Towards the carrier-envelope phase stabilization of a 16 TW 4.5 fs laser system

Master’s Thesis

Emil Thorin (emth0022@student.umu.se)

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Abstract

In the last decades the scientific development has made it possible to produce pulses with durations below the femtosecond time scale \( (1 \text{ fs} = 10^{-15} \text{ s}) \), reaching to attoseconds \( (1 \text{ as} = 10^{-18} \text{ s}) \). This is the time scale of electronic motion inside atoms and molecules. One way to produce isolated attosecond pulses is through high harmonic generation in gases with intense few-cycle laser pulses. This process depends strongly on the electric field shape relative to the pulse envelope, which is characterized by the so called carrier-envelope phase (CEP).

The goal of this master’s thesis is to measure and investigate the possibility to improve the CEP stability of sub-two-cycle laser pulses from the laser, Light Wave Synthesizer 20 (LWS-20). The first step of the master’s thesis was to modify a Labview program used to evaluate the CEP change to be able to reevaluate the already acquired raw data. The measurements are done with an f-to-2f interferometer, which is a spectral interference device, which measures the CEP difference between two pulses. The CEP change of the laser system was measured at three positions: after the multi-pass amplifier of the laser front end (MP), after a hollow-core fiber (HCF), which is used for spectral broadening, and at the end of the laser system. The stability is determined as the RMS error (standard deviation) of the phase change over all shots in one sample (lower RMS is better stability). The measurements show an average stability of \( 160 \pm 20 \) mrad RMS after the MP, \( 280 \pm 31 \) mrad RMS after the HCF and \( 560 \pm 53 \) mrad RMS at the end of the system. The stability at the end of the system could be improved to \( 475 \pm 40 \) mrad RMS after a scan of the pump energy for one of the amplifier stages. The HCF appears to provide a lower limit in stability and influences it only if it is very good after the MP. The alignment of the HCF does also seem to influence the CEP stability and the best stability appears to coincide with maximum output energy. An acousto-optic modulator (Dazzler) has been used to manipulate the CEP change at the end of the system and can thereby compensate for long-term drifts, but the source of the CEP stability degradation at the end of the system should be further investigated.

Supervisor: Peter Fischer (peter.fischer@umu.se)
Examinator: Prof. Laszlo Veisz (laszlo.veisz@umu.se)
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1 Introduction

Ever since the invention of the first ruby laser in the 1960s [1] it has been under constant development of design and performance. Multiple techniques of building pulsed lasers shortly emerged, like Q-switching (1962) [2], and Mode Locking (1963) [2]. Pulsed lasers can generate peak powers of several orders of magnitude higher than of a continuous wave (CW) laser [3, 2] and allow for high time resolution of experiments [4], where pulse durations below 10 femtoseconds (1 fs = $10^{-15}$ s) are achieved by a technique called Kerr lens mode locking [4]. The laser is used as a tool in a wide range of fields, ranging from industrial applications like laser cutting [5], medical applications such as eye- and dental surgery [6], monitoring pollution in the environment [7] and much more.

Apart from these applications, the field of attosecond physics studies ultrafast processes like electronic motion inside atoms and molecules, which uses pulsed lasers to achieve a time resolution in the attosecond range (1 as = $10^{-18}$ s). This time resolution can be achieved using intense few-cycle femtosecond laser pulses for high harmonic generation (HHG) in gases [8], that correspond to attosecond pulses in the time domain. The laser pulse ionizes the gas atom, releasing an electron. The electron is accelerated in the laser field, gaining kinetic energy and has a probability to recombine with the ionized atom and releasing a high-energy photon [8]. Attosecond pulses serve as the "flashes" for our camera when studying ultrafast processes, and to take sharp images isolated attosecond pulses (IAPs) are required. In the HHG process the shape of the electric field with respect to the field envelope, more specifically, the phase of the carrier wave relative to the field envelope (known as the carrier-envelope phase; CEP, see section 2.1), which effectively changes the instantaneous intensity, becomes important when generating IAPs [8]. Depending on the CEP, either a single attosecond pulse is generated if an intensity maximum coincide with the envelope maximum (cosine pulse), or a pair of pulses are generated if there are two symmetric intensity maxima on either side of the envelope maximum (sine pulse) and everything in between. By making sure that the CEP is stable and does not change from pulse to pulse one can guarantee IAPs to be generated from every laser pulse and the experimental conditions are identical, which significantly cuts down the amount of repeated measurements required for the experiments\(^1\). The field of nanophotonics, which studies the interaction of light with nano-scale objects, also involves CEP dependent processes [9]. One common technique to measure the CEP fluctuations is through f-to-2f interferometry [10, 11, 12, 13], which measures the relative CEP change between two pulses. This change can be used in a feedback system to stabilize the CEP, which can be done by electro- or acousto-optic modulators (EOM/AOM) [15, 16] at higher rep. rates (MHz and beyond) or by moving or tilting optical components [10, 11] such as dispersive elements or cavity mirrors at lower rep. rates (few-kHz and below), just to name a few techniques.

\(^1\)Imagine every pulse having the same CEP instead of every tenth or twentieth, then the number of measurements are reduced by a factor of 10 or 20.
A comparison with other similar systems will give an idea of what numbers we can expect for the CEP stability. There are other terawatt systems that has measured CEP stabilities of 670 mrad RMS [12] with a stability of 320 mrad RMS (10 ms averaging) in the kHz system, one system with a CEP stability of 315 mrad [17] with 300 mrad in the kHz system and one group has measured 1.3 rad [13] with 150 mrad (20 ms averaging) in the kHz system. However, averaging the stability measurements gives misleading results [17] and they appear better than their real value. One group even reports a system with a stability of 210 mrad with a front end stability of below 100 mrad [14].

The goal of this master thesis is to measure and investigate the possibility of CEP stabilization of sub-two-cycle laser pulses from the Light Wave Synthesizer 20 (LWS-20). The LWS-20 is located in the Relativistic Attosecond Physics Laboratory (REAL) at Umeå University and is used to generate attosecond pulses for research purposes. As the first step a program for CEP change evaluation will be modified to reevaluate measurements without the spectrometer. The first experimental goal is to get familiar with the CEP measurement device, an f-to-2f interferometer, and perform measurements of the CEP change at different positions in the LWS-20 laser system. Measurements will be made after the compressed multi-pass amplifier of the front-end, after the hollow-core fiber compressor of the front end and at the end of the full LWS-20 laser chain with 16 TW and 4.5 fs pulses. Conclusions about the CEP stabilization of the system will be drawn from the measurements with the f-to-2f interferometer and eventually a commercial hardware will be implemented to stabilize the CEP of LWS-20 in the future.
2 Theory

2.1 Ultrashort Pulses

An ultrashort pulse refers to a laser pulse with a time duration in the picosecond (ps; $10^{-12}$ s), or even femtosecond (fs; $10^{-15}$ s) time scale, and are usually produced by mode locked lasers [3, 4]. The electric field of a laser pulse can be described in the complex representation\(^2\) [3]

$$E(t) = \text{Re}\left\{ A(t)e^{i(\omega_0 t - \phi(t))} \right\} = \frac{1}{2} A(t)e^{i(\omega_0 t - \phi(t))} + \text{c.c.}, \quad (1)$$

where \(A(t)\) is the field envelope, \(\omega_0\) is the carrier frequency, \(\phi(t)\) is the temporal phase of the carrier wave and the second term, c.c., refers to the complex conjugate of the first term. The field envelope \(A(t)\) is usually described as a Gaussian with the intensity full width at half maximum (FWHM) \(\tau_{\text{FWHM}}\), defined below. An example of an ultrashort pulse is shown in figure (1), where the field envelope is a Gaussian, and the temporal phase is constant \(\phi(t) = \phi_{\text{CEP}}\), which is known as the carrier-envelope phase (CEP). The CEP is important especially in few-cycle pulses, where the field properties change significantly with the CEP [11].

Figure 1 – Temporal electric field of a sub-cycle pulse, with reference to the intensity full width at half maximum (FWHM) \(\tau_{\text{FWHM}}=2.5\) fs. A single-cycle would correspond to a period of 2.7 fs. The carrier-envelope phase is the constant offset of the carrier wave with respect to the envelope maximum.

As mentioned above, the full width at half maximum is the pulse duration where the intensity is reduced by half of its peak value. The (instantaneous) intensity of the electric field is given by [19]

\(^2\)Note that this is only the magnitude. To be more correct, this should also include a polarization.
\[ I(t) = n c \epsilon_0 A(t)^2 \cos^2 (\omega_0 t - \phi(t)), \]  

(2)

where \( n \) is the refractive index of the medium, \( c \) is the speed of light and \( \epsilon_0 \) is the permittivity of free space. Since most physical phenomena are not sensitive to the rapid oscillations of the electric field (and instantaneous intensity) only to its cycle-average, the intensity given by [3, 19]

\[ \langle I(t) \rangle = \frac{1}{2} n c \epsilon_0 A(t)^2, \]  

(3)

Since the intensity is proportional to the squared electric field, the electric field FWHM and intensity FWHM differs by a factor of \( \sqrt{2} \) for a Gaussian pulse. This is illustrated in figure (2), which shows peak normalized electric field and intensity envelopes with \( \tau_{FWHM} = 2.5 \) fs.

![Electric Field Envelope](image)

**Figure 2** – Peak normalized electric field envelope, intensity envelope and instantaneous intensity with \( \tau_{FWHM} = 2.5 \) fs and \( \phi(t) = 0 \) (not the same as the pulse in figure (1)).

By Fourier transforming \( E(t) \) the pulse is expressed in the spectral domain and we get information about the frequency components of the pulse. The spectral electric field is given by

\[ \tilde{E}(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} E(t) e^{-i\omega t} dt, \]  

(4)

and we recover the temporal electric field again by inverse Fourier transforming the spectral electric field

\[ E(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \tilde{E}(\omega) e^{i\omega t} d\omega. \]  

(5)
We can define two important physical quantities, which are the spectrum \(^3\) [3]

\[
S(\omega) = |\tilde{E}(\omega)|^2, \quad (6)
\]

and the spectral phase. The spectral phase is defined as the complex phase of the spectral electric field, i.e. [20]

\[
\varphi(\omega) = -\arctan \left\{ \frac{\text{Im}[\tilde{E}(\omega)]}{\text{Re}[\tilde{E}(\omega)]} \right\}. \quad (7)
\]

The spectral phase is important for the properties of a pulse, and the physical meaning is most conveniently described after Taylor expanding \(\varphi(\omega)\) around \(\omega_0\), where it can be analyzed term by term. The Taylor expansion becomes [21]

\[
\varphi(\omega) = \varphi(\omega_0) + \varphi_1 \cdot (\omega - \omega_0) + \varphi_2 \cdot \frac{(\omega - \omega_0)^2}{2!} + \varphi_3 \cdot \frac{(\omega - \omega_0)^3}{3!} + \ldots \quad (8)
\]

where the 0th order term is the CEP in frequency domain, \(\varphi_{\text{CEP}}\), (which is the same as in the time domain, \(\phi_{\text{CEP}}\), since it is a constant), and where

\[
\varphi_1 = \left. \frac{d\varphi(\omega)}{d\omega}\right|_{\omega_0} \quad \text{is the group delay (GD)},
\]

\[
\varphi_2 = \left. \frac{d^2\varphi(\omega)}{d\omega^2}\right|_{\omega_0} \quad \text{is called the group-delay dispersion (GDD)},
\]

\[
\varphi_3 = \left. \frac{d^3\varphi(\omega)}{d\omega^3}\right|_{\omega_0} \quad \text{is called the third order dispersion (TOD)},
\]

followed by fourth order dispersion etc. The effect of the different terms, up to third order, are visualized in figure (3) below.

\(^3\)The spectrum describes the intensity for different wavelengths, i.e., its frequency distribution.
Figure 3 – The effect of first 4 terms of the spectral phase, compared to a cosine reference pulse (maxima of carrier and field envelope at $t = 0$). The reference pulse is chosen to have a wavelength $\lambda = 800$ nm and an intensity FWHM pulse duration of $\tau = 2.5$ fs.

We can see in figure (3) that the zeroth order spectral phase (CEP) shifts the carrier wave within the envelope and the first order phase shifts the envelope in time. The second order spectral phase introduces a temporal frequency shift, a "chirp", and stretches the pulse in time. The third order spectral phase introduces pre-pulses and stretches the pulse.

Two other important physical quantities of ultrashort pulses are the peak power [29]

$$P_{\text{peak}} \approx \frac{E}{\tau},$$

which refers to the power of an individual pulse, and the average power [30]

$$P_{\text{average}} = Ef_{\text{rep}},$$

$^4$Or post-pulses depending on the sign of $\phi_3$. 

6
where \( E \) is the pulse energy, \( \tau \) is the pulse duration and \( f_{\text{rep}} \) is the repetition rate of the laser. The peak power is the power during a single pulse and is important for experiments which require high intensity. The average power is the power integrated over several pulses and is relevant to get useful results of the experiments and good statistics, i.e. repeated observations.

2.2 Nonlinear Optics

If the laser intensity is sufficiently high, we will enter the regime of Nonlinear Optics [19]. The propagation of light in a dielectric medium satisfy Maxwell’s equations, from which one can derive the wave equation for the electric field [3]

\[
\nabla^2 E - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 P}{\partial t^2},
\]

where \( P \) is the polarization density, i.e. the dipole moment per unit volume. One starting point to nonlinear optics is to express \( P \) as an expansion in terms of the applied electric field [19]

\[
P = \varepsilon_0 (\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \ldots),
\]

where \( \chi^{(n)} \) is the \( n \)-th order susceptibility tensor of rank \((n+1)\). We can realize that, since \( \chi^{(n)} \) is a tensor, \( P \parallel E \) does not always hold. This can cause the higher order terms to have different polarization from \( E \), which will be the case for us in second harmonic generation, where the second harmonic wave is perpendicularly polarized to the fundamental [19].

2.2.1 Second Harmonic Generation

Second harmonic generation (SHG) is a special case when the second term in Eq. 12 \((\propto E^2)\) is significant. Consider an incident field \( E \) in a nonlinear medium, with electric field magnitude given by Eq. 1. For simplicity we neglect the pulse envelope (and the phase term) and consider the incident field to be a continuous wave (CW) with magnitude \( E_0 \). Then the second term of the nonlinear polarization in Eq. 12 becomes

\[
P^{(2)}(t) = \frac{1}{4} \varepsilon_0 \chi^{(2)} \left( E_0^2 e^{2i\omega_0 t} + |E_0|^2 \right) + \text{c.c.},
\]

where the first term corresponds to SHG and the second term is called optical rectification (OR), which is of no interest for us. Thus from Eq. 13 there will be a source term with frequency \( 2\omega_0 \) in Eq. 11 which will produce a frequency doubled field.

2.2.2 Phase Matching

In order for most nonlinear effects to occur (SHG for instance) the electric fields needs to constructively interfere and they need to be phase matched. The reason for this is that
the fields have different phase velocities\(^5\) due to frequency dependence in the refractive index [3]. To achieve phase matching, the condition

\[ 2k_\omega = k_{2\omega} \iff n(\omega) = n(2\omega) \tag{14} \]

must be fulfilled. Unfortunately, dispersion prevents this from happening, because in most transparent materials \(dn/d\omega > 0\) [3]. However, this problem can be solved by a property called birefringence, which means that the refractive index depends on the polarization direction. Consider a uniaxial birefringent crystal. The orientation of the molecules define the crystal’s optical axis, which defines a plane together with the direction of propagation. Two kinds of waves can propagate inside the crystal, ordinary waves, which polarization is perpendicular to that plane, and extraordinary waves, which polarization is parallel to that plane. These two axes (ordinary and extraordinary) can have different refractive indices, figure (4), which can allow phase matching between the fundamental and second harmonic waves.

![Figure 4](image)

**Figure 4** – Frequency dependence of the index of refraction of a birefringent material with ordinary refractive index \(n_o\) and extraordinary refractive index \(n_e\). The second harmonic can have the same refractive index as the fundamental if birefringence is utilized. Source: [22].

One way to achieve phase matching for a desired frequency is to tune the angle between the optical axis and propagation direction, also called "angle phase matching". The phase matching condition depends on the refractive index: [3]

\[
\frac{1}{n_e^2(\omega, \theta)} = \frac{\cos^2(\theta)}{n_o^2(\omega)} + \frac{\sin^2(\theta)}{n_o^2(\omega)} \tag{15}
\]

where \(n_e\) is the extraordinary refractive index, \(n_o\) is the ordinary refractive index and \(\theta\) is the angle between the optical axis and direction of propagation. In the case of SHG, angle phase matching is achieved when

\(^5\)Recall the phase velocity: \(v_p = c/n(\omega)\)
n_0(2\omega) = n_e(\omega, \theta). \quad (16)

### 2.2.3 White Light Generation

Consider the third term in Eq. 12, and neglecting second order polarization, Eq. 11 becomes

\[ \nabla^2 E - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \left( \frac{\chi^{(1)}}{c^2} + \frac{\chi^{(3)}|E|^2}{c^2} \right) \frac{\partial^2 E}{\partial t^2}. \] \quad (17)

By rearranging the terms in Eq. 17, the index of refraction becomes [3]

\[ n^2 = 1 + \chi^{(1)} + \chi^{(3)}|E|^2 \equiv n_0^2 + \chi^{(3)}|E|^2, \] \quad (18)

where \( n_0 \) is the traditional linear refractive index. This can be written as

\[ n = n_0 \sqrt{1 + \frac{\chi^{(3)}}{n_0^2}|E|^2} \approx n_0 + \frac{\chi^{(3)}}{2n_0}|E|^2 \equiv n_0 + n_2 I, \] \quad (19)

where \( n_2 \) is the nonlinear refractive index and \( I \) is the intensity. The approximation is valid when \( (\chi^{(3)}/n_0^2)|E|^2 \) is small. This intensity dependence on the refractive index is called the optical Kerr effect. This effect gives rise to self-focusing (and defocusing) and self-phase modulation (SPM). By propagating through a medium SPM causes the pulse to pick up an extra temporal phase [3]

\[ \phi(z,t) = \frac{\omega n_2 I(t)z}{c}, \] \quad (20)

where \( z \) is the distance traveled through the medium. The acquired phase from SPM broadens the spectrum of the pulse [3]. We call this process white light generation (WLG), since we will observe a broader spectrum. The associated spatial self-focusing keeps the beam focused for a long distance (longer than the diffraction would allow), which is called filamentation [24], see figure (5a). Moreover, if the beam profile is not perfectly smooth and the intensity/energy is too high, small scale self-focusing can occur. This means that instead of the beam being focused towards the center causing a single filament, the noise peaks of the beam are locally focused and cause multiple filaments, figure (5b).
2.3 f-to-2f Interferometry

One technique to measure the change of the carrier-envelope phase (carrier-envelope offset; CEO) is by f-to-2f interferometry, which is based on spectral interference [21, 25], that is interference in the frequency domain. Some sources define CEO=CEP [26], but I will denote CEO as the CEP change between two pulses. A laser oscillator is typically said to be CEO stabilized if the CEO is constant, i.e. the change in CEP is constant. However, an amplifier is called CEP-stabilized if all pulses have the same CEP value. The original laser pulse (the fundamental) is frequency doubled using SHG. The CEO can be measured if the fundamental and second harmonic spectra overlap, thereby causing a beating [27], i.e. an interference in the spectrum. In order to get this beating, the fundamental spectrum must be octave spanning, i.e. the spectrum must span a frequency of a factor of two ($f_{\text{max}} \geq 2f_{\text{min}}$ in the fundamental spectrum). This can be done through WLG, described in section 2.2.3. As mentioned in section 2.1, ultrashort pulses are usually generated by mode locked lasers which spectra have a comb structure\(^6\) [10, 11], shown in figure (6). The frequency of the $k$th comb mode is given by [27]

$$f_k = f_{\text{CEO}} + kf_{\text{rep}}, \quad (21)$$

where

$$f_{\text{CEO}} = \frac{\phi_{\text{CEO}}}{2\pi} f_{\text{rep}}, \quad (22)$$

and $\phi_{\text{CEO}}, [0, 2\pi]$ is the carrier-envelope offset and $f_{\text{rep}}$ is the repetition rate of the laser. Using Eq. 21 we can express two comb modes as

$$f_1 = f_{\text{CEO}} + mf_{\text{rep}},$$
$$f_2 = f_{\text{CEO}} + nf_{\text{rep}}, \quad (23)$$

where $f_1$ is a comb line in the red part of the fundamental and $f_2$ is a comb line in the blue part of the fundamental. In the overlapping part of the spectra, we have $n = 2m$

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\(^6\)This can be realized by Fourier transforming an infinite pulse train.
and the frequency doubled fundamental for two neighboring comb lines. These comb lines will interfere with each other and the beat frequency is given by [28]

\[ 2f_1 - f_2 = f_{CEO} \]  \hspace{1cm} (24)

As \( f_{CEO} \) changes, the interference pattern in the spectrum also changes. Thereby we can measure \( \phi_{CEO} \) from the interference.

In single shot measurements we compare the interference pattern between two shots. The CEO, or CEP change between two shots, can be extracted from the beating by cutting out the interfering part of the spectra and applying the Fourier transform algorithm [25], further described in appendix A.

\section*{3 Method/Experimental Setup}

\subsection*{3.1 The Light Wave Synthesizer 20}

The Light Wave Synthesizer 20 (LWS-20), which is a laser providing relativistic intensity few-cycle pulses, shown in figure (7). It is divided into the following parts: the oscillator, the broadband seed generation, the stretcher, the pump laser, the optical parametric synthesizer (OPS) and the compressor [31].
The oscillator is a mode locked Ti:Sapphire laser with a central wavelength of approximately 800 nm operating at a repetition rate of 80 MHz, whose output is split into seed and pump output. The seed is amplified in a multi-pass chirped-pulse amplifier (CPA) at 1 kHz repetition rate and then spectrally broadened (through SPM) in a neon-filled hollow-core fiber (HCF). Chirped-pulse amplification means that the pulse is stretched in time, to reduce its intensity to avoid damage to the amplifier, amplified and then compressed again. Optionally, a cross-polarized wave generation (XPW) setup is used after the HCF to improve pulse contrast. The broadened pulses are then temporally stretched by a grism (grating and prism pair) stretcher before the OPS stages, to avoid damage. The OPS consist of an acousto-optic modulator (AOM; Dazzler) and four NOPA (Non-collinear Optical Parametric Amplification) stages amplifying the pulses at a 10 Hz repetition rate. Without going into details, Optical Parametric Amplification (OPA) transfers energy from a pump pulse to a seed pulse in a nonlinear crystal. The

\[7\text{This is due to pre- and post pulses produced in the amplification process, reflections in the amplifiers, amplitude or phase distortions and/or amplified spontaneous emission.}\]
Dazzler is programmable to control the spectral phase and amplitude of the output pulse, further described in section 3.1.2. The long and short wavelength parts of the pulses are amplified separately by the 1A, 2A and 1B, 2B BBO’s respectively. The pump output of the oscillator is frequency shifted in a photonic crystal fiber (PCF) and goes to a flash lamp pumped Nd:YAG laser, which then pumps the NOPA stages after being frequency doubled and tripled. The pulses are then compressed to about 400 fs by 4 pieces of bulk glass, followed by an adaptive mirror to correct for the aberrations and compressed to approximately Fourier-limited duration using chirp mirrors inside a vacuum chamber. Finally after compression, a part of the pulse is sent into an above-threshold ionization (ATI) phase meter to measure the CEP of the pulse. As seen in figure (7), the output pulses have an energy of 70-80 mJ, a pulse duration of 4.5 fs and a repetition rate of 10 Hz, which from Eq. 9 & 10 gives a peak power of approximately 16 TW and an average power of approximately 750 mW.

There may be several sources of CEP noise and drift, not only from airflow, temperature variations, pressure variations etc., but also from individual components in the system. Different sources of noise may affect the CEP on different time scales. For example, mechanical vibrations may cause noise on a shorter time scale than temperature variations. Thereby we want to investigate how different components and parameters affects the CEP stability, and the measurements are carried out after the multi-pass amplifier (9-pass CPA in figure (7)), after the hollow-core fiber (when the pulses are re-compressed by a chirp-mirror compressor) and in the end of the system.

3.1.1 Oscillator Stabilization

The Ti:Sapphire oscillator is CEO stabilized by modulating its continuous wave pump laser intensity using an acousto-optic modulator (AOM). The oscillator pulses are focused in a Periodically Poled Lithium Niobate (PPLN) crystal where SPM and difference frequency generation (DFG) occurs, which will produce a beat signal with the fundamental [15], which holds information about the CEP drift. SPM broadens the spectrum and the DFG will beat with its red part, in our case generating a signal around 1300-1500 nm. This signal is separated by a dichroic mirror (a mirror that transmits a certain band of wavelength and reflects others) which transmits the beat signal, through a long-pass filter to filter out the unwanted part of the spectrum. The reflected part from the dichroic mirror is sent to the multi-pass amplifier. The beat signal is then coupled to a photo detector and used to lock the CEO using external electronics (Menlo Systems). The phase locking electronics are set to lock the CEO to 1/4 of the repetition rate, which means that every 4-th pulse will have the same CEP. In order to achieve a proper phase locking the electronics require a signal-to-noise ratio of at least 35 dB [34].

These parameters affects the refractive index, which will affect the group and phase velocity, thereby affecting the CEP.
3.1.2 Dazzler, Acousto-Optic Programmable Dispersive Filter

The Dazzler mentioned in section 3.1 is an acousto-optical device (an Acousto-Optic Programmable Dispersive Filter; AOPDF) which can tailor the spectral phase and amplitude of the incident pulse [35]. The pulse travels through a nonlinear birefringent crystal, where it interacts with an acoustic wave traveling along the beam. Due to this interaction with the acoustic wave, the incident pulse has its polarization rotated from the ordinary axis (or extraordinary) to the extraordinary (or ordinary) axis, see figure (8).

![Figure 8](image)

**Figure 8** – Working principle of the Acousto-Optic Programmable Dispersive Filter, a.k.a. the Dazzler, for pulse compression. An uncompressed pulse is sent into the crystal, where it interacts with the acoustic wave. The polarization of the incident pulse is flipped due to the interaction with the acoustic wave and the output pulse can be compressed. Source: [36].

Which frequency of the laser pulse that has its polarization rotated depends on the acoustic wave frequency and the phase matching condition between the optic and acoustic wave [35]. The Dazzler can be programmed to add or subtract a spectral phase, up to 4th order in Eq. 8 or an arbitrary phase from a file, and the spectral amplitude can be tailored for the output pulse [36]. The Dazzler also has a programmable transmission, by which we can transmit a percentage of the pulse.

3.2 f-to-2f Interferometer Experimental Setup

The f-to-2f experimental setup, shown in figure (9), consist of two irises to align the beam position and direction, two focusing mirrors, a 3 mm thick piece of quartz glass for white light generation, a 100 μm thick beta-barium borate (BBO) crystal for second harmonic generation, a polarizer, filters, a focusing lens and a detector (spectrometer). The first iris also controls the input energy of the pulses and the pulse is focused into the quartz glass by the first focusing mirror, where WLG occurs to broaden the spectrum. We want to have enough intensity for WLG, but not so high that we get multiple filaments. The second focusing mirror focuses the beam into the BBO crystal, which can be turned to optimize the phase matching for the SHG. The polarizer is used to equalize the polariza-
tion and match the amplitude of the second harmonic and the fundamental, because the second harmonic produced from the BBO is polarized perpendicular to the fundamental. This is referred to as a type-1 crystal. The polarizer is also used to match the intensities of the fundamental and second harmonic.

**Figure 9** – Schematic figure of the f-to-2f interferometer setup. The setup consists of two irises (I), two focusing mirrors (FM), a 3 mm thick piece of quartz glass (Q), a 100 µm thick BBO crystal (BBO), a polarizer (P), spectral filters (F), a focusing lens(L) and a spectrometer (D). A small portion of the pulse is let through the first iris, illustrated by the thickness of the beam, to not have too high intensity in the quartz glass. The red beam represents the fundamental pulse and the blue beam represents the frequency doubled (second harmonic) pulse.

Then there are some filters to suppress the signal to not damage the spectrometer and to filter out the unnecessary part of the spectrum and only keep the interference region, and the pulses are focused into the input coupler of the spectrometer. The spectrometer (Ocean Optics USB2000+) has a fiber coupled input and detects wavelengths between 200 nm and 900 nm, and has a memory capacity of 3000 spectra. It’s acquisition rate is 500 Hz in the fastest configuration, which means that every 2nd pulse will be measured after the multi-pass amplifier and after the hollow-core fiber. In the end of the system, inside the compressor chamber, the repetition rate is 10 Hz, which means that every pulse is measured. For the measurements at the end the spectrometer uses an external trigger.

### 3.3 Labview Program for Evaluation

The Labview program used for evaluation of the CEP change (CEO) has been made in an earlier master’s thesis [38], and is based on the Fourier transform algorithm [25], explained in more detail in appendix A. A screenshot of the program on the *Single Measurement* tab, figure (10), shows live plots of the measured spectra and the evaluation part described in section 2.3. There is the option to do both single, continuous or high-speed measurements, described further below. There are also some spectrometer settings like integration time, number of measurements to average and the option of background
acquisition together with number of measurements for the background. We always use the fastest possible acquisition and integration time, since we want to perform single-shot measurements and averaging the measurements gives misleading results [17]. Below the plots we can set the regions for the filters in both spectral and quasi-time domains, and the region of averaging for evaluation of the CEP. We can also see the processed spectrum, from which we obtain the phase. The filters are Gaussian and the indicators below the plots show the frequencies/times of the FWHM of these functions. The yellow dashed line in the Spectral Phase plot marks half of the FWHM region.

**Figure 10** – Screen shot of the Labview program for evaluation, displaying the *Single Measurement* tab.

On the *Multiple Measurements* tab, shown in figure (11), we can see the time evolution of the CEP compared to the previous shot, CEP compared to the average value and the acquisition time of the spectra together with appropriate statistics. This data is acquired after pressing the *high-speed measurement* button, which does one measurement, every 2 ms on average when the *free running mode* is selected with 1 ms integration time. The high-speed measurement routine acquires a given number of spectra, up to a maximum of 3000, then evaluates the phase. This tab also have the ability to filter bad spectra, which can happen if the spectrometer is set on free running mode and is not triggered externally. This means that the spectrometer may not be synchronized with the laser and might miss a laser shot. This is the case for the measurements after the multi-pass
amplifier and after the HCF, since the spectrometer acquisition rate matches half the repetition rate of the system. The filtering is basically done by integrating the intensity and comparing it to a certain threshold value.

The CEP to average value is calculated by subtracting a reference phase \( \varphi_{ref} \) from every measurement \( \varphi_k \) and averaging it in half the FWHM spectral range of the filter, which reduces the uncertainty of the evaluation compared to the difference of two measurements [38]. The reference phase is given by

\[
\varphi_{ref}(f) = \frac{1}{N} \sum_{k=1}^{N} (\varphi_k(f) - \varphi_k(f_c)), \tag{25}
\]

where \( N \) is the number of measurements and \( f_c \) is the center frequency of the averaging range in the Fourier transform algorithm. The CEO error graph in figure (11) shows the measurement accuracy, which we have defined as the standard deviation of the phase difference between two shots, further described in appendix A. The actual value for the stability is taken as the standard deviation of the evaluated phases (RMS error) of all measured shots, the values in the CEO window in figure (11).

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{Illustration.png}
\caption{Figure 11 – Screen shot of the Labview program for evaluation, displaying the Multiple Measurement tab.}
\end{figure}
These two tabs described above are the most used tools and the high-speed measurements is the source for most of the results, therefore I will not go into detail about the other tabs in the program. In short, the ect tab has some more real time data about the stabilization, but the data acquired from *Multiple Measurements* are sufficient.

4 Results

4.1 Labview Program Modification

There has been made some modifications to the already existing Labview program for evaluation of the CEP, described in section 3.3, which are described below.

**Offline version (without spectrometer).** A modified version of the program has been made to be used without a spectrometer connected, to evaluate measurements that are already made. In this program the user imports an existing file containing spectra from the measurements and can then use the program as before. This version can be used to test how the evaluation depends on the filter settings and other parameters. One drawback is that there is no time information in the stored spectra, but there is an option to "reconstruct" the time information if the user knows the time delay between the measurements, which can be taken from the original CEP evaluation file. This reconstructed time vector does not contain the exact time stamps, but rather fixed steps in time as average time delay. Visually, the program is more or less identical.

**Super-Gaussian filters.** As mentioned in section 3.3, the evaluation involves filtering the spectrum with a Gaussian filter. This is of course not the only way to filter the signal, and thereby user defined Super-Gaussian filters were implemented. Super-Gaussians are Gaussian functions where the power in the exponential function exceeds 2 and are even integers, i.e. \( e^{xn} \), where \( n = 2 \) is a regular Gaussian. In the limit \( n \to \infty \) this becomes a rectangular function. However, sharp edges in Fourier transformations causes sidebands in the filtered spectra, which can be seen in figure (12), where a super-Gaussian of order 40 is used both in the frequency filtering and the quasi-time filtering.
Figure 12 – Real part of the processed spectrum where a super-Gaussian filter of order 40 is used in both frequency spectrum and quasi-time domain for an f-to-2f signal.

These sidebands do not have a significant influence on the evaluation result for this measurement, but makes it harder to distinguish the correct peak to filter out, see figure (13) (compare with figure (32) appendix A), which is why we will stick with the regular Gaussian filter.

Figure 13 – Absolute value of the quasi-time spectrum where a super-Gaussian filter of order 40 is used in the frequency domain of an f-to-2f signal. We can observe two extra peaks between the middle peak and the beat signal peak.

Angle range. The ability to choose the angle ranges $[0, 2\pi]$ or $[-\pi, \pi]$ was added to the program. This may be useful depending on how the phase is stabilized, i.e. if the CEO is locked to $\pi$, then $[0, 2\pi]$ should be chosen for the angle range to get a good visual representation.

Variable delay. As a method to perform long time measurements with the high-speed measurement function, a variable delay was implemented in the program. The user
can define the time between measurements, which can extend the measurement time. However, we loose the phase information in between the shots. This delay works both in free-running mode and with external trigger.

## 4.2 Measurements after the Multi-pass Amplifier

The first point of measurement is after the multi-pass amplifier (MP; labeled 9-pass CPA in figure (7)). Figure (14) shows the CEP to average for an unstabilized system, over 100 shots. The pulse energy is 10 µJ after the first iris of the f-to-2f setup. The measurement errors in the figures are given by the spectrally averaged standard deviation for the given shot when calculating the phase (described in appendix A). The stability is given as the standard deviation (RMS error) of all shots, which in figure (14) is 14670 mrad and the mean measurement error is 28 mrad. The repetition rate after the MP is 1 kHz and a measurement is done every 2 ms, which means that every 2nd pulse is measured. This influences the result of the unstabilized system since the phase change is added in order to get the phase and half of the information is lost. This is not as much of an issue for a stabilized system since the phase change is not random. A typical spectrum after the multi-pass amplifier is shown in figure (36), appendix B.

![Figure 14](image.png)

**Figure 14** – Measurement of the CEP change after the multi-pass amplifier of an unstabilized system of 100 shots. The CEP changes randomly throughout the measurement.

Figure (15) shows the CEP to average of a stabilized system over 100 shots with an RMS stability of 160 mrad and average measurement error of 19 mrad. The mean stability of several (10) measurements is 170±23 mrad\(^9\). For these measurements, the beat signal for the phase locking electronics was 32 dB.

\(^9\)The error represents the standard deviation of these 10 measurements.
Figure 15 – Measurement of the CEP change after the multi-pass amplifier of a stabilized system of 100 shots. The RMS stability is 160 mrad and the average measurement error is 19 mrad. The phase change is approximately constant.

We are also interested in the stability over a longer time to observe possible instabilities which may occur on a larger time scale, which originate from the multi-pass amplifier (as slow loop stabilization is not included). Figure (16) shows the CEP to average over 3000 shots of the same system as above, where the RMS stability is 200 mrad and the average measurement error is 27 mrad.
There is a slight oscillation of the CEP, but no apparent drift in this measurement. This is unexpected because we do not use any slow loop to correct for any long term drifts. Figure (17) shows a comparison of the stability of 100, 1000, 2000 and 3000 shots, which is the limit of the spectrometer. There is no significant increase in the stability above 1000 shots, but a slight increase from 100 to 1000 shots because we can detect variations on a larger time scale.
Figure 17 – Comparison of measurements for 100, 1000, 2000 and 3000 shots after the multi-pass amplifier. There is a slight increase in RMS stability between 100 and 1000 shots, which indicates variations on a larger time scale.

The beat signal for the phase locking electronics is an important part of the stabilization, therefore a scan of the beat signal was performed to investigate how it affects the CEP stability. Figure (18) shows the stability for beat signals in the range of 24-32 dB, with 10 measurements of 100 shots for each setting. One thing not shown in this figure is the stabilization time, which was affected by the beat signal. This has unfortunately not been measured in detail, but at 32 dB the stabilization could hold for about half an hour, while it could barely hold for a minute for 24 dB.
It is worth mentioning that all of the above measurements were performed on the same day (2018-06-13).

4.3 Measurements after the Hollow Core Fiber

The hollow-core fiber, which is located right after the multi-pass amplifier, broadens the spectrum and thus require less energy to get a sufficient interference signal. A pulse energy of approximately $3 \, \mu$J after the first iris was sufficient. Figure (19) shows the CEP to average for a stabilized system over 100 shots with an RMS stability of 290 mrad and an average measurement error is 8 mrad. The mean stability of the measurement series is $280 \pm 30$ mrad. The mean stability after the multi-pass on this day is $160 \pm 20$ mrad. Note that the measurement after the hollow-core fiber has a lower measurement error than the measurement after the multi-pass. This is because of the fact that the hollow-core fiber provides a broader spectrum and a stronger interference. The measurement is performed 2018-06-19. A typical spectrum after the hollow-core fiber is shown in figure (37), appendix B.
Figure 19 – Measurement of the CEP change after the hollow-core fiber of a stabilized system of 100 shots. The RMS stability is 290 mrad and the average measurement error is 8 mrad.

However, sometimes the measurements after the HCF looks like in figure (20), where the RMS stability is 630 mrad with average measurement error of 10 mrad.

Figure 20 – Measurement of the CEP change after the misaligned hollow-core fiber of a stabilized system of 100 shots. The RMS stability is 630 mrad and the average measurement error is 10 mrad.
This suggests that there might be some dependence of the HCF on the stability. Furthermore, sometimes the measurements show no degradation of the CEP stability after HCF compared to the multi-pass, as shown in figure (21). This measurement was performed 2018-06-25, and the mean stability after the MP was 240±23 mrad and the mean stability after the HCF was 230±28 mrad.

![Figure 21 – Comparison of the CEP stability between the multi-pass amplifier and the hollow-core fiber. The mean stability after the MP was 240±23 mrad and the mean stability after the HCF was 230±28 mrad. Here the HCF has no influence in the stability.](image)

The HCF is mounted on two stages on the optical table, each stage having a horizontal and vertical alignment. The back stage (seen from the laser direction) is scanned, both horizontally and vertically to investigate if this has an effect on the stability. Only the back stage is scanned in these measurements because this stage is less sensitive to misalignments while the front stage is more sensitive and degrades the beam profile and energy. Before the alignment scan, the HCF is aligned to maximum output energy. The result from the horizontal alignment scan is shown in figure (22), where the error bars represent the standard deviation of the measurements. However, since only the first stage is scanned there is no guarantee that this setting is the optimal, which can be seen by comparing figures (22) & (21).
The result from the vertical alignment scan is shown in figure (23).

Figure 22 – Measurement of the CEP change after the hollow-core fiber against horizontal displacement of the back side of the fiber. The error bars represent the standard deviation of the measurements. The magenta measurement (visually not at zero displacement, but in reality it is) is a remeasurement at zero displacement to reproduce the initial result.

Figure 23 – Measurement of the CEP change after the hollow-core fiber against horizontal displacement of the back side of the fiber. The error bars represent the standard deviation of the measurements.
Figures (22) & (23) indicates that the alignment of the HCF might have a significant influence on the stability. To make sure that the stability of the system has not degraded, another measurement was performed after the multi-pass amplifier. The result is shown in figure (24), where the second result after the MP is compared to the first measurement and that after the HCF.

Figure 24 – Comparison of the stability after the MP before (left, labeled MP) and after (right, labeled MP remeasure) alignment scans after the HCF. The stability appears to have improved during remeasurement. This could be a statistical effect, but the system can also have reached steady state.

Figure (24) shows that the stability has not degraded during the time of the HCF measurements, but rather appears to have improved. This can be either just a statistical effect, or the system needed more time to reach a steady state.

4.4 Measurements at the end of the LWS-20 laser system

The measurements at the end of the system are performed inside the compressor vacuum chamber after the chirp-mirrors. The repetition rate is 10 Hz so the CEP changes are observed on a different time scale compared to the MP and the HCF, but may as well depend on the stabilities of them. There are many parameters to scan in the OPS, like pump energies, delay between pump and seed pulse and Dazzler transmission to name a few. Optimally the stability should be the same or at least comparable as after the hollow-core fiber.

Figure (25) shows the CEP to average at the end of the system, with an RMS stability of 560 mrad and mean measurement error of 26 mrad. The mean stability of the measurement series is 560±53 mrad. The measurement was performed 2018-06-19 and the
mean stability after the MP was $160\pm 20$ mrad and the mean stability after the HCF was $280\pm 31$ mrad. A typical spectrum at the end of the system is shown in figure (38), appendix B.

The stability is clearly not as good as after the HCF, which motivates an investigation of how the stability depends on the OPS parameters.

The flash lamp delay for the pump laser was scanned to investigate if it had an influence on the stability at the end of the system. The flash delay scan effectively makes a pump energy scan for the OPA stages. The result is shown in figure (26) and it appears to not have a significant influence in the stability. The flash delay corresponds to a time unit, which is unknown but not that important. The reason there is much more data on flash delay 80 is that this is the default setting for the other measurements and correspond to maximum gain.
Figure 26 – CEP stability measurement at the end of the system of 100 shots for different settings of the flash lamp delay. The different settings have no influence on the stability.

Figure (27) shows the result of a scan of the pump energy for the 1A stage in the OPS. For the 12 mJ there is a slight improvement of the stability of $475\pm40$ mrad, with the best measurement as 410 mrad, compared to the other settings. This indicates that the 1A OPA stage might have an effect on the stability and that should be investigated further. 17 mJ is the original setting that was used this day which is why, as for the flash delay scan, we have more data points at this setting.
Figure 27 – CEP stability measurement at the end of the system of 100 shots for different settings of the 1A stage pump energy. There is an improvement in the stability for the 12 mJ setting. This might indicate that this stage needs to be investigated.

The Dazzler transmission was varied to investigate if it had an influence on the stability. Figure (28) shows the result from varying the Dazzler transmission between 10%, 50% and 100% (default setting). This seems not to improve the stability at the end of the system, however this measurement should be repeated with more data to confirm this statement.
Figure 28 – CEP stability measurement at the end of the system of 100 shots for different settings of the Dazzler transmission. There is no significant improvement in the stability, but there are few measurements for the 10% and 50% settings so one should not draw conclusions.

All of the above measurements are performed on the same day (2018-06-19). Of all measurements above, the 1A pump energy appears to be the only parameter that shows improvement in the stability. There are more parameters to vary in the OPS for further improvement and to find the sources of the shot-to-shot noise in the CEP.

The Dazzler has the ability to manipulate the CEP change (CEO) of the pulses, so it may be interesting to see if that can be used to compensate CEO drifts on a relatively long time scale. However, this cannot compensate for the shot-to-shot stability that we have measured above. To test this idea the Dazzler is programmed to change the CEO between 0 and $\pi/2$ every 4 seconds. In figure (29) the result of such a measurement is shown with a distinct periodic change in the CEO approximately every 4 second. By averaging the blue and black regions in figure (29) separately, the amplitude of the change is estimated by taking the difference between the red lines as $1.70\pm0.17$ rad$^{10}$, which is approximately $\pi/2$ (1.5708). The RMS stability of 100 shots prior to this measurement is $720\pm52$ mrad with a typical measurement error of 40 mrad, but this measurement serves to prove that the CEO can be manipulated so that’s not important.

$^{10}$The measurement error is taken as the square root of the sum of the squares of the standard errors from the black and blue regions respectively, i.e. $s = \sqrt{s_1^2 + s_2^2}$. 

32
We conclude that the Dazzler can be used in a slow loop to compensate for long term drifts in the CEP change.

5 Discussion

Generally, the stability of the laser is similar or even better than other terawatt systems which I mentioned in section 1. However, it is not as good as 315 mrad [17] and we want to optimize the system as far as possible, thereby finding the source of the degradation of the CEP stability. An average stability of 475 mrad is still practically enough for stable generation of isolated attosecond pulses.

The oscillator stabilization has played an important role in this master’s thesis and has been time consuming. There was a problem with acquiring a sufficient beating signal for the stabilization electronics, which was solved through careful alignment of the oscillator to increase the output power and spectrum of the oscillator. The output power could be increased by increasing the pumping power, but it did not always work, probably because the spectrum changed in an unfavorable way for the beat signal point of view, i.e. got narrower. As mentioned in section 3.1.1, a signal of at least 35 dB is required, but as we can see in figure (18) that wasn’t even reached. Although, 32 dB is enough to stabilize the system, but this might reduce the stabilization time. There is no measurement of the stabilization time, but it is observed during other measurements that the stabilization holds for about half an hour with a 32 dB signal and barely a minute with 25 dB. The phase locking electronics does also play an important role in the stabilization, both for
RMS stability and stabilization time and also needed optimization in order to obtain a
good stability of the system. More time was spent on optimizing the electronics before
which might explain the difference in stability after the MP. We should not disregard the
possibility of a coupling between the electronics settings and the beat signal either.

6 Conclusions

The CEP stability has been measured at three different locations in the laser system:
after the multi-pass amplifier, after the hollow-core fiber and at the end of the
system. The result after the multi-pass amplifier shows an average stability of 160 mrad,
with best value of 120 mrad, which is better than the other systems mentioned in sec-
tion 1 where only one system used single-shot measurements. It should be noted that we
only measure every second pulse, but still single-shot, so this number and the number
for the hollow-core fiber might be slightly different. The best thing to obtain a certain
single-shot stability measurement is to get a faster spectrometer to measure every shot
at 1 kHz rate. The result can still be unchanged at 1 kHz rate but it might reveal short
scale variations.

As mentioned earlier, the spectrum of the pulse is broader after the hollow-core fiber,
therefore the average measurement error is lower compared to the multi-pass amplifier.
As shown in figures (21) & (24) the stability after the MP and the HCF are basically the
same, which means that the HCF does not influence the stability. However, for the mea-
surements on 2018-06-19 the stability after the MP was 160 mrad and the stability after
the HCF was 280 mrad, which would imply that the HCF does influence the stability
for larger MP stability values. This could mean that the HCF have some sort of lower
stability limit. The long time measurements (1000, 2000, 3000 shots) for the HCF has
not been mentioned in the results, but behave the same as for the MP, the RMS value
is slightly increased from 100 to 1000 shots. Under certain conditions the stability after
the HCF was at best 230 mrad.

The alignment measurements of the HCF shows that the alignment might have a signif-
icant influence on the stability, which motivates the need for a stationary f-to-2f setup
after the HCF in order to align the HCF properly to have a good stability. If the HCF
is misaligned far enough, plasma will be generated inside the HCF and this can be seen
in the beam profile which will start to flicker. This increased the RMS value quite a lot,
and could explain the measurement in figure (20) with 630 mrad if the HCF was not
aligned properly. However, since only the first stage of the HCF mount was scanned, the
alignment might not be optimal, neither before the alignment scan. Therefore another
alignment scan with both stages should be performed.

At the end of the system we measured every shot since the repetition rate is changed
to 10 Hz from 1 kHz, and here the spectrometer was set to use an external trigger. At the end of the system there are more parameters to investigate than what is done in this thesis. The only parameter that seems to improve the stability at the end of the LWS-20 is the pump energy for the 1A OPA stage, which improved the average stability from 560 mrad down to 475 mrad. This is an important result, as this could tell us how to improve the stability. As mentioned above, the stability is comparable or better than other terawatt systems referenced in section 1, but only one of these use an OPA system and [17] even states that CEP stabilization is technically challenging. However, average stability of 475 mrad is facilitating stable generation of isolated attosecond pulses.

7 Outlook

The source of the degradation of the CEP stability is still unknown and future work will involve improving the stability further and trying to find the source. The next step is to repeat some of the relevant measurements done in this thesis to confirm the results, like the HCF alignment scan with both stages and 1A pump energy. A further step is to measure at the end of the system without the XPW and to do more measurements where the pump energies for the OPA stages are varied, also for other stages than 1A. The pump energy in the second stages could tell us if the problem is saturation there or amplification in the first ones. However, we cannot decrease the pump energies without decreasing the output energy of the LWS-20. Measurements without amplification in the OPA stages is also something to consider, but this might not be possible due to the low energy. The Dazzler can be used to compensate for long-term drifts of the CEP change with a simple feedback system, but this is not suitable to compensate the shot-to-shot noise of the CEP.
References


[38] Linβ, S., Automation of single-shot carrier envelope phase measurement, Master’s Thesis, Department of Physics, Umeå University, (2017)
A Fourier Transform Algorithm

The beating between the fundamental and second harmonic, which contains the information about the carrier-envelope phase, can be evaluated using the Fourier transform algorithm. This technique involves filtering out the interfering part of the original spectrum (figure (30)), in our case using a Gaussian filter, seen in figure (31).

![Figure 30](image.png)

**Figure 30** – Measured wavelength spectrum after the multi-pass amplifier. We can observe the interference pattern (beating) around 500 nm.

The wavelength spectrum is first transformed into frequency spectrum and the Gaussian filter is applied around the interference.
Figure 31 – Frequency spectrum (blue) from the signal in figure (30). The beating is filtered out (red) using a Gaussian filter function (dashed).

The filtered spectrum is inverse Fourier transformed into a "quasi-time domain", seen in figure (32), which is a time representation of the filtered spectrum. It is convenient to use a Gaussian filter, since the Fourier transform of a Gaussian is still a Gaussian [28]. The large peak around zero in figure (32) comes from the Gaussian shape of the filtered signal, where the side peaks appears symmetrically around zero due to the modulation in the spectrum\textsuperscript{11}. Therefore, one of the side peaks is filtered.

Figure 32 – Absolute value of the inverse Fourier transformed filtered signal, which we call the "quasi-time domain". The middle peak appears due to the Gaussian shape of the filter function and the side peaks appears due to the modulation (interference) in the frequency spectrum. One of the side peaks, we choose the right one, is filtered again (red) using a Gaussian filter function (dashed).

\textsuperscript{11}This is a property of the Fourier transform.
The filtered part of the quasi-time domain is Fourier transformed back to the frequency domain, which we call the "processed spectrum". The real part of the processed spectrum is shown in figure (33).

![Figure 33](image)

**Figure 33** – The real part of the Fourier transformed quasi-time domain signal, which we call the "processed spectrum". This is a complex spectrum which contains the phase information of the pulse relative to a reference pulse. Therefore, the change in the phase between two pulses can be determined, but not the absolute phase unless a reference is known.

By using this complex spectrum we have acquired by the Fourier transformations, the spectral phase can be calculated using Eq. 7 in section 2. The calculated phase is shown in figure (34), where the dashed line denotes the FWHM of the Gaussian filter.
Figure 34 – The phase (up to an unknown constant) calculated from the complex processed spectrum. The dashed lines indicate the FWHM of the Gaussian filter in the frequency spectrum.

The above procedure is repeated for another pulse and we subtract the phase from figure (34) from each other. The phase difference is averaged over half of the FWHM region (symmetrically), which is the CEP change between the two pulses, and this average is subtracted from the phase difference. The result is a function close to zero, shown in figure (35).
Figure 35 – The difference between two phases (figure (34)) with their average over half of the FWHM region, i.e. the CEP change between the two pulses, subtracted. The standard deviation of this function over the same region gives the measurement accuracy.

The standard deviation of the function in figure (35) is defined as the measurement accuracy between two shots [38] and will represent the errorbars in section 4.
B Laser Spectra

This section holds the spectra at the measurement locations in the laser system. Figure (36) shows the spectrum after the multi-pass amplifier.

![Spectrum](image)

**Figure 36** – Spectrum measured after the multi-pass amplifier.

Figure (37) shows the spectrum after the hollow-core fiber.
Figure 37 – Spectrum measured after the hollow-core fiber.

Figure (38) shows the spectrum after the optical parametric synthesizer.

Figure 38 – Spectrum measured after the optical parametric synthesizer.
C  Source Data

This section gives the filepaths for the source data used in the results. All paths are given using REALs Attoserver as root folder. Note: Some CEP measurement series may have been shifted by $2\pi$ at some point. This is due to an evaluation from a bad spectra, i.e., the $2\pi$ jump is not real.

Figure (12)
File:\Talks, Posters, Papers, Thesis\Thesis\Emil Master\FTA\Gauss_40.txt

Figure (13)
File:\Talks, Posters, Papers, Thesis\Thesis\Emil Master\FTA\Gauss_40_QT.txt

Figure (14)
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0032--MP_32dB_unstab001_CEP.txt

Figure (15)
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0033--MP_32dB_stab004_CEP.txt

Figure (16)
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0043--MP_32dB_stab021_CEP.txt

Figure (17)
100 shots:
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0033--MP_32dB_stab001_CEP.txt
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0033--MP_32dB_stab002_CEP.txt
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0033--MP_32dB_stab003_CEP.txt
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0033--MP_32dB_stab004_CEP.txt

1000 shots:
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0040--MP_32dB_stab011_CEP.txt
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0040--MP_32dB_stab012_CEP.txt
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0040--MP_32dB_stab013_CEP.txt
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0040--MP_32dB_stab014_CEP.txt

2000 shots:
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0040--MP_32dB_stab015_CEP.txt
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0040--MP_32dB_stab016_CEP.txt
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0040--MP_32dB_stab017_CEP.txt
File:\ExpData\2018\2018 06 13 CEP\f-2f--1806-0040--MP_32dB_stab018_CEP.txt

Figure (18)

32 dB: Same set as 100 shots above.

31 dB:

29 dB:

28 dB:

27 dB:

26 dB:

25 dB:

24 dB:

23 dB:

22 dB:

21 dB:

20 dB:

19 dB:

18 dB: Same set as 100 shots above.
Figure (19)

Figure (20)

Figure (21)

Multi-Pass:
Hollow Core Fiber:

file:\ExpData\2018\2018 06 25 CEP\HCF\f-2f--1806-1439--HCF_32dB_stab001_CEP.txt

file:\ExpData\2018\2018 06 25 CEP\HCF\f-2f--1806-1439--HCF_32dB_stab002_CEP.txt

file:\ExpData\2018\2018 06 25 CEP\HCF\f-2f--1806-1439--HCF_32dB_stab003_CEP.txt

Hollow Core Fiber:

file:\ExpData\2018\2018 06 25 CEP\HCF\f-2f--1806-1439--HCF_32dB_stab004_CEP.txt

file:\ExpData\2018\2018 06 25 CEP\HCF\f-2f--1806-1439--HCF_32dB_stab005_CEP.txt

file:\ExpData\2018\2018 06 25 CEP\HCF\f-2f--1806-1439--HCF_32dB_stab006_CEP.txt

file:\ExpData\2018\2018 06 25 CEP\HCF\f-2f--1806-1439--HCF_32dB_stab007_CEP.txt

file:\ExpData\2018\2018 06 25 CEP\HCF\f-2f--1806-1439--HCF_32dB_stab008_CEP.txt

Figure (22)
ExpData\2018\2018 07 04 CEP\MP\f-2f--1807-1810--MP_30dB_stab017_CEP.txt
ExpData\2018\2018 07 04 CEP\MP\f-2f--1807-1810--MP_30dB_stab018_CEP.txt

HCF: Same set at 0 above,
File:\ExpData\2018\2018 07 04 CEP\HCF\f-2f--1807-1916--HCF_30dB_stab001_CEP.txt...

etc.

MP remeas:
ExpData\2018\2018 07 04 CEP\MP\f-2f--1807-2128--MP_remeas_29dB_stab001_CEP.txt
ExpData\2018\2018 07 04 CEP\MP\f-2f--1807-2128--MP_remeas_29dB_stab002_CEP.txt
ExpData\2018\2018 07 04 CEP\MP\f-2f--1807-2129--MP_remeas_29dB_stab003_CEP.txt
ExpData\2018\2018 07 04 CEP\MP\f-2f--1807-2129--MP_remeas_29dB_stab004_CEP.txt
ExpData\2018\2018 07 04 CEP\MP\f-2f--1807-2129--MP_remeas_29dB_stab005_CEP.txt

Figure (25)
File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1735--db32 056 stable 80 opt_CEP.txt

Figure (26)

FD 80 & pump 17 mJ & Dazzler 100:
File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1734--db32 048 stable 75 opt_CEP.txt
File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1735--db32 049 stable 75 opt_CEP.txt
File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1736--db32 050 stable 75 opt_CEP.txt
File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1737--db32 051 stable 75 opt_CEP.txt

FD 75:
File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1733--db32 045 stable 70 opt_CEP.txt
File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1734--db32 046 stable 70 opt_CEP.txt
File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1734--db32 047 stable 70 opt_CEP.txt

FD 70:
File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1732--db32 043 stable 70 opt_CEP.txt
File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1733--db32 044 stable 70 opt_CEP.txt
File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1734--db32 045 stable 70 opt_CEP.txt

FD 65:
File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-2051--dB32 032 l1a 17mJ long trans 100_CEP.txt

52
Figure (27)

17 mJ (FD 80 & Dazzler 100):
(Same as the set shown above)

File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1734--db32 054 stable 80 opt_CEP.txt
etc.

12 mJ:

File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1734--db32 054 stable 80 opt_CEP.txt

FD 60:

File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1721--db32 032 stable 60 opt_CEP.txt

22 mJ:

File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1955--db32 020 11a 22mJ long_CEP.txt

File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1958--db32 022 11a 22mJ long_CEP.txt

50%:

File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1958--db32 022 11a 22mJ long_CEP.txt

Figure (28)

100% (FD 80 &amp; pump 17 mJ):
(Same as the set shown above)

File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-1734--db32 054 stable 80 opt_CEP.txt
etc.

10%:

File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-2037--db32 036 11a 17mJ long trans 010_CEP.txt

50%:

File:\ExpData\2018\2018 06 19 CEP\Comp\f-2f--1806-2039--db32 038 11a 17mJ long trans 010_CEP.txt

Figure (29)

File:\ExpData\2018\2018 05 14 CEP\Compressor\f-2f--1805-2200--Comp_beating-26_049_CEP.txt