Attosecond radiation sources

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based on the lectures of Fabrizio Carbone and Michele Puppin

Contents

Part I

Lectures

Chapter 1

The main ideas of laser physics

This chapter aims to provide a broad picture of laser physics. The Laser is introduced as a oscillator with an amplifying media. We derive important conditions for laser amplification based on the media (required number of levels, population inversion) and for the cavity (gain/phase conditions). These ideas will be used during the whole course so make sure to understand them.

A Light amplification

The word laser is an acronym standing for *light amplification by stimulated emission of radiation*. Thus, we start our journey in the world of laser physics by emphasizing the challenge of amplifying light. Since we describe lasers, we will stick to the case amplifying oscillators.

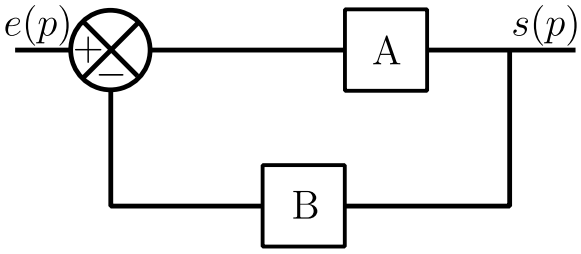

Figure 1.1: General block diagram for any feedback loop in the Laplace domain. Note that A and B can depend on p

A general purpose oscillator can be represented using the block diagram of figure 1.1. The amplification is described by A and the oscillation are generated by the negative

feedback loop, up to a factor of B. Using the formalism of signal theory, we can write that

$$s(p) = A[e(p) - Bs(p)] \quad \Leftrightarrow \quad s(p) = \frac{A}{1+AB}e(p) \tag{1.1}$$
$$= H(p)e(p) \tag{1.2}$$
$$\tag{1.3}$$

where $H(p)$ defines the transfer function of the system. If the system is linear, you can study the system by simply studying the function $H(p)$.

This brief discussion of oscillator physics gives us the two figures of merit of a possible light oscillator, namely a feedback loop and an amplifier. In the case of light, the feedback loop can be achieved using mirrors to send the light back on itself. The amplification on the other hand requires to take advantage of another mechanism : stimulated emission.

Ingredient for oscillator

- A feedback loop in the form of a Fabry-Perot cavity
- An amplifier in the form of stimulated emission.

B Amplification

Before discussing how we can amplify light, we need to discuss the various mechanisms leading to radiations. Following Maxwell's equation, we know that the electromagnetic field originates from a charge movement. For instance, accelerating (Synchrotron) or deccelerating free electrons (Bremsstrahlung) produces such a field. However, the manipulation of free-charges is often experimentally challenging (see for instance the chapter **??** on free-electron lasers).

Another mechanism emitting radiation is of course dipolar transition[1], in which the movement of electrons occurs between different atomic orbitals. This is the historic way of amplifying light, and therefore the one we will study now.

As a first approach, we model our atom as a two-level system, a picture that we will refine in the next paragraphs. This atom interacts with light via thre mechanism :

Absorption A photon is absorbed, an electron goes in an excited state

[1] We briefly recall the results here. Further discussion is to be found in Svelto's book[**?**], chapter 2

Spontaneous emission An excited electron decay toward the ground state, emitting an electron

Stimulated emission A photon triggers the decay of an excited electrons, resulting in two coherent photons propagating in the same directions.

The stimulated emission process is the one allowing amplification, as stated by the acronym LASER, as it increases the total photon number. However, several questions have to be adressed in order to get lasing, for instance :

- How should the spontaneous vs stimulated emission rate compare ?
- What is the condition to have an amplifying media ?
- Is there a limit to the amplification ?

To tackle these questions quantitatively, we will present the general classical framework to study the dynamic of electronic populations in the presence of a light flux.

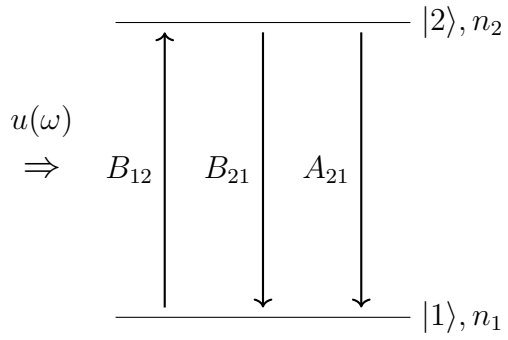

Figure 1.2: Transitions in a two lovel system in the presence of an incoming light $u(\omega)$. B_{12}, B_{21} and A_{21} describe absorption, stimulated emission and spontaneous emission respectively.

1 Two-level system in the steady-state

By reading figure 1.2, we get :

$$\frac{dn_1}{dt} = -\frac{dn_2}{dt} = -B_{12}n_1u(\omega) + B_{21}n_2u(\omega) + A_{21}n_2 \tag{1.4}$$

$$n = n_1 + n_2 \tag{1.5}$$

We are interested in the steady-state solution of the system. The quantity of interest (the one we want to amplify) is the flux. In this case :

$$-B_{12}n_1u(\omega) + B_{21}n_2u(\omega) + A_{21}n_2 = 0 \tag{1.6}$$

$$\Leftrightarrow u(\omega) = \frac{A_{21}}{B_{12}n_1/n_2 - B_{21}} \equiv \frac{\rho(\omega_{21})}{\hbar\omega_{21}} \tag{1.7}$$

where we defined the spectral energy density as $\rho(\omega)$. Assuming that we are at thermal equilibrium, statistical physics tells us that the occupation of a given energy level follows Boltzmann distribution :

$$\frac{N_2}{N_1} = e^{-E_{21}/k_BT} \qquad \Rightarrow \qquad \rho(\omega_{21}) = = \frac{A_{21}}{B_{12}e^{\hbar\omega/k_BT} - B_{21}} \tag{1.8}$$

This last expression should look familiar. It is a less general way of describing Planck's law for Black-body radiation.

$$u(\omega) = \frac{\hbar\omega^3}{\pi^2c^3}\frac{1}{\exp\left(\frac{\hbar\omega}{k_BT}\right) - 1} \tag{1.9}$$

Instead of dealing with various quantified radiations confined in a box, we assume a single quantified radiation between atomic levels. Still, the two formula should identify.

$$\frac{A_{21}}{B_{21}} = \frac{\hbar\omega^3}{\pi^2c^3} \qquad\qquad B_{12} = B_{21} = B \tag{1.10}$$

For the second equation, this holds because we assume no degeneracy.

$$u(\omega) = \frac{\hbar\omega^3}{\pi^2c^3}\frac{1}{\exp\left(\frac{\hbar\omega}{k_BT}\right) - 1} \tag{1.11}$$

Using these result, we can also calculate n_1, n_2 in the steady state, which we can interprete as the media's answer to incoming light

$$n_2 = n_{tot}\frac{Bu}{2Bu + A} \tag{1.12}$$

$$n_1 = n_{tot}\frac{A + Bu}{2Bu + A} \tag{1.13}$$

Let's wrap up what we just did. We treated a physical media as an ensemble of 2 level systems submited to a light flux. We saw that the coefficients describing light-matter interactions aren't independent and can be used to calculate the equilibrium response of the media. This description isWe have introduced here the atom cross-section for absorption σ_{12}) and stimulated emission σ_{21}). They have to be equal according to equation 1.10, as $B = \int_\omega \sigma(\omega) d\omega$. found in many textbooks but misses the key point we want to make : how is light amplified ?

2 Population inversion

To describe actual amplification, we need more something like a balance equation for our medium, in order write something like :

$$I_{out} = H I_{in} \tag{1.14}$$

where H is the transfer function of the medium, hopefully leading to an out intensity greater than one. Following the well-known techniques of infinitesimal calculus, we have according to figure **??** :

$$u(z + dz) = u(z)(1 + \sigma_{21} n_2 - \sigma_{12} n_1) + n_2 A(u_{spont,+} - u_{spont,-}) \tag{1.15}$$

$$\Rightarrow \frac{\partial u}{\partial z} = \sigma u(z)(n_2 - n_1) \tag{1.16}$$

Several comments on this :

- We have introduced here the atom cross-section for absorption σ_{12}) and stimulated emission σ_{21}). They have to be equal according to equation 1.10, as $B = \int_\omega \sigma(\omega) d\omega$.
- We have assumed two directions only for photon emission (forward and backward). The key feature here is that spontaneous emission is isotropic, as opposed to the other mechanisms. Therefore, spontaneous emission doesn't bring any net contribution to the flux and cancel itself.
- Amplification imposes that $\partial u(z)/\partial z > 1$, which translates into $n_2 > n_1$. This is a very important condition known as population inversion.

Back to our 2 level system discussion, we define $N = n_2 - n_1$, and we can readily see using equation 1.13 that $N < 0$. This is an terrible news : we can't have amplification using a two-level system.

3 Toward 3 and 4 level system

Since 2 level aren't suitable for amplification, we have to look for systems with 3 or more levels. This will naturally increase the number of transition rate we have to consider, and the fine interplay between these rates to achieve lasing will be discussed in the first tutorial. REF !! We can nonetheless explain without maths the general mechanisms required in a 3 or 4 level system to achieve amplification.

In the three level case, we pump from level 1 to 3 and we need a very fast decay from 3 to 2, and we want the level 2 to be long-lived. In this case, electrons will start accumulating in this level 2, while the ground state is depleting, leading ultimately to $N = n_2 - n_1 > 0$. This scheme has the disadvantage of emptying the ground state, which can be difficult has many electrons live there.

A four-level system solves this issue as it requires population inversion between the long-lived state 3 and a state which is almost empty, namely 2. To have a proper 4 level laser, we'd like to have a very fast decay from 2 to 1, whereas the 3 level scheme remains untouched.

This theory is nice, but the difficult part is actually to find atoms or systems in real-life that follow such state dynamics, with the proper lifetime between each energy level.

C The feedback-loop

We are done with the basic description of amplification. In principle, we could build our laser by having sufficiently long media in which light gets amplified all the way, which is not convenient and also has some large limitation. The easiest way is therefore to place the amplifying media in some cavity (figure 1.3), so that light passes several time in the medium to reach higher intensity before being released. How does the field evove in this cavity ?

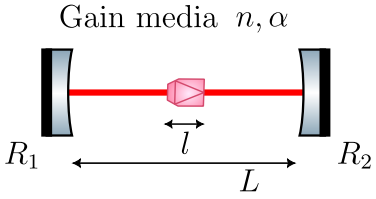

Figure 1.3: The easiest possible laser cavity with an amplifying medium in the center.

To answer this question, we use the ideal case of a plane wave $E(z) = E_0 e^{-ikz)}$,

where k is the wave number. After one round-trip in the cavity, we have that :

$$E(z) = E_0 e^{-2i\omega(L-l)/c} e^{2i\omega(n'+in'')\omega l/c} R_1 R_2 \tag{1.17}$$

This equation has a lot of implications in laser physics. To get lasing, we require constructive interferences between the fields after given number of round-trip. At the threshhold, this implies that :

$$n'' = \sigma_{12} N = \frac{1}{l} \ln(\frac{1}{|R_1 R_2|}) \tag{1.18}$$

$$\nu_p = \frac{pc}{2L'} + c\frac{\phi_1 + \phi_2}{4\pi L'} \qquad L' = L - l + n'l \tag{1.19}$$

In essence, this condition of constructive interference tells that only certain frequency modes are going to lase, provided that they have enough gain.

Chapter 2

Coherence

We saw in the previous chapter that the laser scheme is in essence a way to amplify light. It relies on the stimulated emission process, in which an incoming photon hitting an atom leads to two photons with same energy and momentum. Intuitively, we understand that all photons coming from a laser have a lot in common. If you think of it, this is very different from the usual case of a chaotic light source, where photons have random phases. In this chapter, we want to explore and give flesh to this reasoning by introducing a key quantity in wave physics, namely coherence

A Introduction

1. Elaborate on a model of chaotic source vs laser.
   2. Spatial or temporal picture

B First order coherence

We start our discussion with temporal coherence. We want to construct a quantity that expresses how much a wave originating from an emitter ressembles itself as time passes. A first idea is to measure the wave in amplitude and phase at a time t and then to measure it after some delay τ. We then get :

$$G_{naive}(\tau, t) = E^*(t)E(t+\tau) \tag{2.1}$$

We use the complex conjugate so that for $\tau = 0$, we simply obtain the modulus of the wave. The issue with our observable G_{naive} is that it's defined at a given time, but this time has nothing special. Beyond this, the interest of the concept of wave is

that it describes a flow of energy in time. Therefore, we can integrate this quantity over a time T, where ideally T is as long as possible.

$$G^1(\tau) = \lim_{T\to\infty} \frac{1}{T} \int_T E^*(t)E(t+\tau)dt \tag{2.2}$$

This definition is meaningful for an experimentalist, as it allows one to measure coherence experimentally. However, this definition is utterly inefficient to construct a theory of coherence.

1 Revisiting ergodicity in wave physics

The coherence function we just constructed is an average over time of the field generated by a source. Our idea is to model this source as a large number of individual emitters, in order to use the framework of statistical physics. We will therefore invoke the ergodic hypothesis, which in essence tells us that the time-average is equal to the ensemble average, meaning a probabilistic average over a large number of realization. We shall write :

$$G^1(\tau) = \langle E*(t)E(t+\tau)\rangle \tag{2.3}$$

This definition is way more powerful to construct a theory, because it gives us a recipe to calculate the first order coherence. We start from a probabilistic model for individual emitters, and then we run the calculation of an expectation value. It is in essence an auto-correlation function.

Coherence function using probabilities

This is all we will explain about statistical physics in this lecture. We will see this in greater details during the tutorial. For statistical physics, the interested reader may have a look at ..... Concerning statistical optics, Loudon REFFFF is a good reference.

2 Graphical representation of first order coherence

Let us use our time-average to calculate the coherence function for the easiest type of wave known to man : the plane wave. We write an electric field as $E(t) = Ae^{i\omega t+\phi}$. Then the first order coherence reads :

$$G(\tau) = \frac{A^2}{2} e^{i\omega\tau} \tag{2.4}$$

Since a plane wave is merely a theoretical case, we may need to add a time varying enveloppe $A(t)$ or phase $\phi(t)$. In this case, the coherence usually starts at a maximum and then undergoes damped oscillations. FIGURRE.

3 Wiener-Khinchin theorem

A legitimate question regarding any physical observable is whether or not we can measure it. To measure coherence, we would need to sample the electric field in time with PHz speed, which is unfeasible using simple detector like a photodiode[1]. By chance, we have the Wiener-Khinchin theorem, which states that the spectral power density $S(\nu)$ is simply the Fourier transform of the first order coherence function.

$$S(\nu) = \int G(\tau) e^{i\omega\tau} d\tau \tag{2.5}$$

In this expression, the spectral power density is defined as :

$$S(\nu) = \lim_{T\to\infty} \frac{1}{T} \left\langle \left| \int_T E(t) e^{i\omega t} dt \right|^2 \right\rangle \tag{2.6}$$

You should really take some time to appreciate the importance of this result. We just said that measuring a spectrum, something easy in a laboratory, is a way to assess whether light is coherent or not.

To convince yourself, here is a proof of this theorem. We have to calculate the expectation of the spectral power density.

$$\left\langle \left[\left| \int_0^T E(t) e^{-i\omega t} dt \right|^2 \right] \right\rangle = \left\langle \int u(t) u(t') e^{-i\omega(t-t')} dt dt' \right\rangle \tag{2.7}$$

$$= \int \langle u(t) u(t') \rangle e^{-i\omega(t-t')} dt dt' \tag{2.8}$$

$$= \int G(t-t') e^{-i\omega(t-t')} dt dt' \qquad \text{Definition of correlation} \tag{2.9}$$

$$= \int \Gamma_u(\tau) e^{-i\omega\tau} d\tau dt \qquad \tau = t - t' \tag{2.10}$$

$$= T \times FT[G](\omega) \tag{2.11}$$

[1]Measurement of light is adressed in great detail in chapter 9

Taking the limit when T goes to infinity yields the result.

As a side-note, this explains to you why a plane wave is very coherent and why broadband light isn't. If you have a plane wave, you have a single frequency, leading to oscillation of the coherence function. If you start adding frequencies, you will have interference and coherence measures the time on which all components are in phase. To gain coherence, you can use filtering of frequencies.

4 Michelson interferometer

The previous theorem gives us a way to measure the correlation function. However, there is another method to probe the function $G(\tau)$ directly, using Michelson interferometry. A typical Michelson interferometer is displayed on figure2.1.

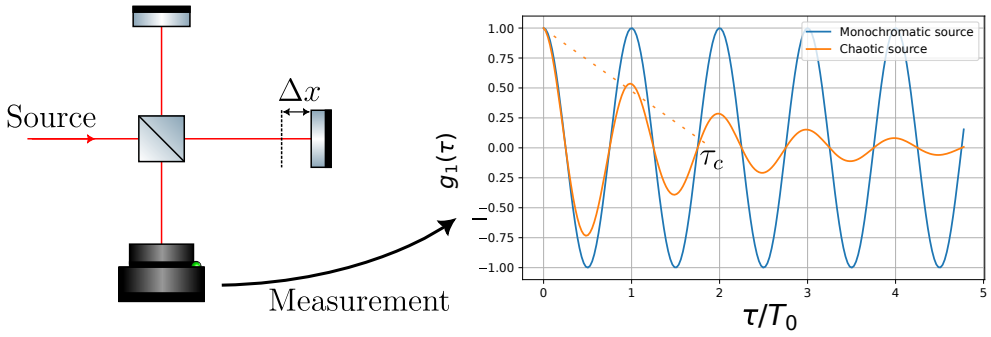

Figure 2.1: Set-up of Michelsion, in which coherence can be measured by moving one arm. On the right, the associated signal is displayed as a function of the delay between the two waves for a coherent and less coherent source.

By moving one arm of the interferometer with respect to the other, we can impose a phase between the two waves, leading to the following intensity :

$$I(\tau) = |E(t) + E(t+\tau)|^2 \tag{2.12}$$
$$= |E(t)|^2 + |E(t+\tau)|^2 + 2E^*(t)E(t+\tau) \tag{2.13}$$

Therefore we see that the result depends on the product $E^*(t)E(t+\tau)$. What we see on our detector is a modulation of intensity when we sweep the delay, which is almost a direct measure of coherence.

A word of caution here. We are presenting here an oversimplistic treatment of the Michelson interferometer, treating it as a simple delay line between our two fields.

However, this set-up has a very good accuracy, and deeper understanding is gained when adding a geometric description of the apparatus (thickness and angle of the Beam splitter). As a matter of fact, this experiment was meant to detect ether, the media allowing propagation of electromagnetic waves. The impossibility to measure sensible variation in the speed of light was an argument against the theory of ether and led to the developpment of special relativity.

5 Spatial coherence

We have seen that a wave can ressemble itself in time. The same idea can be applied in space, leading to the following definition :

$$G(\vec{R}) = \langle E(\vec{r})E(\vec{r}+\vec{R})\rangle \tag{2.14}$$

This concept is best illustrated by Young's slit experiment. FIGURE As you already know, the interference in this experiment originates from the phase difference between waves reaching the same point[2]. Most textbook descriptions of the phenomemon assume a plane wave for Young's slit experiment, so that the two emerging waves are the same.

If this condition isn't fulfilled, at least approximately (say that you can tune the distance between the two slits), your interference pattern will show some symmetry breaking due to lack of coherence. If you were to input maths, CARMINATI optique.

This gives rise to a coherence length for your light.

Next, you can define a coherence volume $V = c\tau_c R_c$. This becomes useful only if

C Higher degrees of coherence

Using the first order coherence function, we move continuously from coherent to incoherent light. Using higher order coherence function, we can instead classify light using the language of phase transition, in which the laser phase is fundamentally different from the chaotic light phase[3].

1 Generalizing coherence

First, we need to generalize our notion of coherence by introducing the n-th order coherence function and degree, defined as :

[2]For good illustration, see 3BLUE1 Brown

[3]For details, see here

$$G_n(X_1, \dots X_n, X'_1, \dots X'_n) = \langle \prod_i E^*(X_i) E(X'_i) \rangle \tag{2.15}$$

$$g_n = \frac{G_n}{\prod \langle I(x_i) \rangle \langle I(x'_i) \rangle} \tag{2.16}$$

In this expression, we introduced the spatio-temporal quartet $X = (x, y, z, ct)$. We introduce X_i, X'_i to stress that coherence is measured between pairs of events (points in time). We could have introduced it by imposing n an even number.

You may check for yourself that G_1, introduced previously, is in agreement with this definition. To differentiate laser light from chaotic light, we have to move to the second order coherence function.

Chapter 3

Brief overview of laser techniques

We have seen the ground principles of laser in chapter 1 and the main figures of merit of a laser in chapter 2. It is now time to leave the abstrac world and apply our formalism to real laser system, meaning solid-state amplifying media, gas laser and the rest. If this chapter looks like a catalog to you, then congrats ! It means that your knowledge of the past chapters allow you to address a wide variety of systems.

A Solid state lasers

As the name suggests, this class of laser has a solid state amplifying media, usually a matrix containing some dopants. It may sounds trivial but as an important consequence. As a matter of fact, we derived properties of laser for a cloud of isolated atoms with specific and well-defined energy levels. As you know, a solid introduces a large degeneracy of this atomic levels leading to the formation of energy bands. Hence, we will sometime deal with bands and still use the term « level », since the lasing transition always occur between atomic levels.

1 The Ruby laser

We start with the first ever created laser, namely the Ruby laser. The operation scheme is pretty much like the ideal case of the first chapter. It consists in an Al_2O_3 matrix doped with Cr atoms. It has a rather poor efficiency, about 10^{-3}. It can be pumped using flashlight or electrical pumping.

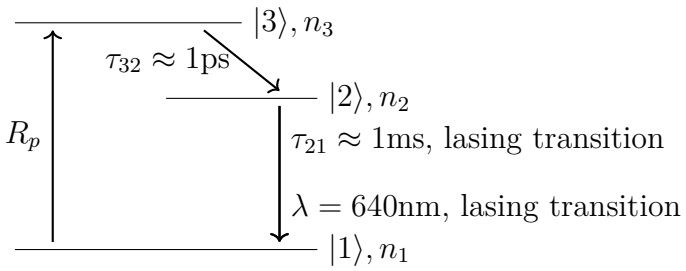

Figure 3.1: Energy levels of the Ruby lasers, showing a long lived excited state allowing population inversion

2 The Nd-YAG laser

This is a superstar in the field : cheap, high efficiency of 3%, easy to manipulate. Nd stands for Neodymium, the lasing atom trapped in an yttrium aluminium garnet $Y_2Al_5O_{12}$. The emission wavelength is $\lambda \approx 1064$nm. Its natural linewidth falls in the kHz range, but due to homogeneous broadening, we have that $\Delta\nu_{vib} = 100$GHz.

Several alternative to this Nd-YAG laser should be mentionned. To change the emission wavelength, we can change the matrix, using for instance YLF (yttrium lithium fluoride). Another possibility is to trap Nd within a glass, which leads to much larger broadening. This can be used in two ways. One way is to produce a tunable source by filtering some wavelength, the other is to use this broad spectrum to go to very short pulses.

This laser are pumped optically using various schemes, as presented on figure 3.2.

3 Semiconductor laser

To discuss semi-conductor laser, we have to take a step back and briefly recap the operating principles of a diode. In a semi-conductor, the valence band is filled with electrons and the conduction band is empty. A diode is in essence a PN junction, that is to say a semiconductor with one side doped with electrons (N doping) and the other is doped with holes (P doping). This results (we admit the result) in a depletion region, in which the bands are twisted, favorizing the creation of electron-hole pairs are created under a bias voltage. The recombination of this exciton creates light. This is the technique underlying LED.

This technology has very high efficiency, ranging typically from 10 to 30%. As a consequence, there is no need for high reflectance mirror, $R \approx 30\%$ is enough, which is about the reflectivity between silicon and air. In other words, a photodiode with

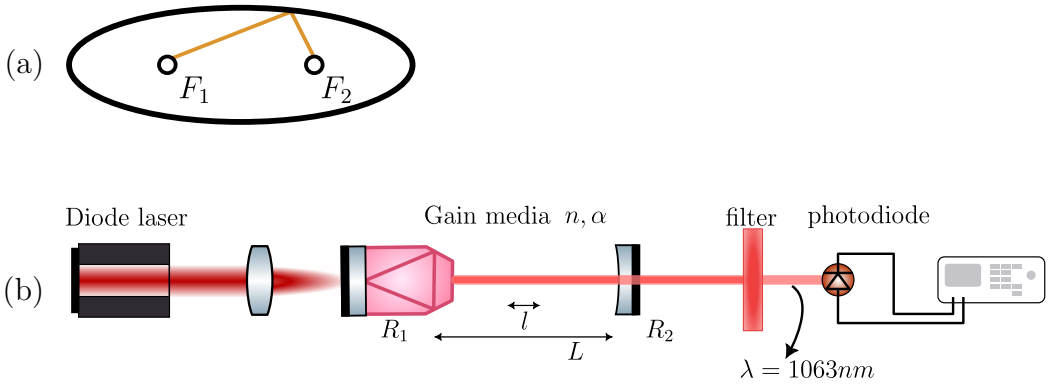

Figure 3.2: (a) Elliptic geometry, a light source is placed in F_1 and the light is used to pump a Nd:YAG crystal in F_2. (b) Laser pumping using a semiconductor laser (pratical set-up)

cavity shape is a laser in itself, and a cheap one at that. Their bandwidth usually fall in the GHz range. The main drawback is their astigmatic emission, since the source is a rectangle, as shown in figure 3.3.

Due to this advantages, they found a broad range of applications. The most common one is to use them as optical pump for other lasers. Many variations are possible for this laser. This is due to the fact that you can engineer the amplifying media using micro-fabrication techniques, allowing high design tunability. We merely quote here quantum cascade laser, monochromatic semiconductor laser and VCSEL.

Quantum cascade laser have been developped in the group of Frederico Capaco. it consists in multiple PN junctions following one another, which allows a tunable gain using cascade emission. Monochromatic laser have a cavity which has the shape of a diffraction grating, that is used to get only one amplified wavelength, killing the others via destructive interference. The VCSEL stands for Vertical Cavity Surface Emitting Lasers. It consists in a mirror at the output of the cavity, allowing emission perpendicular to the emission plane.

B Vibronic Laser

1 Ti-Saphire

We now move to another class of laser in which energy levels are broadened by vibrations. The typical example of such a laser is the Ti-Saphire laser, which consists

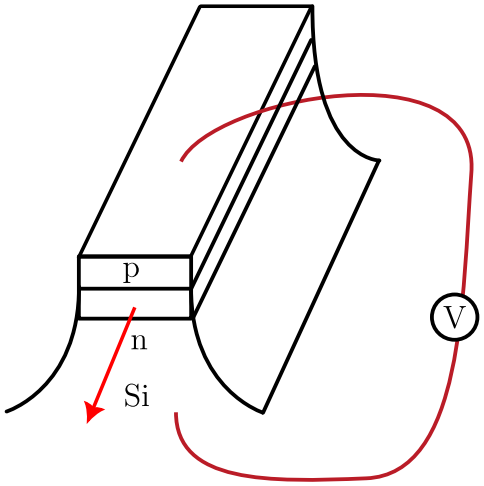

Figure 3.3: A semi-conductor laser schematics. The PN junction is biased, and laser light is emitted in the depletion region (red arrow).

in Al_2O_3 crystal doped with titanium atoms. Due to the level splitting, this laser has a bandwidth ranging from 650nm to 1200nm in continuous wave mode, and 780 ± 20nm in pulsed mode. Efficiency lies aroung 4%.

This broadband source allows tunability in the wavelength, inserting for instance a prism in the cavity to enhance only one wavelength. Conversely, we can use this broadband spectrum to generate short pulses, since the pulse duration is ultimately limited by the bandwidth due to the uncertainty principle.

2 Dye-Lasers

This laser use the vibronic state of a molecule forming a dye. This vibronic state can arise for instance from Jahn-Teller effect. The huge asset of this technique is that to change the properties of the laser, we simply have to change a dye, which is very convenient. This laser are becoming less and less popular due the hazardous chemicals used to produce the dye, and the no less dangerous fact that we let high electric current flows next to this dye.

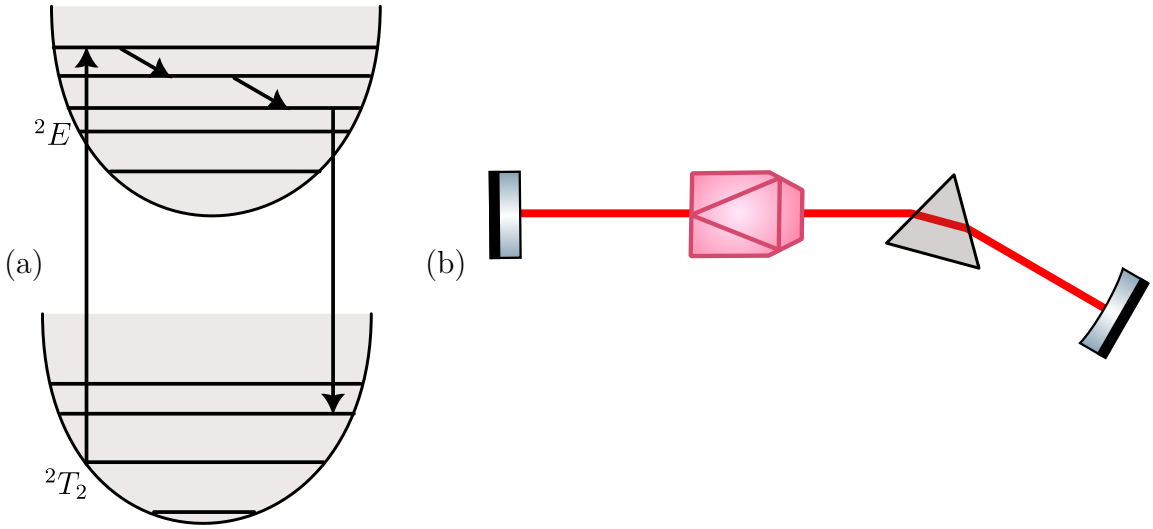

Figure 3.4: (a) Schematic view of the energy levels in Ti-Saph. Due to the large splitting, we can have lots of emitted wavelengths. (b) This wavelengths can be filtered thanks to a tunable wavelength cavity. Using a prism, we can sort the different wavelengths and "purify" our output wavelength.

C Gas laser

D The Helium-Neon laser

Our discussion will be based on the He-Ne laser, whch is the most common laser pointer. This laser has a very narrow band, lying somewhere in the kHz range, which is mostly due to inhomogeneous broadening (Doppler effect). Therefore, this laser is very coherent. It's efficiency lies around 1%.

This laser relies on a mixture of gases. The Helium is pumped using electrical discharge into the excited state 2^1s and 2^3s. This levels Helium are resonant with the $5s$ and $4s$ levels of Neon, which are metastable, meaning electrons can be transferred from Helium to Neon via collisions. This levels will then decay to lower levels of Neon, producing different wavelength (See figure).

To select one precise wavelength, we cut the edge of the He-Ne tube at the Brewster angle, which is wavelength dependent. This allows in addition to get linearly polarized light at the output of the laser.

E The Argon ion laser

This technology is very simple, and quite close to the ideal description of the first chapter. We start by ionizing Argon into Ar^+, and electrical pumping then leads to a classical 4-level scheme. The main asset of this technique is that it sustains very high cw power, leading to about 10W of green light. The efficiency of this laser is however quite low, around 0.1%.

This laser was used to pump every laser back in the days, but the rise of semiconductor laser, with same caracteristics with cheaper prices leads to the decommissioning of this technology. A large issue of this technique is that it requires a very large cooling to remove the heat, which comes next to a large electrical current, making the whole machine quite dangerous.

F Excimers lasers

1 K_2F lasers

We start the discussion with K_2F, which is an excimer, that is to say a short-lived unstable molecule. In this system, population inversion is obtained by electrical pumping. The output wavelength is about 248nm, and efficiency is around 2%. This rather short wavelength in the UV convenient for DUV lithography, as a way to expose a photoresist. It is also used for eye surgery, since the cristallin absorbs in the UV.

2 CO_2 lasers

This is an interesting case, which uses a vibrational transition for lasing instead of an electronic transition. The CO_2 molecule indeed has three vibrational modes, that can be calculated using group theory. Each modes has a precise energy, as reported on the energy diagram of figure 3.5.

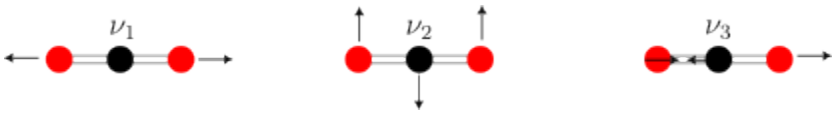

Figure 3.5: Vibrational levels of the CO_2 molecules and associated energy diagram.

The pumping is achieved by direct electron collisions and resonant energy transfer. As the level 3 is metastable, it becomes hard to send the system in the ground state

to pump it again. We therefore add a mixture of N_2 and He to allow proper de-excitation. At a pressure of 10torr, the typical ratio is about $CO_2 : 1, Ne : 1, He : 8$.

Speaking of numbers, we have $\eta \approx 20\%, P = 20\text{W}, \lambda \approx 10\mu\text{m}$.

The last thing we should mention about CO_2 laser is that we need CO_2 circulation in the laser, which is presented in figure **??**.

G Chemical laser

Our example will be the HF laser. In this technique, the energy transition originates from a chemical reaction, namely :

$$F + H_2 \rightarrow HF^* + H \qquad H + F_2 \rightarrow HF^* + F \tag{3.1}$$

The laser transition occurs here due to a decay of the vibrational levels of the HF molecule, leading to photon emission. The emission is around 3nm.

X-ray laser

First, we stress that such a laser has not been experimentally demonstrated yet To obtain a coherent source of X-ray, you can either use free-electron lasers (FEL, see7), or try to find an atomic energy transition that falls in the X-Ray region. A good candidate is a highly charged ion like Se^{24+}. This huge ionization leads to an increase in energy spread between levels, of the order of 20nm.

However, such oxydation can't resist for a very long time, meaning you have to pump very high power. At such low wavelength, new issues arise due to the high dispersion encountered in typical material, making the light way more difficult to amplify.

Another option for this laser would be to use nuclear transitions, that are known to have higher energies.

H Fiber SRS laser

See Aggrawal, it basically relies on Raman scattering using a pump.

Chapter 4

Generation of laser pulses

We have covered so far the main ideas of laser physics, explaining the general scheme leading to light amplification. We then saw how this ideas apply in a wide variety of laser system, reviewing detailed mechanisms on how light can be amplified. In this chapter, we want to start approaching the discussion of pulses generation. To this end, we will give some ideas underlying the transient (time-dependent) analysis of a laser, since pulses are measured in unit of time. We will then discuss two main ideas to produce pulses, namely Q-Switching and mode-locking.

A Transient analysis

1 Do we reach steady-state ?

In the previous chapters, we have discussed every system assuming monochromatic wave, possibly with some broadening, which only makes sense if we reach a steady state. Let us discuss briefly a possible transient scenario for a 4-level laser :

1. Pumping with a rate R_p increases the number of electron in state 3. The increase is linear with time.

2. When we reach population inversion, ϕ starts to increase.

3. The increase of ϕ is depleting level 3, reducing population inversion, possibly down to 0.

4. Now population inversion means no light, therefore we stack electrons again in state 3. And so on and so forth

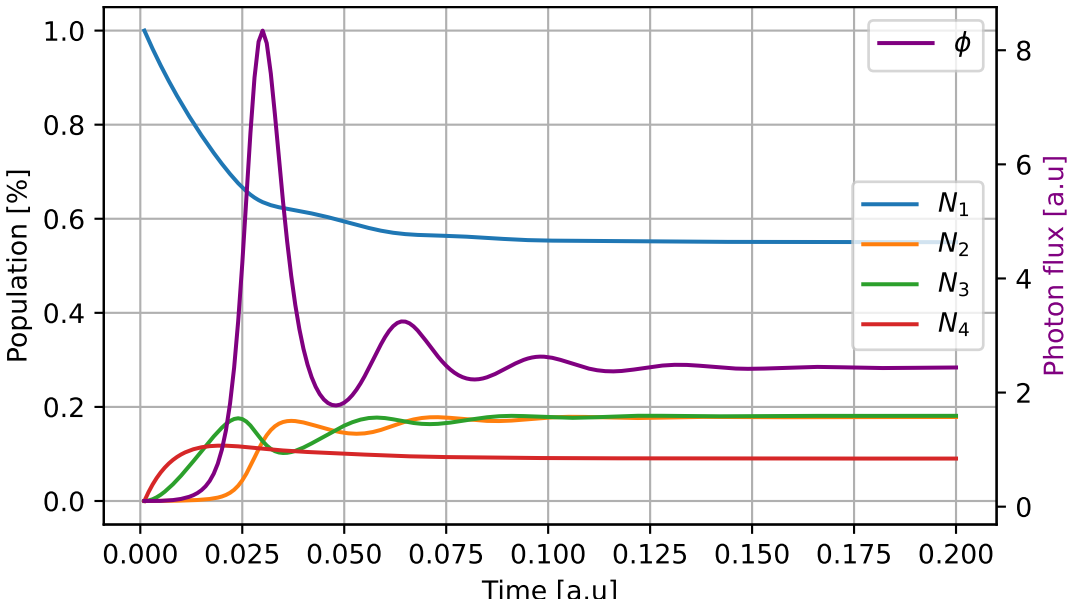

Figure 4.1: Simulation of spiking in a 4-level system. Several ripples are observed in the flux intensity before stabilization. Playing with the parameters, it is possible to have inherently unstable situation.

Playing with the respective decay rate of a 4-level system, using exactly the same model that we described in chapter 1, we can demonstrate such spiking effect, as shown on figure4.1. In some real system, like the ruby laser, the situation is actually never stable and light pulses are emitted following a poissonian distribution. In other system like Nd:YAG, it convereges after a few oscillations.

2 Around pulses

We just saw that pulses are expected in a laser system, but they occur with random phase, intensity and even delay, which isn't acceptable in some cases of experiments like ultrafast science. Therefore, we would like to engineer the system to achieve better control of the pulses. Eventually, these techniques will apply also to continuous-wave laser that reach a non-spiking steady-state.

B Q-Switching

The first idea to generate pulses is to switch the losses of the cavity on and of, hence modifying the quality factor of the cavity Q. We therefore speak of Q-switching. In this technique, the repetition rate and the pulse duration is fixed by the loss-modulating technique. Furthermore, since we pump the system at a constant rate without amplifying light, it means that we can build up a large population inversion, leading eventually to high energy pulses.

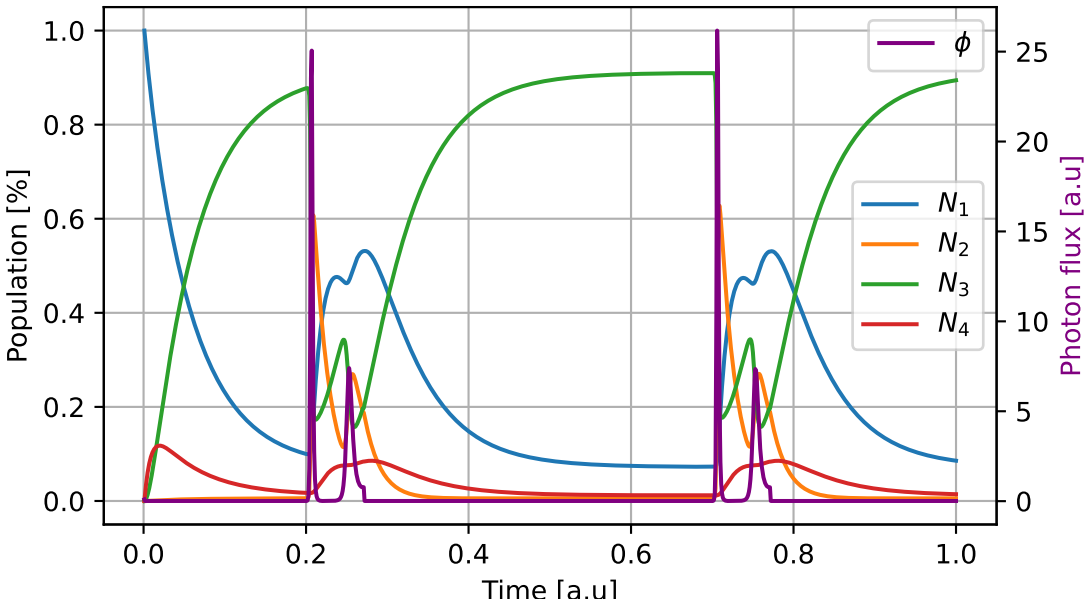

Figure 4.2: Simulation of Q-switch in a 4-level system. The inversion of population is build up for a given time, and then suddenly released. This leads to a sizeable short pulse. The process is then repeated at the repetition rate of the Q-switch system.

We will review here several suitable techniques for Q-switch.

1 Electro-optical

This relies on the Pockels effect, *i.e* the change in the optical index of a material resulting from a voltage. We typically use a non-linear crystal such as KDP (potassium dihydrogen phosphate) or Lithium niobate, and the effect of the Pockels cell is to

rotate the polarization of the light, since $\Delta\phi = k\Delta nL$. In that sense, you can think of it as switchable quarter waveplate. If you place a polarizer, light will be either transmitted or absorbed based on the state of the Pockels cell. The typical pulse duration is about 20ns, as we are limited by electronics.

2 Acoustic-optic switches

Here, we change the refractive index of the material using a pressure wave, putting less constrains on the media we want to use. It then acts like a diffraction grating, meaning the light don't see a cavity anymore. To launche the pulse, we stop the pressure wave, and the cavity is restored. Such a technique has low optical losses, which is interesting for high power applications.

3 Saturable absorber

The two previous cases require an external control over the pulse emission, and falls therefore under the denomination of active Q-switch. We can also have passive Q-switch, mainly using a media that becomes transparent at saturation intensity, and then start bleaching due to spontaneous emission.

This can be treated mathematically, using that $\frac{dI}{dz} = -\alpha(I)I$. Contrary to Beer-Lambert's law, you have α that depends on the intensity. If we model the saturable absorber as a two-level system, we see that if the flux becomes too high, the absorber becomes transparent (cf. tutorial 1).

A limit of passive Q-Switch is that you don't control exactly the pulse emission, and based on the quality of your set-up, you can have rather large jitter.

4 Gain switching

Analogous to Q-switching, we can also play with the gain of the system. This gain depends on the population inversion, which itself depends on the pump rate R_p, which we can tune. However, this technique is rather slow, and not so used anymore.

Is there a difference between gain and Q-switching?

If we have high losses, then we don't amplify, meaning the gain as to be low, right ? If you think this way, you should go back to your favorite signal theory textbook. We model our laser as an oscillator with losses, meaning it follows a differential equation of the form :

$$\frac{1}{\omega_0^2}\ddot{s}(t) + \frac{2\xi}{\omega_0}\dot{s}(t) + s(t) = Ke(t) \tag{4.1}$$

In this expression, we define K as the static gain, the one we use in gain switching. ξ refers to the losses, and is modulated in Q-switching

C Mode Locking

We discussed in chapter 1 the spectral profile of a laser. We can have several oscillating cavity modes at the same time. Most of the time, This give rise to a poor output beam intensity, since we don't synchronize those modes. In other words, all these modes are emitted with random phases.

We will see that if we can engineer the system to have all of our modes in phase, we can produce very short pulses separated by a well-defined delay. Following this discussion, we will discuss several techniques suitable to realize mode-locking in practice.

Qualitatively, if we have N modes and a given bandwidth, then the modes are evenly separated by a frequency $\Delta\nu = \Delta\nu_L/N$, then the output is periodic with period $1/\Delta\nu$ and consist in a serie of pulses of duration $1/\Delta\nu_L$. Now, if we have a laser with a very large bandwidth (gas, dye or solid-state lasers), we can go to very short pulses and very high power.

1 Frequency behavior

The general form of the electric field if we consider N modes is given by :

$$E(t) = \Re(\sum_m E_m e^{i\omega_m t + \phi_m}) \tag{4.2}$$

We recall that modes for a FP cavity obey the relation $\omega_m = \omega_0 + m\Delta\omega_0$, with spacing $\Delta\omega_0 = \frac{2\pi c}{2Ln}$. For a 25 cm cavity, this falls in the GHz range. If the modes

are uncorrelated, we have an intensity that reads $I(t) = \overline{I}(t) + \Delta I$, which looks like speckle. If we now work on the modes, imposing $E_m = E_0$ and $\phi_m = \phi$, it turns out that :

$$I(t) = \Re(\sum_m E_m e^{i\omega_m t + \phi_m}) \tag{4.3}$$

$$= |E_0|^2 |\sum_m e^{i\omega_m t} e^{\phi}|^2 \tag{4.4}$$

$$= I_0 \left| e^{i\omega_0 t} \frac{1 - e^{iN(\Delta\omega_0)t}}{1 - e^{i\Delta\omega_0 t}} \right|^2 \tag{4.5}$$

$$= I_0 \frac{\sin(N\Delta\omega_0 t/2)}{\sin(\Delta\omega_0 t/2)} \tag{4.6}$$

Hence, we see that we obtain light pulses of duration $\tau = T/N$ with period $T = \frac{2\pi}{\Delta\omega_0}$. FIGURES !! shows the peak intensity as a function of time for different numbers of modes. As you can see, the more modes, the shorter the pulse and the higher the pulse power. The remaining question for any experimentalist is the following : How to synchronize the modes in practice ?

2 Active mode-locking

In the case of active mode-locking, we use an external source to synchronize our modes. We will limite our discussion to amplitude modulation, in which the losses are oscillating at a frequency $\Delta\nu = c/2Ln$. You can work out the math to derive synchronization, but there is a more intuitive way to explain this phenomenon.

Consider a light pulse (with lot of frequencies) travelling through a media with non-uniform losses. Then, some parts of the pulse will be depleted, and others will be transmitted (FIGURE). Repeating this scheme, what we obtain in practice is a very narrow pulse sitting at the minimum of the losses.

3 Passive mode-locking

In the passive case, we want the system to synchronize the modes by itself, that is to say produce lasing in pulsed mode only. To this end, several approach are possible, but we will limit ourselves to Kerr-lens mode-locking.

The Kerr-effect refers to the fact that the optical index is a function of intensity, $n(I) = n_0 + n_1 I$. Now, the intensity of the beam we send through the media has some transverse structure, typically $I(r)$ can have a gaussian profile. Therefore, different

region of the beam will experience different $n(r)$. It can be shown (cf exercices) that this leads to self-focusing of the beam, *i.e* the media behaves as a lens. The idea to achieve mode-locking is to then place an iris at the focal length of the media to let the self-focused beam pass while blocking the other components of the light. This way, we will give more amplification to the pulse compared to the continuous wave regime.

Tutorial 4

We will see the example of mode-locking in practicals based on Ti-Saph.

4 Cavity dispersion

Now that we started discussing short pulses, the question is : How short a pulse can ultimately be ? To answer, we have to consider dispersion in the cavity.

First, why should a pulse experience broadening ? We can define several velocities to describe a wave, the phase and group velocities. The phases are traveling at different speed, so that the pulse broaden. To quantize this broadening, we define the group velocity dispersion.

$$GVD = \left(\frac{d^2\beta}{d\omega^2}\right)_{\omega_L} = \left(\frac{d(1/v_g)}{d\omega}\right)_{\omega_L} \qquad \beta \text{ propagation constant}$$

With this tool, you can show how limited a pulse can be and think of optical set-ups to compensate those issues.

Chapter 5

Pulse amplifiers

We saw in the past chapter how one is able to generate light pulses. We now want to look at two main features of pulsed laser, the first one being pulse amplification and the second being propagation.

We want to look at the physics of pulse laser. We characterize a pulse using mainly 3 values :

Pulse duration the full width at half maximum of the pulse

Average power the power averaged over many pulses in time

Peak power maximum power in a pulse

In laser physics with mode-locked ([Ti-saph](Ti-Saph-Laser)) or Q Switch (Nd-YAG), this peak power is typically in the MW range, meaning we have to take into account non-linear and perturbative effects. Upon reaching GW peak power, you have other effects that we won't discuss here.

A Master oscillator / Power amplifier

One main difficulty for pulse amplification is that you lack a suitable pump power for such high intensity. The idea is therefore to use a pulse picker to select one pulse, and then send this pulse through an amplifying media.

1 Model

Such a media can be described as a 4-Level system and in general, one chooses a media s.t $\tau_1 < \tau_p < \tau_2$. The amplification then relies on stimulated emission. To gain

understanding, it is possible to derive differential equations for N, I in this amplifier.

$$\frac{\partial N}{\partial t} = I\frac{N}{\Gamma_s} \qquad \Gamma_s = \frac{h\nu}{\sigma} \tag{5.1}$$

$$\frac{\partial I}{\partial t} = \left(\frac{\partial I}{\partial t}\right)_{se} + \left(\frac{\partial I}{\partial t}\right)_{losses} + \left(\frac{\partial I}{\partial t}\right)_{flux} \tag{5.2}$$

$$= \sigma I N - \alpha I - \frac{\partial I}{\partial z} \tag{5.3}$$

This is usually hard to solve analytically. Assuming one pulse per time, we have that:

$$N(z,t) = \ N_0 e^{-\Gamma/\Gamma_s} \qquad \Gamma = \int_0^t I(t)dt$$

Next, integrating the second differential equation with respect to time, we get :

$$\frac{d\Gamma}{dz} = g\Gamma_s(1 - e^{-\Gamma/\Gamma_s}) - \alpha\Gamma$$

Assuming small losses, we can even derive the output energy :

$$\Gamma(l) = \Gamma_s \ln(1 + e^{-gl}(e^{\Gamma_i n/\Gamma_s} - 1))$$

We can make several comments on this :

1. Based on the value of Γ_{in} wrt to Γ_s, we observe two distinct regime. The first one gives linear gain amplification, the second one is then saturated.

2. Since the tails of a pulse have by definition less intensity, they undergo higher amplification, leading to a time-broadening of the pulse

3. This expression doesn't account for the shape of the pulse. As the pulse passes, N is depleted, which means the back of the pulse is less amplify.

4. To get a more accurate description, we need to take into account the internal losses. We can note that usually, optical damage arise before the losses ($\Gamma_d = 10J/cm^2$)

5. Thermal and non-linear effect tend to distort the pulse in time.

6. The gain medium itself can have parasitic lasing, since it lives with population inversion.

2 Mirrorless laser

We should mention here that such an amplifier can in fact behaves as a laser. Usually, for a rod-like cavity, the gain might be written as $G \propto e^{\sigma N l}$. This means that we can have a low directivity laser in this system ($\Omega = \frac{\pi D^2}{4l^2}$).

3 Geometry of the amplifying media

You can have :

Rods Standard and high gain

Slabs High gain and better cooling due to higher surface

Disks Very efficient cooling but low gain since it's a reflecting amplifier

Fibers immune to thermal effects, high gain, easy in set-up, but large non-linear effects due to the small core

B Amplifying system in cascade

Usually, the amplifiers are limited in gain. Instead of asking too much of one amplifier, the idea is to use a chain of amplifiers to achieve the same gain with less distortions. The first amplifiers will have high gain and distortion, the last amplifiers will have low gain and distortion. In between, you want to use filters to shape the pulse.

1 Optical isolators

As discussed before, you can have parasitic lenses in the amplifiers. Another possibility is that a fraction of a laser pulse is reflected at the input of an amplifier and retro-propagate into the past optical system. Bottom-line : You will have a beam propagating in the wrong direction, and getting amplified in the wrong direction. To avoid this, you want to use an optical isolator.

![image](uploads/c379a4ae03f817b9d121af0823452429/image.png)

By using a waveplate, you turn the polarization of the light, meaning the beam can only propagate in one direction.

2 Regenerative amplifier

![image](uploads/393ccc7393f382a3d3c6f04ac3879840/image.png)

This amplifier uses the same gain medium several time using the switching of a Pockels cell. - When the cell is on, an incoming light pulse will have proper polarization to stay trap in the system. - When the cell is off, the light pulse is expelled from the system.

This way, you can use the same amplifier to amplify the beam up to saturation.

3 Multi-pass amplifiers

The idea of this amplifiers is focus the pulse in different regions of the amplifying media (using a large media), so that the same media contribute to the amplification via different regions. However, aligning the optics for this set-up is rather complex.

![image](uploads/4b5d20590414e3cfaced8aa7f3f9874d/image.png)

C Chirped pulse amplifier

Taking into account Kerr-non-linearity, at some point it becomes impossible to amplify a pulse any further. The non-linearity becomes simply too large. Self-focusing becomes the ultimate limit for pulse amplification. To quantify this, people use the B-integral, defined as :

$$B = \frac{2\pi}{\lambda} \int_0^L n_2 I(z) dz$$

We want to minimize this quantity. To this end, we start by stretching the pulse, meaning lower peak intensity. Then, we amplify this easier pulse that has less self-focusing effect. Last, we contract the pulse again using a pair of gratings.

D Pulse propagation

1 Why is it relevant

When discussing wave propagation, most textbooks assume a plane wave. Then, the discussion rapidly shifts toward properties of the dielectric function of the encountered media, the wave incoming angle, its polarization and so on. This properties are all wavelength dependent, and a pulse is in essence a wavepacket containing lots of different wavelength. The shorter the pulse, the more wavelength, and the more

dispersion we may face. A dramatic example of this is that a 5fs pulse at the laser output, when travelling through few cm of glass, can be as broad as 100fs. Therefore, we need to revisit Fourier signal theory to study pulse propagation.

2 Maths

The basics of the pulse propagation are well-captured by Fourier-theory. The idea is to define a pulse like :

$$E(t) = \sqrt{I(t)}e^{i\phi(t)} \Leftrightarrow \tilde{(}E)(\omega) = \sqrt{S(\omega)}e^{-i\phi(\omega)} \tag{5.4}$$

Then, we focuse the discussion on the spectral phase $\phi(\omega)$, which is the main feature that determines the shape of a pulse.

Tutorial on pulses

This aspects will be treated in great detail during the exercice session. The curious reader may look at the notebook, itself largely inspired from the femtoUp summer school.

More details in ursula Keller

3 Example : Gaussian pulse

We define a Gaussian pulse as an enveloppe with a fast oscillating wave.

$$E(t) = e^{-\Gamma t^2}e^{i\omega_0 t} \tag{5.5}$$

We want to compute the associated

E Measuring the duration of a pulse

Chapter 6

Non-linear optics in laser physics

The first purpose of this page is to summarize Michele's lecture in the Laser course. It will grow over time to add content on this huge topic.

A Perturbative non-linear optics

1 Example of non linear system

One of the easiest sytem that exhibits non-linear wave behavior is a speaker amplifier. If you ask for too much amplification, the system will saturate, and the consequence of this saturation is the creation of new harmonics in the spectrum.

![image](uploads/c60a63b75e13b698cf19faf9c6c2c21d/image.png)

Another example is the anharmonicity of nuclear vibration. Usually, you solve quantum mechanical problem assuming a parabolic potential, but in fact this potential is asymetric, leading to non-linear effect.

2 The case of light

For the light, linearity means that the polarization is related to the electric field up to some tensor $\chi, \vec{P(t)} = \epsilon_0 \chi \vec{E(t)}$. Here, we assume instantaneous response of the media when the field is applied, which is of course a simplification.

Of course, the polarization doesn't have to follow the electric field linearly. In the contexte of weak perturbation, we may use the physicist's favorite tool, the Taylor-expansion.

$$P(t) = \epsilon_0(\chi^{(1)}E(t) + \chi^{(2)}E(t)^2 + \chi^{(3)}E(t)^3 + \dots$$

To keep the discussion simple, we assume scalar fields here. For a more rigorous definition taking into account the vector nature of the field, see [here](Formal approach of non-linear optics). Usually, we have that $\chi^{(2)} \ll \chi^{(1)}$, and therefore we have to reach very large field ($\approx 1/(\chi^{(2)})$ to observe non linear-effects. This explains why the field of non-linear optics was boosted by the invention of laser, allowing such high fields.

3 The non-centrosymmetric condition

The previous paragraph defines the susceptibility as a pure mathematical construction, a fitting parameter. It tells nothing about the physical properties modulating this parameter. Such a treatment is beyond our scope, but we can still mention that a centro-symmetric crystal implies a vanishing $\chi^{(2)}$. Indeed, this would imply that :

$$-P(t) = \chi^{(2)}(-E(t))^2 = \chi^{(2)}(E(t))^2 = P(t) \Rightarrow \chi^{(2)} = 0$$

This restricts the type of crystal we may use for this type of studies.

B Frequency generation

C The zoo of mixing effects

Consider an electric field made up with two plane wave at frequencies ω_1, ω_2 entering a non linear crystal. At the output, you generate polarizations at : - $2\omega_i$, aka second harmonic generation - $\omega_1 \pm \omega 2$, aka sum-difference frequency generation - 0, aka optical rectification

If you now remember that the polarization acts as a source term for the wave equation, it becomes clear that the crystal generates fields at various frequencies.

D Parametric processes

In a parametric process, there is no exchange of energy/ momentum between the field and the system (crystal). In other words, the crystal allows the mixing but isn't contributing in the energy balance or the rate equations. The previously mentioned process are parametric processes

The canonical process in those systems is second harmonic generation (SHG), in which two photons at a given frequency combine to produce a new photon at twice this frequency.

Such processes can be used for optical parametric amplification. You have a large input beam that feeds the signal you want to amplify. This is widely used in fs laser physics to recover a high fluence after some frequency generation (which leads to large energy losses).

![image](uploads/149c60997aef90930a33e74ee650e685/image.png)

To go beyond OPA, the concept of OPCPA has been introduced. To this end, you split a pulse in two. One will be used as pump after undergoing SHG, and the other will be used as signal amplified by this pump. That way, you don't have the limitation of the chirped pulse amplification.

1 Non-linear non-parametric processes

The easiest and already mentionned example is saturable absorption. In this case, the material indeed gains energy.

Another example is two photon absorption. A single photon couldn't be absorbed by the material, but two photons can.

2 Treatment in classical electrodynamic

We assume again a three wave mixing process (SFG). The core idea is to solve the wave equation using the polarization as a source term, which will mix the various components of the field.

$$-\Delta E - \frac{n^2\omega^2}{c^2}E = \frac{4\pi\omega^2}{c^2}P_n^{NL}$$

We can't write the fields as plane wave anymore, since their amplitudes are varying as they propagate in the material. We write $E_i = A_i(r)e^{ikz}$, which leads to the equation :

$$-\frac{d^2A_3}{dz^2} + 2ik_3\frac{dA_3}{dz} = \frac{-16\pi d_{eff}\omega_3^3}{c^2}A_1A_2e^{i(k_1+k_2-k_3)z}$$

Then, in the slowly varying wave approximation (neglect second order derivatives) :

$$\frac{dA_3}{dz} = \frac{8i\pi d_{eff}\omega_3^3}{k^3c^2} A_1 A_2 e^{i(k_1+k_2-k_3)z} \tag{6.1}$$
$$\frac{dA_1}{dz} = \frac{8i\pi d_{eff}\omega_3^3}{k^3c^2} A_3 A_2^* e^{i(-k_1-k_2+k_3)z} \tag{6.2}$$
$$\frac{dA_2}{dz} = \frac{8i\pi d_{eff}\omega_3^3}{k^3c^2} A_3 A_1^* e^{i(-k_1-k_2+k_3)z} \tag{6.3}$$
$$\tag{6.4}$$

This equation can be exactly solved in the undepleted wave limit. Solving this equation leads to a quadratic growth of intensity in the medium as a function of L. This approximation is valid if we are matched, and if the crystal is short. If you let a very long crystal, the field will oscillate between frequencies as a function of distance (plot)

3 Phase matching conditions

So far, we didn't impose any geometrical considerations on this system. However, this is crucial to understand non-linear materials. We saw that parametric process rely on a phase matching condition, meaning $n_1\omega_1 + n_2\ \omega_2 = n_3\omega_3$ and $\omega_1\ + \omega_2 = \omega_3$. However, we know that n is an increasing function of frequency. To fulfill the previous condition, we can : - place ourselves near absorption where n starts fluctuating (anomalous dispersion) - Use birefringent properties of the media. This means that the optical index depends on the polarization of the incoming light. This holds in many crystals, but not cubic ones as they are fully isotropic !

| System | Optical classificiation | | —— | ———————— | | Triclinic, Monoclinic, Orthorombic | Biaxial | | Trigonal, tetragonal, hexagonal | Uniaxial | | Cubic | Isotropic |

Birefringence

Birefringence will be discuss in more details in the incoming tutorial, both experimentally (you will have to optimize the angle) and theoritically (working out the math of type I and II phase-matching and using SNLO).

Chapter 7

Free electron laser

This page summarizes the physics of Free electron lasers (FEL). There is a book to go beyond, UV and Soft XRAY FEL (Schmüser, 2009)

This starts with the need of developing lasers at very low wavelength, typically X-Ray (200eV - 300keV). This way, you get a coherent X-ray source to study a lattice, since $\lambda \approx 10^{-10} m$.

A The tool kit

To perform computations, we will need several formula :

$$\gamma = \frac{1}{\sqrt{1-(v/c)^2}} \qquad E^2 = p^2c^2 + (mc^2)^2 \tag{7.1}$$

$$E = \frac{hc}{\lambda} \qquad = \gamma mc^2 \tag{7.2}$$

$$p = \gamma mv \tag{7.3}$$

B The historical idea : Inverse Compton scattering

The basic idea is to change the photon wavelength using charged particles. Therefore, we model the scattering event between one electron E, p and a photon of energy $h\nu$. After scattering, the electron has energy E' and the photon has energy $h\nu'$. We assume : 1. $E, E' >> mc^2$, that is the electron is and remains relativistic 2. $E - E' = \Delta E \ll E$, which allows for Taylor expansion.

We want to compute the change of energy for the electron to first order.

$$\Delta E = \frac{dE}{dp}\Delta p \qquad E = \sqrt{p^2c^2 + (mc^2)^2} \tag{7.4}$$

$$\frac{1}{2}\frac{c^2 \times 2p}{\sqrt{p^2c^2 + (mc^2)^2}}\Delta p \tag{7.5}$$

$$\frac{pc^2}{E}\Delta p \tag{7.6}$$

For the photon, we have :

$$\Delta E = h\nu' - h\nu \tag{7.7}$$

$$\Delta p = \frac{h}{c}(\nu' + \nu) \tag{7.8}$$

Since we have to conserve energy and momentum, we can use this two quantities into the equation for electrons. We get :

$$h\nu' - h\nu = \frac{pc^2}{E}\frac{h}{c}(\nu' + \nu) \qquad \frac{pc}{E} = \frac{v}{c} \tag{7.9}$$

$$\Rightarrow \left(1 - \frac{v}{c}\right)\nu' = \left(1 + \frac{v}{c}\right)\nu \tag{7.10}$$

Rk : We need a counter-propagating photon in the description. Otherwise, we can't realize any frequency conversion. This is in fact due to the fact that we can't fulfill momentum conservation if we don't add the photon.

As a rule of thumb, we have $\nu' = 4\gamma^2\nu$. Speaking of numbers, if we aim for $\lambda' = 10^{-10}m$ starting from IR laser $\lambda = 1\mu m$, we need $\gamma = 50$.

We can also see that with this equation, we fulfill the relativistic limit $v_e \approx 0.9998c$ and $\Delta E \approx 10keV \ll 25MeV \approx E$.

So everything good right ? We have a suitable way of converting light frequency using charged particles, and it depends on very simple parameters. Sadly, we overlooked a key quantity in light-matter interaction : the cross-section, which is veryyyy small in this case. Therefore, this process is very inefficient in terms of energy. Therefore, we have to look at another set-up.

C The undulators / wiggler set-up

Instead of having an electron interacting with only one photon, we use an undulator which is a series of magnets with alternated axis. The electron is therefore accelerated up and down, thus emitting a radiation. The nice thing is that you can play with this magnet in your experiment, giving you a new degree of freedom.

Therefore, we model an electron propagating in a field $B_y(z) = B_0 \cos\left(\frac{2\pi}{\Lambda} z\right)$. In reality, the field would be more rough.

We model the interaction classically, meaning that $\vec{F} = -e\vec{v} \wedge \vec{B}$. We further assume that $p_z \gg p_x$. Then:

$$p_x(z) = p_x^{(o)} \sin\left(\frac{2\pi}{\Lambda} z\right) \tag{7.11}$$

$$\frac{dp_x}{dt} = p_x^{(o)} \cos\left(\frac{2\pi}{\Lambda} z\right) \frac{2\pi}{\Lambda} \frac{dz}{dt} \tag{7.12}$$

$$= -e\vec{v} \wedge \vec{B} \tag{7.13}$$

$$\Rightarrow p_x^{(0)} = -evB \frac{\Lambda}{2\pi \langle v_z \rangle} \tag{7.14}$$

The mean value for velocity along z comes from our assumption that the electron has very small momentum in transverse directions. Another quantity of interest is the maximum angle the electron is having wrt the z-axis.

$$\theta_m = \frac{p_x}{p_z} \approx \frac{eB}{2\pi} \frac{v}{v_z} \frac{\Lambda}{p} \approx \frac{eB\Lambda}{2\pi cm\gamma} = \frac{\kappa}{\gamma}$$

We defined here κ as the undulator factor. It depends only on physical constant and construction parameters. To get strong emission, you want a large κ. However, you can see that you mainly emit when you change the momentum in the x-direction. If κ is too large, you get two different sources, leading to much broader emission. We therefore have : - The undulator : stays self-superimposing - The wiggler : With two emission spots

The typical undulator has $\kappa \approx 1 - 4$. For synchrotron, it can be as high as 150. We still miss a part of the picture, which is the link to emitted radiation. To perform this, we have to compute the trajectory of the electron.

$$v_z = \frac{p_z}{\gamma m}\sqrt{1-\frac{{p_x}^2}{p}} = v\sqrt{1-\left(\frac{\kappa}{\gamma}\right)^2 \sin^2\left(\frac{2\pi z}{\Lambda}\right)} \tag{7.15}$$

$$= c\sqrt{\left(1-\frac{1}{\gamma^2}\right)\left(1-\sin^2\left(\frac{2\pi z}{\Lambda}\right)\right)} \tag{7.16}$$

$$= c\left(1-\frac{1}{2\gamma^2}\left[1+\kappa^2\sin^2\left(\frac{2\pi z}{\Lambda}\right)\right]\right) \tag{7.17}$$

We have a deviation around an average position. If we perform the same computation, not in the lab frame but in the electron frame, we see that electron follows a kind of 8 shape, and in the proper limits, it gives a dipolar type of radiation.

1 End of undulator physics

Last time, we discussed the fluctuations in the speed of the electrons along x, z and how this means that we have emission of light. However, we didn't actually connected this parameters to the emission wavelength of the system. To do this, we write the period in the rest frame of one electron, which reads :

$$T' = \frac{\Lambda'}{\langle v_z\rangle} = \frac{\Lambda}{\langle v_z\rangle}\frac{1}{\gamma}$$

In our case, we want the wavelength, or the frequency, in the lab frame, meaning we have to correct the Doppler shift :

$$\nu = \sqrt{\frac{1+\beta}{1-\beta}}\nu' \tag{7.18}$$

$$= \frac{\langle v_z\rangle}{\Lambda}\frac{1}{1-\frac{\langle v_z\rangle}{c}} \tag{7.19}$$

$$= \frac{c}{\Lambda}\frac{1}{\frac{1}{2\gamma^2}\left(1+\frac{\kappa^2}{2}\right)} \tag{7.20}$$

$$\tag{7.21}$$

This translates in wavelength

$$\lambda = \Lambda\frac{1+\kappa^2/2}{2\gamma^2}$$

This means that to go to angstrom size for the wavelength, considering $\Lambda \approx 10^-2m, \kappa \approx 1-5$, we need to accelerate the electrons in the GeV range.

2 Compare ICS to undulators

We want to make here a simple mathematical argument explaining why ICS emits weaker field than an undulator.

For an undulator, we have $\kappa = \frac{eB\Lambda}{2\pi mc}$, and one can show that $P \propto \kappa^2$, and $\lambda' = \frac{\Lambda}{2\gamma^2}(1+\kappa^2/2)$.

On the other hand, for ICS, we have $P_{ICS} \propto \frac{E_0^2\lambda^2}{4\gamma^2c^2}$, and $\lambda' = \frac{lambda}{4\gamma^2}$.

This tells us that for the same wavelength, ICS require less energy per electron than undulator, which seems nice. However, it emits way less power than an undulator, making it unsuitable in practice.

3 The FEL

Now that we have seen the undulator, there is only one small ingredient we miss to describe the FEL. This ingredient is the interaction of an electron with the emitted field. As a matter of fact, since the electron is travelling at 99.99...

The copropagation of the electron with the field is what causes amplification or damping, based on the relative phase between the electron and the photon. If the phase difference is zero, electrons is feeding the field, but if we have a pi-shift, the field is feeding the electron.

Another way to see the process is to see the field as a potential, and electrons will minimize their energy by seating at the lowest point of this potential. This leads to pockets of electrons propagating together at low energy points of the field, which is known as micro-bunching. At this moment, the electrons and the field are almost in phase, leading to sizeable gain for the field.

The issue here is that each pulse is random in amplitude and in phase ! The reason for this is that microbunching is a stochastic feature that appear at a different region of the undulator for each pulse, leading to different gain each time. Typical values at swiss light are 80

A way to circumvene this issue, that is implemented in Trieste, is to send a UV pulse together with the electron pulse to stabilize the beam. However, also this method doesn't allow full reproducibility.

Therefore, performing pump probe experiment using a FEL is challenging, because instead of imposing a given delay between pump and probe, we have to record a wide variety of delays, and then retrieve for each measurement the corresponding

delay. This can be achieved for instance by recording a reference pulse on a camera after a grating and a SiN membrane. The role of the grating is to link the spatial coordinates to the time arrival of the pulse. Next, you send your FEL pulse through the membrane, modulating it's optical index on a very narrow region, leading to a deformed reference pulse on the camera. This tells you exactly the delay between FEL and reference.

Chapter 8

High Harmonic generation

We have now seen how to generate femtosecond pulses and how to manipulate them. We want to go one step further and explain attosecond pulses, as advertised in the course title. To this end, we will discuss the process known as High-harmonic generation. We will start by explaining the phenomenon using a semi-classical three-step-model. This will lead us to the generation of an attosecond train-pulse, and we will then adress the means to turn it into a single attosecond-pulse.

A The limits of perturbative non-linear optics

As a brief reminder, in [non-linear optics](Non-linear-effects), one can write the polarization as :

$$P = \epsilon_0(\chi^1 E + \chi^2 E^2 + \ldots)$$

This can lead to various non-linear processes. Among them, you have second harmonic generation, for which :

$$I_{2\omega} \propto I_\omega^2 \qquad \tau_{2\omega} = \frac{\tau\omega}{\sqrt{2}} \qquad \tau q\omega = \frac{\tau\omega}{\sqrt{q}}$$

This scaling laws imply that we can reduce the pulse duration by using higher-harmonics, at the cost of the energy. Therefore, we nee high energy in our pulses, as discussed previously on pulse amplifier.

However, instead of following this scaling law, the power spectrum rather show a plateau as we go to very high harmonic. Our aim is to explain why this happens and how we can describe this situation.

1 The Keldysh parameter

First we need a tool that helps us switching from one description to another one. To this end, let's come back to the physical meaning of polarization. The polarization can be seen classically as the deformation of the electronic cloud around an atom when submitted to a field. In the case of a low field, it's tempting to use a simple linear scaling $P = \epsilon E$. However, as we dramatically increase the energy of the field, we introduce non-linear effects by polarizing more and more. The natural physicist's tool to address the problem is a perturbative treatment. Now, if we increase again the intensity of the light, we will polarize the atom to a point where it will ionize. Therefore, we need a quantity comparing the work function (measuring ionization) vs ponderomotive energy bringing the electron into motion.

This leads to the Keldysh parameter $\gamma = \sqrt{W_i/2U_p}$, where W_i is the work function for ionization (a material property) and U_p the ponderomotive energy. A large Keldysh parameter ($\ll 1$) means that we can use the usual perturbative description, and a small one means we give enough energy to the electron to ionize the atom.

In this case, we need to calculate the ponderomotive energy. Starting from Newton's equation, we get :

$$m_e \frac{d\vec{v}}{dt} = eE_0 \cos(\omega t) \Rightarrow \vec{v} = \frac{eE_O}{m_e \omega} \sin \omega t$$

This leads to a kinetic energy $U_p = \frac{e^2 E_O^2}{4 m_e \omega^2}$.

If you think of the traditional energy profile binding an electron to an atom, you can now think of it as a deformed profile, lowering the energy for ionization.

B Three-step model of HHG

This semi-classical approach allows to describe in a neat way HHG.

1 Ionization

The ionization occurs via tunneling of the electron out of the atomic shells. The terms tunneling is not ill-defined, since after the event, the electron reaches a Volkov state, which differs from thermal hopping.

The characteristic tunneling time can be defined st:

$$t_{tu} = \frac{l_{barrier}}{v} = \frac{W_i}{eE_0} \sqrt{\frac{m_e}{2W_i}} = \sqrt{\frac{m_e W_i}{2}} \frac{1}{E_0} = \gamma T_0$$

This allows us to revisit the Keldysh parameter. It is the time needed to cross Coulomb barrier over the time over which the Barrier is lowered. If the tunneling time becomes too long, we don't ionize significantly, meaning a perturbative treatment still holds.

2 Acceleration in the field

We have ionized our atom, meaning we have a free electron in a field. In classical terms, it follows the following equation of motion (1D to stay simple) :

$$\ddot{x} = -\frac{e}{m_e} E_0 \cos(\omega t)$$

The shape of the field is given by the fact that we assume a plane wave. If we then assumes no kinetic energy at emission, and place the atom at the origin of axis, then $\dot{x(t_0)} = x(t_0) = 0$

$$x(t) = \frac{eE_0}{m_e \omega^2} \left[\cos(\omega t) - \cos(\omega t_0) + \omega \sin(\omega t_0)(t - t_0)\right]$$

3 Recombination

Your atom is accelerated in an oscillatory field, meaning it can eventually recombine with the ion. Its excess energy will then be consume to emit a photon. The question we have to adress is to know if an electron comes back to its original position, and with how much energy.

In essence, this problem amounts to solve $x(t > t_0) = 0$, and then find $\dot{x(t)}$ to calculate the kinetic energy. Since the previous equation of trajectory don't have explicit solution, we can solve graphically. We can then observe two behaviors, one where electrons come back to the atom, and one where they don't. PLOT !

Then, you may look at the energy of each electron when it goes back to 0. Interestingly enough, we observe a chirp, with a given emission time which is optimal. The emitted photon has therefore energy :

$$\hbar\omega(t) = W_i + 2U_p(\sin(\omega t) - \sin(\omega t_0))^2 \approx W_i + 3.17U_p$$

We can define that way the cut-off frequency of emission. This results tells us that to get higher harmonics, we have to move to high ponderomotive energies. This is why in practice rare gas are used for HHG cells.

Some limits : - We are not forced to have recombination of the electron with the parent atom

4 Attosecond pulse train

There is a last feature we didn't explain about HHG yet, namely the 2ω spacing in the HHG spectrum. This comes from the fact that an attosecond train is emitted at each extremum of the exciting laser pulse, meaning with a frequency 2ω. The result of such a system is an attosecond pulse train, because the exciting light pulse is likely to present several oscillations of light. The next difficulty is to select one pulse among this train.

C Toward single attosecond pulse

1 Motivation and scheme

As already discussed, the time-resolution involved in the study of a given phenomenon is ultimately fixed by the duration of a pulse. So far we have discuss the generation of a train of pulses. This artificially broaden our pulse, leading to blurring of the data.

To go to single attosecond pulses, the main idea is to use the largest attosecond pulse from the train, which contains all the highest harmonics, and especially the one of the cut-off. Therefore, we want to use high-pass spectral filtering tools to obtain such pulses.

This involve several technical challenges, as summarized on figure (FIGURE).

2 Nonlinear frequency broadening

The first step toward single-attosecond pulse is to generate a train containing only few pulses. To this end, we need a Fourier-limited optical pulse at the input of the HHG. This can be generated chirped technique[1].

However, the shorter the pulse, the broader the spectrum, the more dispersion we face. To give numbers, a Fourier limited 5fs pulse broadens to around 100fs after propagation in 1m of air. This means that the pulse has to be chirped so that the dispersion on the gas jet will be minimal.

One technical side-note here. For very broad spectrum, you can't find material reflecting identically over the whole spectrum with constant phase. This led to the developpment of chirped mirror, that are specially design to reflect pulses with an engineered phase. FIGURE !!

[1] cf notebook femto-up

3 Carrier-enveloppe phase

The next challeng we should adress for attosecond pulse generation is the control of the carrier-enveloppe phase ϕ_{ce}. This concept appears whenever we have a sinusoidal wave trapped within a slowly varying enveloppe. In essence, this is the phase between the maximum of the pulse enveloppe and the maximum of the wave lying in this enveloppe. SEE FIGURE.

As discussed above, the emission rate of electron in HHG (first step) scales exponentially with the field amplitude. If we don't control ϕ_{ce}, the amplitude of the maximal peak will fluctuate, leading to fluctuations in the spectrum of our attosecond pulse. This is a serious issue, since we want to use our attosecond pulse pump or probe a phenomenon. This requires stability.

The easiest way to measure this phase is to use a $f-2f$ interferometer, also called octave-spanning oscillator in analogy with acoustic waves. As a matter of fact, the output of HHG is a frequency comb, where all frequencies have an offset which is the carrier enveloppe phase. More precisely :

$$f_N = Nf_{rep} + f_{ce} \qquad f_{2N} = 2Nf_{rep} + f_{ce} \Rightarrow 2f_N - f_{2N} = f_{ce} \tag{8.1}$$

This technique implies that you have a sufficient number of harmonics in your frequency comb. Once you can measure this phase, you can correct it using a suitable feedback loop.

4 Single-pulse selection

As explained before, the highest frequencies of the high-harmonic spectrum arise solely from the highest pulse, if we have a reduced train. To get only one pulse, we just filter out the lower frequencies. This is achieved for instance using a zirconium mirror, but other techniques have actually been proposed.

At this stage, it is worth pointing out that the suitable optics for visible light are failing in the XUV range. As we saw, air is leading to broadening, meaning experiments have to be performed under high vacuum. Then, at such low wavelength, you start beeing sensitive to the atomic nature of matter, which drastically changes the way light is propagating in the media. Therefore, scientists are using elliptical or even toroidal mirrors at well-defined incidence angles to guide such light.

5 Brief conclusion

Let us conclude this part by summarizing what we learnt. In order to generate attosecond pulses, we rely on High-harmonic generation, a highly non-linear process. To efficiently control our attosecond pulse, the first requirement is to have the shortest possible visible pulse as an input. The second requirement is to controll the carrier-enveloppe phase of this input pulse, so that we always ionize our gaz the same way. Using proper filtering after the HHG, we can select a single pulse.

Of course, this claim is backed with proper experiment to measure the pulse duration. In this case, what has been used is the concept of attosecond streak camera, in which time is mapped over a direction of space by converting the light pulse into an electron pulse.

Chapter 9

Measuring short pulses

By now, we have discussed the main techniques involved in the generation of short pulses, whether in the ns-ps regime (Q-Switching) or in ps-fs regime (Mode-locking) or even in the as regime (High-harmonic generation). As hinted previously, we left a rather fundamental question behind us : How can we measure durations on such short timescales ? In this chapter, we want to adress this question by explaining the difficulties in classical time-measurement techniques, and then present the various measurement techniques used for pulse measurement.

A Why usual method don't work

B Autocorrelation techniques

1 Autocorrelator

The autocorrelation scheme is in essence an interference technique, as depicted on figure : FIG

The incoming pulse is split in two halves and we change the optical path of one line with respect to the other, before letting the two interfere in a non-linear crystal. In this crystal, we take advantage of second-harmonic generation, so that the output intensity is a convolution of the two pulses. This way, if the two signals don't overlap, we measure no intensity, and if the two signal overlap, we have a non-zero result.

By changing the delay, we are therefore able to measure the distance over which pulses overlap and give non zero contribution. In practice, we modify this delay by moving a piezo-electric crystal, since the required scanning distance is very small.

The main issue with this technique is that we have to make a guess about the shape of the incoming pulse that would generate the measured pattern. As a matter of fact, Different signals with different shapes can lead to the same autocorrelation trace. Therefore, this method is lacking precision.

2 FROG

This method is widely used down to the femto-second timescale. It reuses the main features of autocorrelation, the main difference beeing the detector. Instead of a photodiode measuring the intensity, we use a spectrometer to resolve the different frequencies of the pulse as a function of time, pretty much like the Gabor-transform discussed in tutorial.

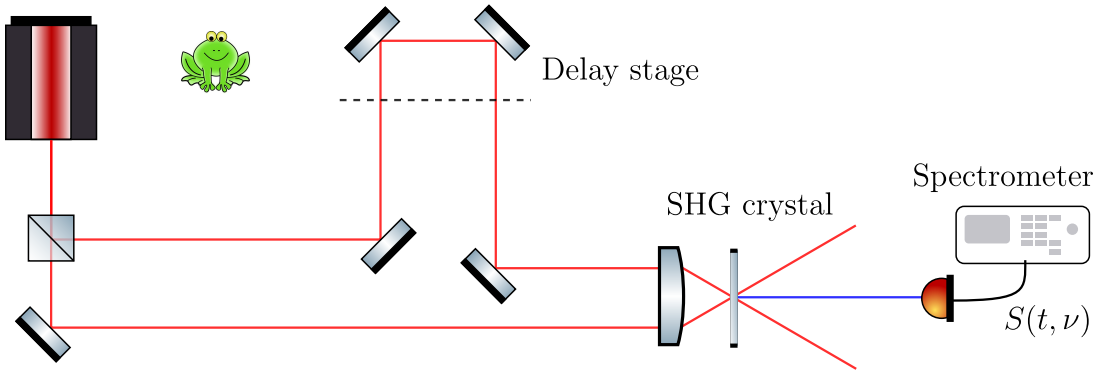

Figure 9.1: Optical set-up of the FROG. The autocorrelation is performed by sending the two pulses in the non-linear crystal, with a delay that is fixed by the delay stage. It is then recorded on the spectrometer. For autocorrelation, a simple photodiode is used, leading to less information

This allows to obtain richer information, and thanks to the proper FROG algorithm, it is possible to converge toward the real pulse. This algorithm is in essence based on gradient descent. For more discussion about FROG, see here REF PAPER

3 TADPOLE

When developped, the FROG hat pJ sensitivity thershold. However, the generation of pulses with high intensities was still a technical challenge, calling for methods sensitive to fJ pulse energy.

In this case, it is useful to use not only the emitted short pulse but also the source pulse, which usually has much higher intensity. This lead to the TADPOLE

method, standing for temporal analysis by dispersing a pair of light E-fields. It relies on spatial interferometry between the pulse and the source. Is requires very good knowledge of the source pulse.

4 GRENOUILLE

The GRENOUILLE scheme (Grating-eliminated no-nonsense observation of ultrafast incident laser light e-field[1]) is in essence another optical set-up that allows to record the spectrogramm of the pulse, and hence the pulse duration. (FIGURE).

Contrary to FROG, the delay between the two pulses is fixed using a Fresnel biprism, which is dividing the pulse in two. This way, the two subpulses will interact at different position in a thick non-linear crystal, mapping time over a spatial direction. Why do we need a thick crystal ? As you know, the momentum conservation condition imposes that the second-harmonic has a wavevector that isn't colinear to the incoming light, meaning over a large distance, we will disperse light according to its frequency. We therefore end up in a situation where time is mapped along one direction, and frequency on another direction. Using a CCD camera, we therefore measure directly the spectrogramm, in a single shot experiment.

C Reconstruction of attosecond beating by interference of two-photon transition RABBIT

The FROG method provides great result but is not not suited if we were to measure the duration of pulses in an attosecond pulse train[2] out of a gas jet. A first practical reason for this is that it is hard to find suitable optics for this range of frequencies. Another reason, more fundamental, lies in tha fact that we will correlate different pulses from a given pulse train, which prevents finding information about a single pulse.

Fortunately, another method is possible and relies on the fact that we have a pulse train with $2\omega_0$ spacing in frequency between the modes, ω_0 beeing the frequency of the exciting pulse. It is known as reconstruction of attosecond beating by interference of two-photon transition, hinting toward an interference effect.

As a matter of fact, the intensity of an APS can be written as

$$I(t) = \left| \sum_q A_q e^{i(\omega_q t + \phi_q(\omega_q))} \right| \tag{9.1}$$

[1]Sorry, I couldn't deny myself the pleasure

[2]See the chapter on HHG

where $\phi_q(\omega_q)$ is a phase that reflects the imperfect phase match of the modes in the APS. As you can guess, the information about the duration of a single pulse is contained in this phase. To keep the discussion simple, we consider the case of a linear chirp, which leads to a Fourier limited pulse : $\phi_q(\omega_q) = q\omega_0 t_e$, where t_e is the harmonic emission time. The shortest pulse duration is then given by the difference between the highest and lowest emission time[3].

Still, how should we measure the phase of the pulse ? The idea is to use a second gas jet on which the attosecond pulse train and the exciting pulse are focused. The APS will therefore develop sidebands at $\pm\omega_0$ due to it's interaction with the IR pulse, leading to interference effect between it's different Harmonics. Therefore, If you place a spectrometer after this gas jet, you will observe a large 2ω spacing as well as a ω modulation of the intensity, which depends on the delay between IR and APS.

Finally, we stress that this method is in fact giving much more information about the pulse than the sole duration. It can be extented to study other characteristics of an attosecond pulse train, and also to study atomic physics. Think of it, you now have a mean to study the answer of matter submitted to a field on the as timescale !

D Streak camera

The last technique we will discuss is the so-called streak camera, which consists in mapping time over a direction of space.

1 Streak camera

The operating principle of a streak camera is displayed on figure **??**. The idea is to send the laser pulse onto a target to extract electrons. Owing to energy/momentum conservation, this electron will travel at a given speed, thus reproducing the temporal profile of the pulse. We then submit our electron to a linearly increasing electric field, that will deflect electrons more and more as time passes. We record this trace on a phosphorous screen, and that's it. The longer the pulse, the broader the electron trace.

[3]For a more detailed discussion, see Mairesse et al

2 Attosecond streak camera

When moving down to the attosecond scale, an alternative technique can be proposed. At such low timescale, it is hard to have a field that is varying enough to sort electrons out, without mentionning the other technical challlenges. Therefore, the idea is to use the electric field of a light pulse instead of the field of a capacitance. Let's give more details.

Once more, the XUV pulse is send on a gas jet, leading to ionization, hence emission of electrons. Then, this electrons are accelerated by the much higher IR field, leading to a dispersion in energy based on the value of the IR field. We will show in the tutoral that the overall momentum distribution reads :

$$\Delta p = \sqrt{\Delta p_0^2 + \left(\frac{dp}{dt}\tau_x\right)^2} \tag{9.2}$$

Therefore, the recorded pattern contains information on both the IR and the XUV pulse. Once more, this is a way to study atomic physics. Especially, it has been demonstrated that depending on the atomic orbital from which the electron comes from has an effect on the momentum dispersion. At such small timescales, it is not possible to treat the emission as a simple scattering event and one typically has to treat this as a coherent process.

Chapter 10

Toward pulses of electrons

By now, you should have a satisfying view of the field of laser physics. We have seen how to generate pulses, how to guide them, how to manipulate them, and how to measure them. We have pushed the description down to the attosecond level. To give a broader perspective, and also because it is core to our research at LUMES, we now want to tackle the physics of electron pulses. We will basically cover the same steps as before, namely generation, optics, and measurements, in a condensed fashion.

A Introduction

In this course, we are mostly interested in electrons for microscopy purposes. So let's review very briefly the pros and cons of electrons over light.

First criterion is resolution. If you accelerate your electrons to some 100keV, the electron wavelength can be calculated using De Broglie's relation. It appears to be on the picometer scale, meaning that we can reach much smaller resolution with electrons than with traditional visible or even UV light. As a matter of fact, state of the art electron microscopes don't reach the diffraction limit and are still limited by aberrations. Resolution is at best around 10pm, still sufficient to observe individual atoms.

The next feature of electrons is that they developp lots of different interactions with a sample, such as diffraction with the lattice, scattering by individual atoms, magnetic contrast, inelastic scattering and so on. Provided that you have the right set of detectors, lenses and aperture, you can then probe very different features of a gievn sample with a unique system. In addition, this interactions have a high chance of occuring, leading to a a contrast which is significantly higher than for X-rays.

There are still difficulties when electrons are used. The most obvious one is

that electrons repel each other due to the Coulomb force. Therefore, a given pulse of electrons will be broaden in time when propagating. Another difficulty is that electron may damage the sample, which is refered to as irradiation damage. This is particularly at stake when trying to image biomolecule, that are very fragile.

B Electrons sources

The first question we will adress is the one of electron generation. FIGURES !! We first note that this sources are all chaotic, leading to lower coherence than laser light. The spatial coherence of the pulse will be generally given by the size of the source and increase the further away we move from this source FIGURE. For higher coherence, we need smaller sources. Temporal coherence on the other hand is scaling as the inverse of the energy spread of the electrons. It is therefore a parameter we can tune.

Thermionic sources The idea is to run a current in a cathode to increase the temperature. This allows the emission of hot electrons, which have enough energy to overcome the work function W. In such systems, typically LaB_6 or Ta tips, the temporal coherence scales as the inverse of the temperature.

Field emission gun (FEG) Here, you place a tip in a strong electric field to extract electrons. To gain spatial coherence, a single-atom tip is used, coming at no cost on brightness.

Cold FEG Same as before, except you cool your tip down to 20K to enhance your coherence time by reducing the temperature.

Photoemission gun This technique is particularly suited to produce electron pulses. By sending light with energy $\hbar\omega > W$, you can extract electrons via the photoemission process. If you have a light pulse, you will end up with an electron pulse. For this source, the coherence time is evolving as $T = 1/(\hbar\omega - W)$. It is worth noticing that in general, W is slightly evolving in time due to contamination and tip deterioration, so there is no point in engineering the light to achieve perfect coherence. Besides, you would still be limited by coherence at some point. You also pay spatial coherence because the light impinging the cathode is usually on the micron scale, much bigger than an atom.

BEC source This is for the moment an experimental proposal[1]. The idea is to trap atoms in an optical lattice and cool them with light. The temperature reached

[1]PAPER

by this atoms is below 1K, allowing very high coherence. By pumping these atoms with light, it is possible to extract electrons.

RF guns This technique relies on a field emission gun cascaded with a RF-cavity. This cavity is imposing an electric field to chop the beam, allowing operation in either continuous or pulsed mode, with the coherence properties of a FEG. You can also play easily with the repetition rate of your pulses by playing with the cavity, up to some GHz. The main drawback is the pulse duration, which is rather long because it is limited by electronics.

C Electron optics

1 Spatial lenses

To guide our electrons and form images of our sample, we have to design proper lenses. An exhaustive description of electromagnetic lens is to be found at: REFF !

The idea is to use a coil surrounded by ferromagnets to focuse electrons.

2 Temporal lenses

This term refers to the fact that an electron pulse we tend to get broader in time due to Coulomb force. The idea of the temporal lense is to have a device that will brake the front electrons of the pulse and accelerate the back electrons so that the pulse will have minimal duration when interacting with the sample. FIGURE.

To fix ideas, we can give some typical numbers. At 300keV, a 100fs pulse of electrons containing 1000 charges is extending up to 10 ps after 1m of propagation, and a TEM is typically longer than 1m. This is a tremendous loss of temporal resolution. Experimental realization of temporal lenses are able to recompress the pulse, but they remain experimentally challenging. You need high cooling to have mK precision on the temperature, and fs precision to launch the electric field.

D Interaction with the sample

When electrons are reaching the sample, various interactions can take place, such as scattering by single atoms, diffraction by the lattice, and so on. An extenive discussion is beyond the scope of this course[2], and we will focuse here on direct

[2] ADD REF

interaction between electrons and light.

1 PINEM

2 Transition radiation

Part II

Tutorials / Practicals

Part III

Bibliography

This should be better organized. Just a few hint to go further.
Svelto, principles of lasers
Keller, Ultrafast lasers
Zenghu, attosecond pulses
Nonlinear optics, Boyd
Fundamentals of photonics, Saleh
Quantum theory of light, Loudon
Fiber optics, Aggrawal
Carter and Williams, TEM
Not to mention papers and things.