P$ has its axis of symmetry at $\Delta P = 0$ displaced with respect to the first mode transmission by the amount given in Eq. (79).

The change in $P$ corresponding to a mode spacing is:

$$\Delta P_{mode} = \Delta P_{j+1} - \Delta P_j = -\frac{P_0 \frac{1}{\tau_{RT}}}{f_0 + \frac{N}{\tau_{RT}}} = -\frac{P_0}{\tau_{RT} \nu_N} = -\lambda \frac{\nu_p}{v_p}.$$ 

(81)

It is not exactly one wavelength, because of the difference between group and phase velocities in the laser cavity. It follows from Eqs. (79, 81) that the ratio $\Delta P_1/\Delta P_{mode}$ is also the ratio of the CEO to the mode.

23
\[ \frac{\Delta P_1}{\Delta P_{\text{mode}}} = f_0 \tau_{\text{RT}} = \frac{f_0}{\nu_{\text{RT}}}, \]  

which is clearly visible in Fig. 7(b) for the case that \( f_0 = \nu_{\text{RT}}/2 \). In conclusion, the Fabry-Perot cavity allows for an easy measurement of the CEO \( f_0 \), provided the reference cavity has been well characterized. It is not necessary to have a Fabry-Perot with zero CEO. In the case of a non-zero CEO for the reference cavity \( f_{\text{ref}} \), the measurement of Fig. 7 provides the laser CEO relative to the reference cavity. The laser pulse train in that case has a CEO of \( f_0 + f_{\text{ref}} \).

### 5.2.3 Beating two pulse trains of the same repetition rate

A third method in measuring CEO is to beat a pulse train to a reference pulse train of known CEO \( f_{\text{ref}} \) and the same repetition rate, as sketched in Fig. 8. A delay line is used to ensure that every successive pulse of one pulse train meets its correspondent of the other train on a detector. Since the teeth of the frequency comb are equally spaced, the two frequency combs are translated with respect to each other by the frequency \( f_0 - f_{\text{ref}} \). This frequency is the lowest frequency component of the beat note spectrum. This is the only method that is applicable for all pulse durations. We will show in the following section that such an elementary measurement is the key to a new family of laser sensors based on intracavity phase interferometry (IPI).

### 6 Unconventional application of CEP and CEO in mode-locked laser sensors

#### 6.1 Intracavity phase interferometry (IPI)

We have seen in the previous section how the interference of two pulse trains of the same repetition rate gives a direct measurement of the CEO. A whole new class of laser sensors is based on the ability to

- Create such pulse trains of identical repetition rate inside a laser cavity
- Make the CEO of these pulse trains proportional to a physical parameter to be measured (angular velocity, acceleration, Fresnel drag, magnetic field, linear and non-linear index of refraction, elongation etc...)

A key question is thus: which cavity element, or laser design, can result in producing two synchronized pulse trains? Two solutions to this problem will be presented in the following subsections: repetition rate control by saturable absorption and repetition rate control in synchronized parametric oscillators. Stabilizing the round-trip time inside a laser, without acting on the other parameter that defines the output pulse trains, is a key issue that will be addressed again in Section 7.1 dealing with coherent interaction inside a laser cavity.
6.1.1 Repetition rate control by saturable absorption

Any mode-locked laser requires some mechanism of amplitude modulation to initiate a circulating pulse in its cavity. This mechanism can be saturable absorption or Kerr lensing [20]. It is little known that this type of intensity dependent loss is responsible for locking the average group velocities of two intracavity pulses to each other, leaving the carrier frequencies uncoupled.

To appreciate this envelope coupling, let us consider a saturable absorber of smaller longitudinal dimensions than the optical pulse, as sketched in Fig. 9. In the figure, the meeting point of the two pulses is on the left of the saturable absorber. Therefore, the pulse $A$ coming from the right enters first the absorber, and its leading edge is attenuated. The absorption is saturated when the two pulses meet in the absorber. When two pulses of equal intensity counterpropagate in a saturable absorber, the effective saturation intensity is reduced by a factor 3 [20]. The pulse $B$ coming from the left is still partly in the absorber when pulse $A$ has left the absorber. Therefore, its tail will be more absorbed. The net effect is a shift of the center of gravity of both pulses, such that at the next round-trip they will meet closer to the middle of the absorber.

\[ n_i = n_2 (I_i + 2I_j), \]

where $I_i$ and $I_j$ are pulse intensities in corresponding directions. The fact that the temporal envelopes are coupled in no way implies that the spectral envelopes are coupled. In fact, in the case of this particular Kerr-lensing mutual coupling, the spectral envelopes of the two pulse overlap only partially. The beat note observed between the two trains of pulses is still due solely to the mode splitting.

6.1.2 Cavities producing two synchronized pulse trains

Whether with saturable absorber or with Kerr lensing, the configurations are as sketched in Fig. 10. The cavity can be linear [in (a)] or ring [Fig. (b)]. In both cases, $A$ is either a saturable absorber, dye jet or multiple quantum
well, or a nonlinear crystal such as ZnS., which favors mode-locked operation with two pulses crossing at the location $A$.

Figure 10: (a) Linear cavity with a nonlinear (saturable) absorber $A$ in the middle. The configuration of minimum losses is that with two pulses circulating symmetrically in that cavity, meeting in the middle. The output interferometer is shown, leading to a beat note as an envelope of the pulse train. (b) Ring configuration, where the two pulses are circulating in opposite directions. $G$ is the gain medium, $P$ dispersive prisms, $I$ iris aperture, $O$ output coupler.

In the case of the linear cavity of Fig. (a), the crossing point of the two circulating pulses is set by the saturable absorber to be in position $A$, in the middle of the cavity. The sample $S$ to be measured is an element that produces a different phase shift for the “red” pulse (shown on the left) as compared to the phase shift for the “blue pulse” on the right. Examples are measurement of the nonlinear index (differential phase shift dependent on the difference of intensity between the two pulses), electro-optic coefficient (Section 6.2.1), or displacements (Section 6.2.2). The output coupler is a beam splitter that deflects the “red” pulse upwards towards an optical delay line, the “blue” pulse downwards. After an appropriate optical delay, the output red pulse is combined with the blue one on a 50% beam splitter, and the interfering signals are sent to a detector where the beat note is recorded. The frequency of this beat note is the phase shift produced on the “red” pulse divided by the round-trip time. Beat frequency bandwidths of less than 1 Hz have been observed, implying a phase resolution of $\Delta \phi = (1 \text{Hz} \times 2 \pi \tau_{RT} \approx 10^{-7}$.

For the case of the ring laser, two pulses, represented as “red” and “blue”, are circulating in opposite sense in the ring cavity, and meeting at the saturable absorber $A$. The two outputs at the output coupler $O$ are delayed with respect to each other (delay line not shown) before being recombined via a beam splitter, and sent to a detector recording the beat note. The sample $S$, responsible for a different phase shift for the “blue” and red pulses, is located 1/4 cavity perimeter away from the pulse crossing point $A$, in order to be traversed at equal time intervals by either pulses. An example of application where the ring cavity is advantageous is the measurement of magnetic fields [9]. For such a measurement the sample $S$ is a transparent material (preferably with high Verdet constant such as TGG) between two quarter wave plates. The linear polarization of the counter circulating beams is converted in opposite circular polarization. In presence of a magnetic field along the $k$ vector of the light, there is a different index of refraction for the two sense of circular polarization. As a result of the different phase shift for the counter- and clockwise circulating pulses, a beat note is recorded, proportional to the magnetic field. We demonstrated recently a sensitivity of 30 nT, using for $S$ a 10 mm long sample of TGG crystal. In the case of the ring laser, $S$ can represent a flowing gas (phase shift and beat note resulting from Fresnel Drag [28]). or even be absent (phase shift between counter-circulating pulses resulting from rotation [29]).

6.1.3 Repetition rate control in synchronized parametric oscillators

Flowing dye jets are not the most popular device. A synchronously pumped parametric oscillator is another means to create a set of two pulses circulating in a cavity with the same repetition rate. In such a device, the gain exists only for the duration of a pump pulse, hence the repetition of the pulse(s) generated in the signal
cavity is locked to that of the pump laser cavity. Two types of possible cavities are sketched in Fig. 11, in (a) a mode-locked lasers pumps an OPO cavity twice as long, and in (b) the OPO crystal share the pump and OPO cavities (both of equal length). In the extracavity pumped OPO, the two signal pulses are separated by the pump pulse cavity round-trip length, which is equal to the length of the signal cavity. Therefore, the two intracavity signal pulses will hit the opposite ends of the cavity at the same time, as in the case of the cavity of Fig. 10 (a). Application to intracavity measurements imply the insertion of the same sample $S$ to be measured (to create a differential phase between the two pulses) and an output coupling with interferometric delay line as shown in Fig. 10 (a) [not reproduced here in Fig. 11 (a)]. In this configuration (a), there is less pump energy (hence less gain) available in the OPO crystal, but it has the advantage over (b) that the two cavities — pump and signal — are not coupled. In order to extract more efficiently power from the pump laser (hence have larger gain), the

Figure 11: Examples of cavities with two pulses circulating in them. (a) Extracavity synchronously pumped OPO. The signal cavity has twice the length of the pump cavity. $T$ is the Ti:sapphire gain medium, $P$ dispersive prisms. (b) Ti:sapphire linear laser pumping synchronously an intracavity OPO [30]. A PPLN crystal shares the pump and signal (ring) cavity. Two pulses are generated for each pulse in a round trip of Ti: Sapphire due to double passage in PPLN. The signal cavity has the same length as the pump cavity.

OPO crystal can be pumped inside the pump cavity as in Fig. 11 (b). Another advantage of this configuration is that it can be used to pump a ring laser as shown in the figure. A pump pulse traverses the OPO crystal from right to left, generating a signal pulse that circulates clockwise in the ring cavity. After hitting the end mirror $M$ of the pump cavity, the pump pulse traverses again the crystal, generating a signal pulse that circulates counter-clockwise in the ring. The crossing point of the two pulses in the signal cavity can be controlled by adjusting the position of the end mirror $M$. The two intracavity pulses are extracted through the output mirror $O$, and made to interfere on detector $D$, after being given appropriate delay in an interferometric delay line.

The intracavity pumping satisfies the important requirement that the two pulses of the OPO cavity share the same spatial mode. Even though its operation has been demonstrated [30], stable operation of an OPO pumped intracavity by a mode-locked Ti:sapphire laser is very difficult to implement. It has been demonstrated that a Q-switched-mode-locked tendency of the Ti:sapphire is enhanced by coupling with the OPO cavity. In addition, there is a strong tendency for unidirectional operation. Both instabilities are rooted in the coupling between pump and signal through depletion of the pump pulse inside its cavity. These instabilities can be corrected by inserting a strong negative feedback in the OPO cavity, which prevents the energy of a circulating pulse to grow excessively, depleting the pump, or grow at the expense of the other OPO pulse. A second harmonic crystal (approximately 1% conversion) inserted in the cavity provides the desired negative feedback [31, 32, 33].

One important point in both of the configurations of Fig. 11 is that the group velocity in the pump cavity controls solely the repetition rate of both pulses, independently of the length of the signal cavity. In turn, the CEO of the signal pulse trains is solely determined by the signal cavity, hence the beat note which is the differential CEO is solely controlled by the sample element $S$, and is independent of the pump cavity. A synchronously pumped OPO has the unique property that the controls of repetition rate and CEO are perfectly
orthogonal, an important advantage when attempting to stabilize the pulse train. In the section 7.1 that follows, a method of locking the repetition rate of a laser to an atomic transition will be discussed.

6.2 Selected examples of IPI

The sample $S$ in Figs. 10 determines the nature of the measurement, by converting a physical parameter (flow velocity, electric field, magnetic field, rotation, displacement) into a phase shift for one of the two intracavity pulses. Whether to use a ring or linear cavity depends on the quantity to be measured. The ring laser is sensitive to rotation [29], and to Fresnel drag in one of its arms [28]. Without any rotation or modulation a mode-locked ring laser has a beat frequency offset of at least 100 Hz and often as high as 100 kHz [26]. This is a result of the asymmetry in the CW and CCW pulse. Due to presence of nonlinear intracavity elements, the order in which the pulse encounters the optical elements will affect the pulsewidth and pulse amplitude [34, 35, 20]. Since the pulses in a linear cavity travel through the same optical elements in the same order, there is no asymmetry. Therefore one advantage of a linear cavity versus a ring geometry is the absence of natural bias. In the ring geometry, any variation in pulse amplitude or pulsewidth will result into a beat signal. Monitoring the differential pulse phase in a laser cavity, an IPI can be exploited to measure a nonlinear index of refraction [36], magnetic fields [37] small displacements [38] and electro-optic effects [39]. The latter two applications will be presented in the following two subsections.

6.2.1 Measurement of electro-optic coefficient

Measurements of the electro-optic coefficient has been performed successively with the linear and ring lasers of Fig. 10 as well as the linear synch pumped OPO of Fig. 11 (b). In the case of the linear lasers, the RF signal recorded on a detector at one end of the laser is filtered, its frequency divided by 2 in a ECL logic. The resulting signal is amplified and applied to the crystal $S$ (of Fig. 10) of which the electro-optic coefficient is to be measured.

A typical beat note is shown in Fig. 12 (a), with its Fourier Transform in Fig. 12 (b). The crystal is a sample of LiNbO$_3$ of thickness $\ell = 2$ mm, at Brewster’s angle. As the electronic delay of the signal applied to the sample is varied, the beat note shows a sinusoidal dependence, as shown in Fig. 12 (c), which is a plot of the beat frequency versus the delay. The optimum timing occurs when one pulse sees a voltage on the sample of $+V_0$ and the second pulse sees a voltage of $-V_0$ at the sample. The line plotted in Fig. 12 is not a fit, but a plot of $V_0 \sin\left(\frac{2\pi c}{2L} \tau_D - \phi_0\right)$, where the fixed phase, $\phi_0$, was the only free parameter and $\tau_D$ is the delay in applied voltage.

Figure 12: (a) Beat signal versus time(second). (b) Fourier Transform of (a). The upper scale shows the displacement $\Delta P$, in pm, corresponding to the scale of beat note $\Delta \nu$. (c) Delay dependence of the beat note frequency.
The phase change $\Delta \varphi = 2\pi \Delta n l / \lambda$ induced by the applied field on the crystal is equivalent to a phase change $\Delta \varphi = 2\pi \Delta P / \lambda$ due to an optical path change $\Delta P$. One can therefore express the beat note $\Delta \nu = \Delta \varphi / (2\pi \tau RT)$ as:

$$\Delta \nu = \frac{\Delta P}{P} \nu. \quad (84)$$

To appreciate the sensitivity of the measurement presented in Fig. 12, a scale representing the change in optical path (in pm) is added to the Fourier Transform in (b). It is clear that the intracavity measurement easily resolves a change in optical path of less than 0.01 pm, provided that optical path is changed periodically at the cavity repetition rate. In the next section we consider extending this sensitivity to the determination of a small elongation, without any restriction of periodicity.

### 6.2.2 Measurements of small displacements

The intracavity measurement of small displacements is conducted in a linear cavity such as in Fig. 10 (a) or as in Fig. 11 (b). One end mirror is replaced by a branching that reminds of a Michelson interferometer, in which a Pockel’s cell switches every other pulse in another arm, as sketched in Fig. 13. The detector $D_1$ records a pulse train at 180 MHz, or twice the cavity repetition rate. This signal goes through a filter, is divided by 2, and the amplified 90 MHz signal is sent to the Pockel’s cell with appropriate phase and bias, in order to derive the “red” intracavity pulse towards the “sample” arm. With a half wave voltage applied to the Pockel’s cell, the beat note observed on detector $D$ is the same $\Delta \nu = \nu \Delta P / P$ as in the previous section.

Figure 13: Intracavity distance measurement. The linear laser is mode-locked with two intracavity pulses. A Pockel’s cell and a polarizing beam splitter direct alternatively one of the pulses towards the reference mirror $REF$, and the other towards the sample reflector $S$. The beat note is detected on detector $D$. The Pockel’s cell is driven at the cavity round-trip frequency (90 MHz). For applications that require a low power on the sample, only a small voltage is applied to the Pockel’s cell, thus diverting only a fraction of the red pulse towards the sample. In order to operate the laser at minimum loss, a feedback loop is introduced to control the position of the reference mirror, as to minimize the 90 MHz component of the signal detected on $D_1$.

Practical applications require often a reduced laser power on the sample [38, 40]. It is therefore interesting to consider the operation of the laser with an arbitrary voltage on the Pockel’s cell, resulting in a phase difference $\Delta$ between the components of the polarization along the optic axis and orthogonal. Before entering the modulator, the light is vertically polarized, along an axis $\hat{y}$, orthogonal to an horizontal axis $\hat{x}$. The initial field is $|\hat{y} = 1 \cdot \exp(i\omega t)|$. We project that field on the main axis $\hat{y}_m$ and $\hat{x}_m$ of the modulator which is oriented at $45^\circ$ with respect to the lab frame. The modulator introduces a difference in phase $\Delta$ (which we will put on the axis $y_m$). At the output of the modulator:

$$E_{xm} = \cos \theta_1 = \frac{1}{\sqrt{2}} e^{i\omega t}$$
\[ E_{ym} = \sin \theta_1 = \frac{1}{\sqrt{2}} e^{i(\omega t + \Delta)} \]  

The polarizing beam splitter selects components along a vertical \( \hat{y} \) and horizontal \( \hat{x} \) axis. The returning beams recombine after having been given a relative phase shift \( \phi \):

\[ \hat{E}_x = \frac{1}{2} \left( 1 - e^{i\Delta} \right) e^{i\varphi} \]
\[ \hat{E}_y = \frac{1}{2} \left( 1 + e^{i\Delta} \right) \]  

(86)

The modulator imparts another phase change of \( \Delta \) on the \( \hat{ym} \) component \( E_{ym} \rightarrow E_{ym} \times \exp(i\Delta) \). The components along \( \hat{y} \) and \( \hat{x} \) of the pulse re-entering the main cavity are:

\[ E_x = \frac{1}{4} \left\{ \left( 1 - e^{i\Delta} \right) \left( 1 + e^{i\varphi} \right) \left( 1 + e^{i\Delta} \right) \right\} e^{i\omega t} \]
\[ E_y = \frac{1}{4} \left\{ \left( 1 + e^{i\Delta} \right)^2 + e^{i\varphi} \left( 1 - e^{i\Delta} \right)^2 \right\} e^{i\omega t}. \]  

(87)

As expected, the ideal condition is the half wave retardation, which corresponds to \( \Delta = \pi \), leading to \( E_{x1} = 0 \) and \( E_{y1} = \exp(i\varphi) \). Thus, the beat note that will be measured will have exactly the frequency \( \Delta \nu = \varphi/(2\pi \tau_{RT}) \).

It is often not desirable to have the full laser power applied to the element under investigation. Instead of a full half wave retardation, a smaller voltage (hence smaller \( \Delta \)) can be applied to the Pockel’s cell. It is obvious from inspection of Eq. (87) that the loss component \( E_{x1} \) will be large, unless \( \varphi \approx \pi \). The beat note corresponding to \( \varphi = \pi \) coincides with half the repetition rate. Accurate monitoring of the “elongation” of the sample arm can be made by doubling the beat note, and monitoring the frequency difference between the double beat note and the repetition rate.

The small \( \Delta \) approximation of Eq. (87) is:

\[ \left\{ E_x \approx -\frac{1}{2} i \Delta \left( 1 + e^{i\varphi} \right), E_y \approx \left[ 1 + i \Delta - \frac{\Delta^2}{4} \left( 1 + e^{i\varphi} \right) \right] \right\}. \]  

(88)

If \( \varphi \) is close to \( \pi \), the losses are practically zero, but the sensitivity to phase shift is multiplied by \( \Delta^2/4 (\Delta \ll 1) \).

An example of measurement is shown in Fig. 14. The linear cavity is that of a Nd:vanadate laser mode-locked with 4 quantum wells on a GaAs substrate, located in the optical middle of the cavity. Because the saturable absorber is fixed in position, there is a coupling (dead band) resulting from backscattering at the surface of the sample. Because of this coupling, the beat note response is nonlinear, and has harmonics. It can therefore be detected near the repetition rate frequency, as sidebands of the repetition rate peak. For a voltage of 216 V, the sidebands are separated by 290 kHz [Fig. 14 (a)]. The arrow points to this position, as the mirror is moved by 72 nm and 131 nm, and the sidebands move.

The problem of reduced response — ratio of beat note to elongation — due to the coupling between pulses at the solid saturable absorber can be solved by the use of the synchronously pumped OPO described in Section 6.1.3. It remains however that the position of the sample reflector is relative to that of the reference mirror. Therefore, accurate measurements require a stabilization of the repetition rate \( 1/\tau_{RT} \) and CEO \( f_0 \) of the reference cavity. One advantage of the configuration described in Section 6.1.3 is that the repetition rate and CEO are decoupled from one another: control of the repetition rate is achieved by acting on the repetition rate of the pump laser, while the CEO is independently controlled by adjusting the signal cavity length. Complex electronic feedback loops have been developed for this purpose (Menlo System Inc). Another approach is being investigated by using the dispersive properties of atomic resonances in order to stabilize a frequency comb. The wavelength of a laser will be stabilized if the dispersion of the cavity is such that the index of refraction (hence the optical length of the cavity) increases when the wavelength decreases [41]. It is demonstrated in the next section that atomic transitions can also be exploited to stabilize the repetition rate of a laser.
7 Repetition rate spectroscopy

7.1 Introduction

Laser sources had brought a revolution in spectroscopy techniques [42]. Stabilized continuous laser sources with narrow linewidth [43] enable high resolution spectroscopy [44]. By mixing light frequencies one can expand the interrogation of the system to nonlinear regime in ever expanding Coherent Anti-Stoke Raman Scattering (CARS) [45, 46]. Coherent interactions with two and three level systems have made it possible to interrogate atomic and molecular systems within their linewidth [20, 47]. A new level of accuracy in spectroscopy is made possible through the frequency comb of a mode-locked laser. Since the spacing between each adjacent teeth of the comb is the same across the spectrum, the accuracy of one frequency standard can be extended to any part of the spectrum, which has important applications in astrophysics. The mode locked frequency comb can also be seen as a coherent superposition of narrow line continuous frequency sources, that are phase locked. Since the source can access each individual mode of a complex system, this suggests a new type of spectroscopy where one looks for molecular finger prints [48]. The new level of accuracy and control of the frequency comb leads in turn to the extraction and amplification of single pulse of well defined carrier to envelope phase. A single optical pulse can be seen as bursts of electric field, which can be exploited to time resolve electron wave packet motions [49]. In the latter case, the duration, energy and carrier to envelope phase of the single pulse plays a role.

Our approach is not to seek extreme time resolution, or expanding the finest frequency resolution across the spectrum, but to exploit the perfect periodicity of the frequency comb. Instead — or in addition to — tuning the optical frequency of the laser, one can tune the repetition rate of the mode locked laser. The concept is shown in Fig 15. If the timing between successive pulses is an integer multiple of a resonance, or, in frequency picture, if the comb is adjusted so that two teeth of the comb coincide with the frequency difference, the condition of a low frequency resonance is met, while the optical frequency \( \nu_N \) is maintained on resonance with the \( 1 \to 3 \) and \( 2 \to 3 \) transitions. With a typical Ti:sapphire laser one has both access to the optical (hundreds of THz) and RF (0.1-5 GHz) frequency ranges. Resonances in the RF range are accessed by tuning the repetition rate of the laser, while keeping the optical frequency constant. As an example of resonance that can be accessed through this technique is the hyperfine splitting in \( \text{Rb}_{87} \). The Section 7.2 that follows presents a brief review of prior work involving coherence induced between the forbidden ground states, coherence which manifests itself through Coherent Population Trapping (CPT) [50]. It will be shown in Section 7.3 how a simple repetition rate scanning can induce a narrow dark line resonance independently of the CEO of the pulse train. Experimental confirmation is presented in Section 7.4.
Figure 15: (a) In Frequency domain one can consider to stretch or contract the mode comb in Accordion motion, so that the repetition rate matched a submultiple of the desired resonance \( \nu_{RT} = \frac{\nu_{res}}{N} \). (b) In time domain one can picture the pulse train coming at integer multiple of the period of the resonance \( \tau_{RT} = N\tau_{res} \). In contrast to the situation considered in Section 6 on IPI and sensors, the interaction is independent of the CEO and CEP.

7.2 Coherent Population Trapping

In a three level system consisting in two hyperfine ground state levels and an upper level, a particular laser excitation can provide a resonance condition in which destructive interference occurs between excitation pathways. The population is reduced in the upper state and trapped within the two ground states, creating a “dark state”. The population is reduced in the upper state and trapped within the two ground states, creating a “dark state”. The reduction of fluorescence and absorption had given the name “dark line” to this effect [51, 52, 50, 53]. Since the transition between ground states of an alkali metal is electric dipole forbidden and has a long lifetime, the narrow resonance feature of this effect could be exploited as an attractive RF frequency standard [54]. Numerous studies have searched for the optimal parameters — such as laser power, light polarization, magnetic field and cell temperature — for narrowest width and best contrast of the resonance [55, 56, 57, 58]. Other “tricks” have been found to reduce the linewidth, such as introduction of a buffer gas [59] and coating the cell walls with paraffin [60].

CEP experiments were usually performed by inducing coherence between the hyperfine split states either with an RF field [55], or tuning two ultra-narrow CW lasers to each branch of the optical transition [59]. Either approach requires at least one high precision optical source accurately tunable and narrow bandwidth. Instead, as shown in the next two Sections, the coherence can be established by tuning solely the repetition rate of a frequency comb, provided the spectral envelope of the comb covers the optical transitions being considered.

7.3 Theory of Coherent Population Trapping by repetition rate spectroscopy

The physical situation to be modelled is the interaction of a train of picosecond pulses with the atomic system consisting in the two split \( 5S_{1/2} \) ground state hyperfine levels of \( ^{87}\text{Rb} \), and the excited \( 5P_{1/2} \) level [61]. The spectrum of the picosecond pulse covers both optical transitions, keeping each branch of the \( \Lambda \) transition on resonance. It will be shown that the population is trapped between the ground states, when the repetition rate of the pulse train is a submultiple of the hyperfine splitting.

The configuration under study is atom interaction with co-propagating beams. The transition in this case is Doppler broadened by 500 MHz. In the \( \Lambda \) transition of the \( D_1 \) line, one can say that a right circularly polarized photon creates a \( \sigma_+ \) transition and a left circularly polarized one induces the \( \sigma_- \) transition. The ps pulse is broad enough to cover the two hyperfine ground state splitting of 6.8 GHz and the modes are fine enough to see the separation. The levels involved in this transition are labeled in Fig. 16, with the fine structure splitting in the excited level “3” being neglected in the present calculations [62]. Let us establish the density matrix equations of the three levels driven by an arbitrary electric field \( E \). The evolution of density matrix is given by:

\[
i\hbar \dot{\rho} = [H, \rho] = [H_0 + H', \rho] + \Gamma \rho,
\]  

\[ \text{(89)} \]
Figure 16: Three level simplified picture of $^{87}$Rb. Here level 1 corresponds to $F=1$ in $5S_{1/2}$, level 2 is $F=2$ in $5S_{1/2}$ and level 3 is $5P_{1/2}$ with both $F=1$ and $F=2$

where the perturbation of the Hamiltonian of the atom $H$ is $H' = -p.E$, “$p$” is the dipole moment, “$E$” is the electric field and “$\Gamma$” is the decay rate. Note that there is no dipole transition between the two ground states: the $V_{12}$ and $V_{21}$ components of the Hamiltonian matrix is zero. The matrix commutation relation excluding the decay leads to:

$$i\hbar \begin{pmatrix} \dot{\rho}_{11} & \dot{\rho}_{12} & \dot{\rho}_{13} \\ \dot{\rho}_{21} & \dot{\rho}_{22} & \dot{\rho}_{23} \\ \dot{\rho}_{31} & \dot{\rho}_{32} & \dot{\rho}_{33} \end{pmatrix} = \begin{pmatrix} H_{11} & 0 & V_{13} \\ 0 & H_{22} & V_{23} \\ V_{31} & V_{32} & H_{33} \end{pmatrix} \cdot \begin{pmatrix} \rho_{11} & \rho_{12} & \rho_{13} \\ \rho_{21} & \rho_{22} & \rho_{23} \\ \rho_{31} & \rho_{32} & \rho_{33} \end{pmatrix} - \begin{pmatrix} H_{11} & 0 & V_{13} \\ 0 & H_{22} & V_{23} \\ V_{31} & V_{32} & H_{33} \end{pmatrix} \cdot \begin{pmatrix} \rho_{11} & \rho_{12} & \rho_{13} \\ \rho_{21} & \rho_{22} & \rho_{23} \\ \rho_{31} & \rho_{32} & \rho_{33} \end{pmatrix},$$

where the tilde over components of $||\rho||$ indicates a complex number (the diagonal matrix elements are real).

The diagonal terms of the Hamiltonian matrix are the eigenvalues of the energy of the unperturbed atom, and the off diagonal elements the electric dipole moment operator. The rate of change of each matrix element is

$$\frac{d\rho_{11}}{dt} = -\frac{i}{\hbar} \left[ (\hbar \omega_{11} \rho_{11} + V_{13}\tilde{\rho}_{31}) - (\hbar \omega_{11} \rho_{11} + V_{31}\tilde{\rho}_{13}) \right] + \frac{\Gamma_{3}}{2} \rho_{33}$$

$$\frac{d\rho_{22}}{dt} = -\frac{i}{\hbar} \left[ (\hbar \omega_{22} \rho_{22} + V_{23}\tilde{\rho}_{32}) - (\hbar \omega_{22} \rho_{22} + V_{32}\tilde{\rho}_{23}) \right] + \frac{\Gamma_{3}}{2} \rho_{33}$$

$$\frac{d\rho_{33}}{dt} = -\frac{i}{\hbar} \left[ (\hbar \omega_{33} \rho_{33} + V_{31}\tilde{\rho}_{13} + V_{32}\tilde{\rho}_{23} + \hbar \omega_{33} \rho_{33}) - (V_{13}\tilde{\rho}_{31} + V_{23}\tilde{\rho}_{32} + \hbar \omega_{33} \rho_{33}) \right] - \frac{\Gamma_{3}}{2} \rho_{33}$$

$$\frac{d\tilde{\rho}_{21}}{dt} = -\frac{i}{\hbar} \left[ (V_{23}\tilde{\rho}_{31} + \hbar \omega_{21} \tilde{\rho}_{21}) - (V_{31}\tilde{\rho}_{23} + \hbar \omega_{12} \tilde{\rho}_{21}) \right] - \frac{\Gamma_{21}}{2} \tilde{\rho}_{21}$$
\[
\frac{d\tilde{\rho}_{31}}{dt} = -\frac{i}{\hbar} \left[ (\hbar(\omega_2 - \omega_1)\tilde{\rho}_{21} + V_{32}^*\tilde{\rho}_{32} - V_{31}^*\tilde{\rho}_{31}) - \Gamma_{21}\tilde{\rho}_{21} \right] \\
\frac{d\tilde{\rho}_{32}}{dt} = -\frac{i}{\hbar} \left[ (\hbar(\omega_3 - \omega_1)\tilde{\rho}_{31} + V_{32}\tilde{\rho}_{21} - V_{31}(\rho_{33} - \rho_{11}) - \Gamma_{31}\tilde{\rho}_{31} \right]
\]

(91)

In the above system of equations, \(\hbar\omega_1\), \(\hbar\omega_2\) and \(\hbar\omega_3\) are the energies of the lowest ground state, the intermediate state and the upper state. Of all the processes that contribute to the relaxation rates \(\Gamma_{ij}\), it is the transit time broadening that is relevant in reaching the steady state condition in the experimental condition outlined in the next section. For \(^{87}\text{Rb}\) at room temperature, the average thermal velocity is 138 m/s, which implies that an atom crosses a 2 mm beam diameter in 0.145 \(\mu\)s. A pulse train with a period of 8 ns establishes coherence with an atom which leaves the interaction region after interacting with 17000 pulses.

The interaction potential \(V_{ij}\) is the electric dipole strength or \(|\Psi_i\rangle - |\vec{p}.\vec{E}|\Psi_j\rangle\), with the following properties

\[
p_{ij} = p_{ji} \\
V_{ij} = -p_{ij}E \\
V_{ij}^* = V_{ji}.
\]

(92)

In order to eliminate the high frequency oscillation of the density matrix elements, the central frequency of the light \(\omega\) is chosen in the rotating wave approximation, with

\[
\tilde{\rho}_{32} = \frac{\tilde{\sigma}_{32}e^{i\omega t}}{2} \\
\tilde{\rho}_{31} = \frac{\tilde{\sigma}_{31}e^{i\omega t}}{2}.
\]

(93)

The electric field can be described as a pulse envelope with central carrier frequency

\[
E = \frac{\tilde{E}e^{i\omega t}}{2}.
\]

(94)

Note that \(\tilde{E}\) is a complex electric field amplitude which entails the eventual chirp of the pulse. The pulse train picture in time can be represented by pulse envelopes (for example a Gaussian pulse with FWHM of \(\tau_p = 1\) ps) separated in time with pulse period \(\tau_{RT}\), which creates a phase shift \(\omega\tau_{RT}\) from pulse to pulse. This can mathematically be stated as:

\[
E = \sum_{k=0}^{N-1} \tilde{E}(t - k\tau_{RT})e^{i\omega(t - k\tau_{RT})} \\
= e^{i\omega t} \sum_{k=0}^{N-1} \tilde{E}(t - k\tau_{RT})e^{i\phi_k},
\]

(95)

where \(\phi_k\) describes the phase relation between pulses. The evolution of density matrix components can be monitored by the electric field envelope \(\tilde{E}\), the frequency of light \(\omega\) and dipole transition values. Making use of
the property that $\text{Im}(A) = -i(A - A^*)$ leads to the following system for the density matrix equation:

\[
\begin{align*}
\frac{d\rho_{11}}{dt} &= -\frac{p_{31}}{2\hbar} \text{Im}(\tilde{E}^* \tilde{\sigma}_{31}) + \frac{\Gamma_3}{2} \rho_{33} \\
\frac{d\rho_{22}}{dt} &= -\frac{p_{32}}{2\hbar} \text{Im}(\tilde{E}^* \tilde{\sigma}_{32}) + \frac{\Gamma_3}{2} \rho_{33} \\
\frac{d\rho_{33}}{dt} &= \frac{p_{32}}{2\hbar} \text{Im}(\tilde{E}^* \tilde{\sigma}_{32}) + \frac{p_{31}}{2\hbar} \text{Im}(\tilde{E}^* \tilde{\sigma}_{31}) - \Gamma_3 \rho_{33} \\
\frac{d\tilde{\rho}_{21}}{dt} &= -i(\omega_2 - \omega_1)\tilde{\rho}_{21} + i\frac{p_{32}}{4\hbar} \tilde{E}^* \tilde{\sigma}_{31} - i\frac{p_{31}}{4\hbar} \tilde{E} \tilde{\sigma}_{32} - \Gamma_{21} \rho_{21} \\
\frac{d\tilde{\sigma}_{31}}{dt} &= -i(\omega_3 - \omega_1 - \omega)\tilde{\sigma}_{31} - i\frac{p_{31}}{\hbar} (\rho_{33} - \rho_{11}) + i\frac{p_{32}}{\hbar} \tilde{\rho}_{21} - \Gamma_{31} \tilde{\sigma}_{31} \\
\frac{d\tilde{\sigma}_{32}}{dt} &= -i(\omega_3 - \omega_2 - \omega)\tilde{\sigma}_{32} - i\frac{p_{32}}{\hbar} (\rho_{33} - \rho_{22}) + i\frac{p_{31}}{\hbar} \tilde{\rho}_{21} - \Gamma_{32} \tilde{\sigma}_{32}
\end{align*}
\]

(96)

For the three level calculation $p_{31} = p_{32} = 2.537 \times 10^{-27}$ C.m and $\Gamma_3 = 2\pi/27.7$ GHz and $\Gamma_{12} = 2\pi/69$ kHz for the transit time interaction of Rb at room temperature with a beam size 2 mm. The lower state hyperfine splitting is 6.834682 GHz. The resonance tuning frequency is $\omega_3 - \omega_1 - \omega$ which is zero when the transition “1 $\iff$ 3” is in resonance. The system of differential equations is defined by the frequency tuning and the electric field envelope through the equations Eq. (94) and (95). Taking $\rho_{13}$ and $\rho_{23}$, off diagonal matrix elements, in a frame rotating at the optical frequency $\omega$, the fastest oscillation in the system is due to the ground state splitting.

This system of differential equations (96) is solved using the Butcher predictor-corrector method [63] which has been found to be most stable for solving Shrödinger type equations [64]. It has been verified that the elements of the density matrix converge towards a steady-state value after a sufficient number of pulses. The value of the density matrix element interacting with 900 pulses is taken as the steady state value. The pulse

Figure 17: Plots of the $\rho_{33}$ as a function of pulse period, for area 0.1. The first dip corresponds to the case where repetition rate is 6834.7 GHz which is the hyperfine splitting in the ground state. The other dips are for $1/2$, $1/3$ and higher fractions of this value.

envelope is chosen to be the Gaussian $E = E_0 \exp[-(t/\tau_G)^2]$, with a duration $\tau_P = \sqrt{2 \ln 2} \tau_G = 1$ ps for most calculations, although the result has been tested for $\tau_P = 100$ fs and 10 ps. The time integral of the Rabi
frequency
\[
\text{area} = \int \frac{p_{ij} E}{\hbar} \, dt
\] (97)
over a single pulse of the train is the pulse area, a parameter that defines how hard the transition is driven.

The diagonal element $\rho_{33}$, proportional to the fluorescence induced by the pulses, is plotted as a function of the repetition rate in Figure 17. A series of dark lines are observed as the repetition rate of the pulses is being scanned. The dark line is a coherence feature observed when the repetition rate of the pulse train equals a submultiple of the hyperfine splitting.

7.3.1 Bandwidth and frequency considerations

The level structure of rubidium puts some practical limitations on the pulse duration and spectrum for which the interaction can be reasonably modelled. With femtosecond pulses, two photon transitions can no longer be neglected, and complicate significantly the interaction. On the other hand, the pulse bandwidth should be large compared with the hyperfine splitting of 6 GHz. An important question to answer is whether the resonance frequencies observed in Fig. 17 are affected by carrier frequency values and fluctuations. Several simulations tested the value of the repetition at resonance, for various scenarios of carrier frequency perturbations, in a range of ± 500 MHz. Either the pulse carrier frequency was tuned off resonance, or given a chirp leading to a bandwidth increase of 500 MHz, or random pulse to pulse carrier frequencies were introduced within that range. Figure 18 shows an example of simulation where the dark line resonance is seen to occur at the same value of repetition rate, when the spectrum is tune off resonance by up to 500 MHz. As a result one can expect to observe the coherent feature without stabilizing the spectrum. This property also shows that Doppler broadening does not — to first order [65] — affect the dark line feature.

Figure 18: Plots of the $\rho_{33}$ as a function of repetition rate. The carrier frequency of the light had been set to off resonant values. As there is less light in the group of interacting modes, the amplitude of the signal is less, however the dark line structure is unchanged.

7.3.2 Spatial effects

There are two types of spatial effects that have to be considered: the dependence of the dark line resonance on intensity, and the reaction of the medium on the pulses. The shape of the dark line is very sensitive to the
laser power, as illustrated by the successive plots of Fig. 19. The curves are labeled by the value of the area, time integral of the Rabi frequency for an individual pulse. The repetition rate is chosen to be close to the hyperfine splitting, to study the dependence of the first of the dark line series in Fig. 17, on optical power. The laser pulse duration is set to 1 ps. For areas below 0.01 the dark line is only a small perturbation on a weak fluorescence. For the area between 0.01 and 0.1 the dark line has a narrow width. As the area increase above 0.5 the broadening of the dark line can be seen in the picture, leading to complete extinction of the dark line feature for an area equal to 2.

The result of the power broadening calculation in Fig. 19 helps us select the right beam size and power for optimum observation of the dark line. Ideally, a super-Gaussian beam should be used, with a flat top corresponding to an area/pulse of less than 0.05. With the use of a Gaussian beam, some averaging between the curves corresponding to $\theta = 0.05$ to $\theta = 0.5$ will occur. In the experiment however, the width of the resonance is simply limited by the transit time broadening (time required to the atoms at the average thermal velocity to cross the 2.3 mm diameter beam).

In the experiment described in the next section, the rubidium cell is optically thin, and the modification of the beam profile due to rubidium absorption is negligible. One can however predict that, with an optially thick cell, the beam profile may stabilize to a flat top profile at an intensity corresponding to the area of minimum (dark line) absorption (at the repetition rate resonance). Another rubidium-induced beam distortion is due to the spatial phase modulation which, to first order, would self-focus or de-focus the beam. This effect is clearly negligible in the case of the cell considered in the next section, which is short compared to both the absorption length and the Rayleigh range of the 2.3 mm diameter beam. The situation is different when the rubidium is inserted in a laser cavity, where a small repetition rate dependent focusing or defocusing may have an impact on the cavity losses. This effect is presently under investigation experimentally, with a ring laser of variable repetition rate that incorporates a rubidium cell in its cavity.

7.4 Experimental demonstration of repetition rate spectroscopy

In order to have higher intensity at each mode and better selection of the atomic levels, the laser is tailored to produce ps pulses. The laser operation is not disturbed when the repetition rate is tuned over $\pm 2\%$ of the repetition rate $\nu_{rep}$. The resonance of interest is the hyperfine splitting in Rb$_{87}$, inducing coherence between the
forbidden ground states, coherence which manifests itself through Coherent Population Trapping (CPT). The

pulse spectrum should be broad enough to cover the hyperfine splitting, on the other hand having a too broad spectrum results in exciting unwanted transitions and having lower power in the modes that are on resonance within 500 MHz doppler bandwidth. A few ps mode locked pulse seems to be an ideal case, while talking about mode comb is still legitimate. The laser source is a Ti:Sapphire pumped by a frequency doubled Nd Vanadate laser (Coherent Verdi-5W). Mode locking and spectrum control are achieved through proper design of multiple quantum wells in reflection [66]. A 4.6 mm quartz birefringent filter enabled fine tuning of the pulse frequency, and a restriction of the pulse bandwidth (central frequency 795 nm and bandwidth 2 nm).

In view of the result of the simulation, the experimental setup can be as simple as sketched in Fig. 20, and electronic stabilization [9] can be bypassed. The output of the mode-locked laser is sent to the $^{87}$Rb cell at room temperature. The chopped fluorescence signal from the side of the cell is detected with a locking amplifier. The signal at chopped frequency is sent to the computer where a lab view program records simultaneously the repetition rate of the laser and the fluorescence. The repetition rate is scanned by translating the output coupler, and monitored by a frequency counter synchronized to a 10 MHz calibration signal of a Novatech Rb frequency standard.

The earth magnetic field is suppressed with Helmholtz coils in order to keep the hyperfine levels magnetically degenerate. The proper choice of quarter waveplate (QWP) and half waveplate is for polarization dependence studies. The plot of fluorescence (Fig. 21) shows a dip when the repetition rate is 1/57 of the hyperfine splitting. The width of the darkline is 11 kHz, corresponding to 1/57th of the transit time broadening of a 2.3 mm diameter of a Gaussian beam with area 0.23.
Figure 21: Measurement of fluorescence as function of repetition rate. The dip in fluorescence corresponds to the repetition rate equal to 1/57 of hyperfine splitting.

7.5 Implication of Dark line spectroscopy

Within the range of repetition rate of Fig. 21, the absorption and fluorescence have a dip at a fraction of the hyperfine splitting frequency. There is a resonant dispersion corresponding to such a negative absorption feature. The Kramers-Kronig relation imply that, at resonance, the index of refraction will increase with repetition rate. On the other hand, in a laser cavity, the repetition rate decreases with increasing cavity length. Both effects are illustrated in the sketch of Fig. 22. Incorporating rubidium in the cavity, the response of the repetition rate to a cavity fluctuation will be compensated by the dispersive response of the dark line, resulting in an autostabilization of the repetition of the laser. Such a stabilization scheme can applied to the pump laser of the synchronously pumped OPO described in Section 6.1.3, resulting in the having the pulses in the signal cavity locked to an atomic reference.

8 Conclusion

A mode-locked laser emits a frequency and temporal comb, with properties that remains to be fully exploited. A remarkable property is the tooth spacing in the frequency domain is constant across the bandwidth of a single pulse. This paper has demonstrated how this property can be exploited, by devising a laser with two intracavity pulses, producing pulse trains with equal spacing, but unequal CEO. By making the two pulse trains interfere on a detector, a beat note is detected, that has the frequency equal to the ratio of the difference in phase between the two pulses circulating inside the laser to the cavity round-trip time. The exquisite sensitivity of this Intracavity Phase Interferometry (IPI) stems from the fact that a phase difference is measured by converting it into a frequency. A sensitivity to a phase difference of $2\pi10^{-7}$ has been demonstrated with unstabilized lasers (an even better resolution can be achieved through stabilization). This corresponds to a difference in optical path (seen by the two intracavity pulses) of only $10^{-7}\lambda$, of the order of 100 fm. This property has been exploited to measure Fresnel drag [28], rotation [29, 67], nonlinear index [30], and electro-optic coefficient [39] and magnetic field [37]. This paper discusses the extension of these measurements to absolute distance measurements,
with the same accuracy. The method involves alternating each of the two intracavity pulses into a reference and sample subcavity with an electro-optic modulator. Such a technique requires absolute stabilization of the “reference” frequency comb. A simple stabilization method involving interaction between a frequency comb and a three level system has been proposed.

The main property of the comb that is exploited in the applications considered here is the “ruler” in the frequency domain. The “sensors” considered utilize the property that the CEO is proportionally affected by any change in phase of the intracavity pulse. The measurements are based on the interference of two combs of the same pitch. Exploiting these properties have resulted in a spatial resolution of \(10^{-7} \times \lambda\). One might wonder if a time-space analogy exist for this technique, which should lead to a temporal resolution of \(10^{-7} \times T \approx 1 \times 10^{-3}\) as. Instead of two frequency combs, one could consider the superposition of two pulse trains with the same carrier frequency, but different repetition rates \(\tau_{RT1} = \tau_{RT}\) and \(\tau_{RT2} = \tau_{RT} + \epsilon\). Using the description of Eq. 10, the sum of the two pulse trains is:

\[
E_{\text{train1}} + E_{\text{train2}} = e^{i\omega t} \left\{ \sum_{q=0}^{\infty} \left[ \tilde{E}_1(t - q\tau_{RT1})e^{iq\varphi_{p1}} + \tilde{E}_2(t - q\tau_{RT2})e^{iq\varphi_{p2}} \right] \right\}
\] (98)

The field can only interfere in the region where the pulses temporally overlap, for instance if for a certain value \(q_0\) of \(q\), \(q_0\tau_{RT1} = (q_0 + 1)\tau_{RT2}\) or \(q_0\epsilon = \tau_{RT}\). Since \(\varphi_{p1} = q\tau_{RT1} - 2N\pi\) and \(\varphi_{p2} = (q + 1)\tau_{RT1} - 2N\pi\), the two fields are in phase when overlapping. If the field given by Eq. (98) impinges on a detector, the detected signal will be a train of electrical pulses spaced by \(T = q_0\tau_{RT}\), from which one determines \(\epsilon = \tau_{RT}^2/T\). Take \(T = 1\ s\), \(\tau_{RT} = 1\ ns\), \(\epsilon = 10^{-9}\ ns = 1\ as\).

At this point one can only speculate which kind of “Gedanke Experiment” could take advantage of such a resolution. Let us consider a laser cavity injected by a pulse train from another laser. Because of the injection, the carrier frequency of that laser is slaved by the injecting laser. Some nonlinear process to be measured may affect the repetition rate. For instance, if some high harmonic generation is taking place in that cavity, it should be possible to determine where the most energy is taken out of the few cycle pulse. If it is on the leading edge
of each cycle, there will be a slight shift of the envelope at each passage, hence a minuscule change in group velocity. This could be measured by having the beam divided in a portion that does not interact, which will be interfered on a detector with the interacting portion.

The authors gratefully acknowledge support from the W. M. Keck Foundation, and the National Science Foundation under grant Nb. ECS 0601612.
References


