1 Mathematical description of ultrashort laser pulses

1.1 We first perform the Fourier transform directly on the Gaussian electric field:

\[ E(\omega) = \mathcal{F}[E(t)] = \int_{-\infty}^{\infty} A_0 e^{-4 \ln^2 \left( \frac{\epsilon}{T_{FWHM}} \right)^2 e^{i(\omega_0 t + \varphi_{CE})} e^{-i\omega t} dt, \]

\[ = A_0 e^{i\varphi_{CE}} \int_{-\infty}^{\infty} e^{-4 \ln^2 \left( \frac{\epsilon}{T_{FWHM}} \right)^2 e^{i(\omega_0 - \omega) t} dt, \]

\[ = A_0 e^{i\varphi_{CE}} \int_{-\infty}^{\infty} e^{-4 \ln^2 \left( \frac{\epsilon}{T_{FWHM}} \right)^2 + i(\omega_0 - \omega) t} dt. \] (1)

By utilizing the identity:

\[ \int_{-\infty}^{\infty} e^{-ax^2} e^{-2bx^2} dx = \sqrt{\pi} a \sqrt{\frac{a}{a}} \left( a > 0 \right), \] (3)

with \( a = 4 \ln 2 / T_{FWHM}^2 \) and \( b = -i(\omega_0 - \omega)/2 \) we could arrive to:

\[ E(\omega) = A_0 e^{i\varphi_{CE}} \sqrt{\frac{\pi}{4 \ln 2 / T_{FWHM}^2}} e^{\frac{(-i(\omega_0 - \omega)^2)}{4 \ln 2 / T_{FWHM}^2}}, \]

\[ = A_0 e^{i\varphi_{CE}} T_{FWHM} \sqrt{\frac{\pi}{4 \ln 2}} e^{-\frac{T_{FWHM}^2(\omega_0 - \omega)^2}{4 \ln 2}}, \]

\[ = B_0 e^{i\varphi_{CE}} e^{-\left( \frac{\omega - \omega_0}{\Delta \omega_0} \right)^2}, \] (4)

where \( B_0 = A_0 T_{FWHM} \sqrt{\frac{\pi}{4 \ln 2}} \) and \( \Delta \omega_0 = 4 \sqrt{\ln 2 / T_{FWHM}} \). Therefore the full width at half maximum of \( S(\omega) \) is \( \Delta \omega_0 \sqrt{2 \ln 2} \) so we can derive the time-bandwidth product:

\[ \Delta t \cdot \Delta \omega = \frac{T_{FWHM}}{\sqrt{2}} \cdot \Delta \omega_0 \sqrt{2 \ln 2}, \]

\[ = \frac{T_{FWHM}}{\sqrt{2}} \cdot 4 \sqrt{\ln 2 / T_{FWHM}} \sqrt{2 \ln 2}, \]

\[ = 4 \ln 2. \] (5)

From this relation, if we want to have a 1 fs Gaussian pulse, we should have at least 1.83 eV bandwidth, regardless of the carrier frequency. Note that if one wants to calculate the bandwidth in wavelength (nm), the carrier (central) frequency should be taken into account.

1.2 Now we would perform the inverse Fourier transform on a simplistic electric field in the frequency domain \( E(\omega) \) (without Gaussian approximation) by applying an additional spectral phase \( \varphi_m(\omega) = \varphi'(\omega_0)(\omega - \omega_0) \) to get the temporal profile of the electric field after propagation through a medium, utilizing the definition \( E_m(\omega) = E(\omega) e^{i\varphi_m(\omega)} \) which can be proven using the wave equation:
\[ E_m(t) = \mathcal{F}^{-1}[E_m(\omega)] = \frac{1}{2\pi} \int_{-\infty}^{\infty} E_m(\omega)e^{i\omega t} d\omega, \]

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

\[ = \frac{1}{2\pi} \int_{-\infty}^{\infty} E(\omega)e^{i\varphi(\omega)(\omega-\omega_0)}e^{i\omega t} d\omega, \]

\[ = e^{-i\varphi(\omega_0)\omega_0} \frac{1}{2\pi} \int_{-\infty}^{\infty} E(\omega)e^{i\omega(t+\varphi'(\omega_0))} d\omega, \]

\[ = e^{-i\varphi(\omega_0)\omega_0} E(t+\varphi'(\omega_0)). \quad (6) \]

As a result, applying a group delay in the frequency domain corresponds to a shift in time plus a change of the carrier envelope phase.

Similarly we could apply the group delay dispersion \( \varphi_m(\omega) = \frac{1}{2} \varphi''(\omega_0)(\omega - \omega_0)^2 \), now for convenience, we use the Gaussian approximation \( E(\omega) = E_0 e^{-4 \ln 2 \left( \frac{\omega - \omega_0}{\sqrt{2}\Delta\omega} \right)^2} \).

\[ E_m(t) = \mathcal{F}^{-1}[E_m(\omega)] = \frac{1}{2\pi} \int_{-\infty}^{\infty} E(\omega)e^{i\varphi_m(\omega)}e^{i\omega t} d\omega, \]

\[ = \frac{1}{2\pi} \int_{-\infty}^{\infty} E(\omega)e^{t\frac{i}{2} \varphi''(\omega_0)(\omega-\omega_0)^2}e^{i\omega t} d\omega, \]

\[ = \frac{1}{2\pi} \int_{-\infty}^{\infty} E_0 e^{-4 \ln 2 \left( \frac{\omega - \omega_0}{\sqrt{2}\Delta\omega} \right)^2} e^{t\frac{i}{2} \varphi''(\omega_0)(\omega-\omega_0)^2} e^{i\omega t} d\omega, \]

\[ = E_0 e^{-t\varphi(\omega_0)} \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{\left( -\frac{4 \ln 2}{\Delta\omega} \right) \left( \omega - \omega_0 \right)^2 + t\frac{i}{2} \varphi''(\omega_0)} \omega^2 e^{i\omega t} d\omega' \quad \text{with} \quad \omega' = \omega - \omega_0, \]

\[ = E_0 e^{-t\varphi(\omega_0)} \frac{1}{2\pi} \sqrt{\frac{\pi}{\left( -\frac{4 \ln 2}{\Delta\omega} \right) + t\frac{i}{2} \varphi''(\omega_0)}} e^{\left( \frac{-i t/2}{\Delta\omega} \right)^2}, \quad (7) \]

where we have used the identity 3 with \( a = -\left( -\frac{4 \ln 2}{2\Delta\omega} \right) + t\frac{i}{2} \varphi''(\omega_0) \) and \( b = -it/2 \).

Equation 7 can be rewritten as:

\[ E_m(t) = C_0 e^{i\omega_0 t} e^{-\left( \frac{-it/2}{\Delta\omega} \right)^2}, \]

\[ = C_0 e^{i\omega_0 t} e^{-\left( -\frac{4 \ln 2}{2\Delta\omega} + i \frac{1}{2} \varphi''(\omega_0) \right)^2}, \]

\[ = C_0 e^{i\omega_0 t} e^{-\frac{4 \ln 2}{2\Delta\omega} - 2i \varphi''(\omega_0) \sqrt{2 \ln 2}} \]

so the width of the intensity profile of \( E_m(t) \) is \( \Delta t_m = \sqrt{\frac{4 \ln 2}{2\Delta\omega} - 2i \varphi''(\omega_0) \sqrt{2 \ln 2}} \) while the width of the intensity profile without the group delay is \( \Delta t = 4 \ln 2 / \Delta\omega \). Therefore \( \Delta t_m = \)
\[ \Delta t \sqrt{1 - \frac{2i\varphi''(\omega_0)\Delta \omega^2}{16(\ln 2)^2}} \] whose absolute value is larger than \( \Delta t \) for all non-zero \( \varphi''(\omega_0) \).

More details on ultrashort laser pulses in particular and ultrafast optics in general can be found in [1, 2, 3, 4].

2 Generation of ultrashort laser pulses

2.1 a) Within a bandwidth of 1.5 GHz, there are \( \frac{1500 \text{ MHz}}{75 \text{ MHz}} = 20 \) modes, which can be excited.

b) For the Ti:Sa laser, we have to convert the bandwidth of 340 nm around a wavelength of 800 nm to frequencies, which gives us 167 THz. From this we get \( \frac{167 \text{ THz}}{75 \text{ MHz}} = 2.23 \cdot 10^6 \) excitable modes.

The small gain bandwidth of the helium-neon laser allows just a few modes, whereas the large bandwidth of the Ti:Sa allows many modes. Usually one would prefer shorter cavities for a helium-neon laser, to allow for single/dual mode operation (Fig. 1 left). In case of the Ti:Sa one tries to get as many modes as possible to generate a wide spectrum, which supports short pulses in time domain.

2.2 For sufficiently many modes with arbitrary phases the output intensity is a constant value over time, for a fixed phase relation pulses will occur in time. This fact can be described by Fourier transform of the given power spectrum and assuming in the first case arbitrary phases, in the second a constant phase. The two cases are shown for the electric field in Fig. 2.

Using the above time-bandwidth product relation for the Gaussian pulse, we arrive to 667 ps for the helium-neon case and 5.99 fs for the Ti:Sa laser.
Figure 2: Phase dependence of the intensity distribution of output of an oscillator. The top panel represents the output of 20 modes (in and out of phase). The bottom panel represents 5 modes.
2.3 The energy per pulse $E$ is calculated from the average power $P$ and the pulse repetition rate $f_{\text{rep}}$:

$$E = \frac{P}{f_{\text{rep}}} = \frac{500 \text{ mW}}{75 \text{ MHz}} = 6.6 \text{ nJ}.$$ 

The corresponding peak power $P_{\text{max}}$ depends on the used pulse width $\Delta t$:

$$P_{\text{max}} = \frac{E}{\Delta t} = \frac{6.6 \text{ nJ}}{5.99 \text{ fs}} = 1.1 \text{ MW}.$$ 

We now calculate the ratio between peak power and average power:

$$\frac{P_{\text{max}}}{P} = \frac{1.1 \text{ MW}}{500 \text{ mW}} = 2.2 \cdot 10^6.$$ 

One can recognize that this represents the number of modes, which are oscillating in phase, as long as we are assuming that all modes contribute with the same amplitude. Since the ratio is the same for power or intensity, we look at the intensity ratio. The intensity is given by:

$$I(t) = \left| \sum_m A_m \exp^{-im\Delta \omega t} \right|^2$$

where $A_m$ are the field amplitudes of the mode $m$ and $\Delta \omega$ is the mode spacing. For the case of mode locking the maximum intensity is reached if all modes are in phase, which means the exponential term is for all modes equal to one. This gives for the assumption that all modes have the same amplitude $A_m = A_0$:

$$I_{\text{max}} = \left| \sum_m A_m \right|^2 = |A_0|^2 \cdot M^2.$$ 

For the non mode-locked case we have to write the absolute square of the field and do a temporal average over each mode:

$$I_{\text{avg}} = \sum_{m,m'} A_m^* \cdot A_{m'} \left\langle \exp^{-i(m'-m)\Delta \omega t} \right\rangle = \sum_m |A_m|^2 = |A_0|^2 \cdot M.$$ 

The ratio $\frac{I_{\text{max}}}{I_{\text{avg}}} = M$ is equal to the number of excited modes.

### 3 Pump-probe experiments and attosecond pulses

3.1 A typical pump-probe experiment consists of a pulsed light source which is split into two beams. One beam is delayed with respect to the other one and then recombined to a common beam path. The path length difference between the two arms of the setup determines the time delay between the pump and the probe and must be adjustable. A possible setup, such as realized in our laboratory, is shown in Fig. 3. In this case pump and probe path lengths only differ by a few micrometers.

3.2 Light travels the distance $d = 3 \text{ nm}$ in $t = 10 \text{ as}$. In order to perform such an experiment one has to be able to change the length of the probe arm in steps of $3 \text{ nm}$ and the relative stability between the two paths has to be better than that.
Figure 3: Schematic drawing of a pump-probe setup. More details can be found in [5].
3.3 The timescale of a pump-probe experiment is determined by several factors:

a) The length of the used pulses. In general it is not possible to resolve dynamics that is faster than the pulse duration.

b) The stability of the optical setup. Fluctuations of the path length lead to an averaging of the corresponding delays.

c) The design of the delay-generating element. Normally nanosecond time-scale experiments, which require path length differences in the order of tens of centimeters can not be performed with the same setup as femtosecond experiments which require spatial movements in the micrometer range.

3.4 Within the Gaussian approximation, a pulse shorter than 1 fs would require at least 1.83 eV bandwidth (proven above). This condition automatically creates the problem that if the carrier wave is within the visible range of the electromagnetic spectrum, i.e. $1.7 < f_0 < 3.1$ eV ($700 > \lambda_0 > 400$ nm), the low-frequency wing of the spectrum would extend further than 0 eV towards the negative part of the frequency axis. This is physically not possible. Therefore if we want to generate an ultrashort laser pulse while keeping the carrier wavelength in the visible range, we have to abandon the Gaussian approximation. Indeed, sub-femtosecond laser pulses were created and characterized with the carrier wave staying well in the visible range of the electromagnetic spectrum [6, 7]. Its spectrum which is almost top-hat covers more than two octaves, ranging from 270 – 1100 nm. Technical details and its challenges can be found in [8, 7].

On the other hand, if the carrier wavelength is shifted to the XUV range of the electromagnetic spectrum, the above requirement can be physically fulfilled. However, there are technical challenges associated with working in the XUV range, to name a few:

a) The absorption of gases is high, which means that the propagation of XUV is only possible under vacuum conditions.

b) The reflectivity of mirrors is quite low. Best results are archived under grazing incidence.

c) There are hardly any transmissive optical elements available.

4 Reconstruction of attosecond beating by interference of two-photon transitions (RABITT)

4.1 The strong laser field deforms the potential of the atom and allows an electron to tunnel through the barrier (step 1). The free electron is then accelerated in the laser field (step 2). Since the oscillating field changes sign, it is driven back towards the ion and may collide with the parent ion. In this second step the electron acquires energy from the laser field.

If the electron recombines with the parent ion, it releases the additional energy as a photon (step 3). Since this procedure is repeated in every half cycle of the laser field, a train of light bursts is generated. Also because of this, even harmonics interfere destructively if the driving field is long enough (a few cycles or more).

4.2 a) Kerr-lens mode-locked Ti:Sa oscillator.

b) Chirped pulse amplifier (stretcher, amplification stages, grating compressor).
c) Gas-filled hollow core fiber and set of chirped mirrors.

4.3 A thin (200 nm) aluminum foil blocks the IR. Since the divergence of the XUV is much less than for the IR, one can remove a significant amount of IR by using an iris or perforated mirror.

4.4 See figure 4 in the paper.

4.5 a) Reconstruction of the pulse train. The RABITT analysis provides an attosecond pulse duration that is the average over the durations of the individual pulses. The complete attosecond pulse train can be reconstructed by the FROG-CRAB technique [9].

b) Determination of material properties as shown in section 3.2 of the paper.

c) Measurements of atomic phases. The derivative of the atomic (scattering) phase with respect to energy provides the attosecond photoionization delay (see chapter 4 of the lecture). In this way phase differences between different ionized atomic shells are obtained. If the pulse train is known, the scattering phases can also be determined directly.

d) Measurements of ionization delays associated with different electronic states of a molecular cation.

References


