Ultrafast Pulses

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Abstract

Ultrafast phenomena are processes that occur with durations in the range of nano- to femtoseconds. This essay discusses the general principles of laser beam generation, focusing on ultrafast pulse generation by mode-locking. Mode-locking of pulses is simulated and the results are shown. The use of autocorrelation as a means of determining the duration of ultrafast pulses is also discussed and other techniques are highlighted as well. Using experimental data, characteristics of an ultrafast pulse such as its pulse repetition rate, bandwidth and duration are also determined.

Contents

1. General Introduction

'And God said, "Let there be light"' is a text often quoted from the Christian scriptures. The phenomenon that is light is one that has both fascinated and puzzled man for centuries. Light, as we know it, has been harnessed for various applications—from basic illumination to more sophisticated fibre optic communications. Light is just one form of electromagnetic radiation and as such, is a form of energy. Hence, from a quantum mechanical perspective, light could be thought of to consist of discrete packets called photons. From quantum mechanics, it is known that matter consists of different discrete energy levels and, in order to move from a lower energy level, E_n to a higher one, E_m, requires an input of energy (usually a photon) which is governed by the equation

$$E_m - E_n = (\Delta E)_{m,n} = h\nu, \tag{1.1}$$

where

$$h \approx 6.625 \times 10^{-34} \quad \text{Js}$$

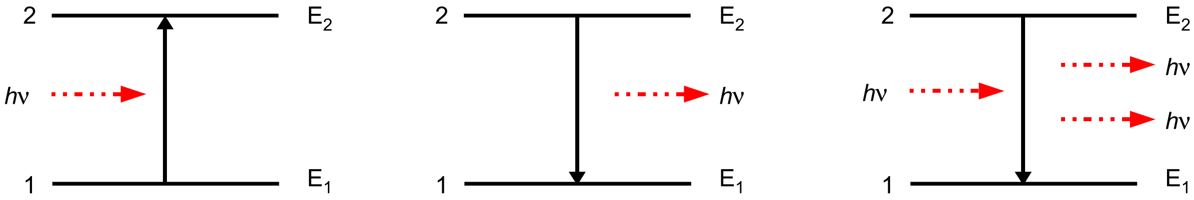
is Planck's constant, ν is the frequency of the radiation incident upon the material and m, n are the transition states [1]. In essence, the material *absorbs* some energy in order to make this transition. On the other hand, a material can 'lose' energy in order to drop from a higher to a lower energy level. In this case, the material *emits* or *radiates* energy as it makes this transition. This energy can be emitted in the form of light [2]. It is this property of materials that enables lasers to be generated.

1.1 Absorption, Spontaneous and Stimulated Emission

In nature, materials usually exist in the transition state of lowest energy (usually referred to as their ground state[1]). **Absorption** occurs when an atom receives an input of energy that causes it to transit from this lower transition level to a higher one. The frequency of the exciting radiation is governed by equation (1.1). However, some atoms or molecules exist in an excited state and, without the external excitation, will drop to a state of lower energy, releasing photons in the process. This natural emission of photons is referred to as **spontaneous emission** and is a non-radioactive decay of a material [3]. Spontaneous emission is the source of most light around us, e.g. from the sun, stars, light bulbs and fluorescent light. Einstein proposed that a second type of emission could be induced [2]. Instead of undergoing a natural decay, an excited atom could be *stimulated* to release its energy as a photon of light. This is what is referred to as **stimulated emission**. In this case, an atom emits light due to some external stimulation by a photon. This is the key concept that drives the generation of lasers. The figure below illustrates the differences between these two methods of emission.

In fig (1.1(a)), the atom receives an input of energy causing it to move from level 1 to level 2. The probability that the absorption process occurs is determined by the *absorption rate* which is

[1]The term 'ground state' used here does not refer to the state of the system at absolute zero but at thermal equilibrium.



(a) Illustration of absorption.

(b) Illustration of spontaneous emission.

(c) Illustration of stimulated emission.

Figure 1.1: Diagram illustrating the processes of absorption, spontaneous emission and stimulated emission

proportional to the population of atoms in the energy level 1 [3]. Thus, we can write

$$\dot{N}_1 = -W_{12} N_1 \quad , \tag{1.2}$$

where $\dot{N}_i = dN_i/dt$, N_i is the number of atoms in level i and W_{12} is a constant defined by [3]

$$W_{12} = \sigma_{12} I_\nu \quad . \tag{1.3}$$

σ_{12} is referred to as the absorption cross section (and depends on the particular transition) and I_ν is the intensity of the incident wave (energy per time per m^2). In the case of spontaneous emission, (fig 1.1(b)), as the atom drops from level 2 to level 1, a photon is emitted and the rate at which this occurs is governed by $\dot{N}_2 = -A_{21} N_2$; where A_{21} is the Einstein coefficient for the transition. On the other hand, for stimulated emission (fig 1.1(c)), the energy from the 'stimulating' photon causes the release of another photon, which is in phase with it, as the atom drops from level 2 to level 1. As in the case of absorption, the rate of stimulated emission is proportional to the population of the atoms in the higher energy level 2 and we can write

$$\dot{N}_2 = -W_{21} N_2 \quad , \tag{1.4}$$

and the coefficient W_{21} is [3]

$$W_{21} = \sigma_{21} I_\nu \quad . \tag{1.5}$$

σ_{21} is called the stimulated emission cross section and is also dependent on the characteristics of the transition.

1.1.1 Gain

For a material with two energy levels 1 and 2 let the populations per unit volume of these levels be N_1 and N_2 respectively. Consider a plane wave of intensity $I_\nu(z)$ travelling through the material as shown in figure (1.2) below. This wave undergoes an elemental change in its intensity $dI_\nu(z)$ as it travels through the elemental length dz of the material due to the absorption and stimulated emission processes that it induces in the shaded region. If A is the cross sectional area of the wave, then the change in the number of photons being absorbed and emitted is $AdI_\nu(z)$. This must be equal to the difference between the population of photons being absorbed and emitted in the shaded volume $dV = Adz$ per unit time i.e.

$$\begin{aligned} AdI_\nu(z) &= (\dot{N}_1 - \dot{N}_2)Adz \\ &= (W_{21}N_2 - W_{12}N_1)Adz \quad . \end{aligned} \tag{1.6}$$

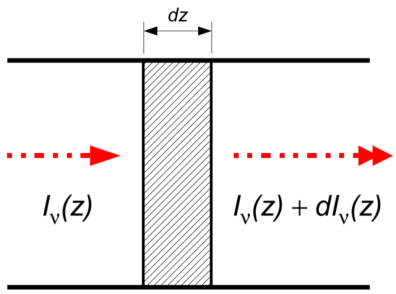


Figure 1.2: A photon of intensity $I_\nu(z)$ undergoing a change $dI_\nu(z)$ as it travels through a distance dz in a material.

Using equations (1.3) and (1.5), we obtain

$$\begin{aligned} dI_\nu(z) = (\sigma_{21}N_2 - \sigma_{12}N_1)I_\nu(z)dz &= \sigma_{21}\left[N_2 - \frac{\sigma_{12}}{\sigma_{21}}N_1\right]I_\nu(z)dz \\ \Rightarrow \quad dI_\nu(z) &= \sigma_{21}\left[N_2 - \frac{g_2}{g_1}N_1\right]I_\nu(z)dz \quad , \end{aligned} \tag{1.7}$$

where g_1, g_2 are the degeneracies of the energy levels and are related to the respective cross sections by the Einstein relation [3]

$$g_2\sigma_{21} = g_1\sigma_{12} \quad .$$

Solving equation (1.7), with the boundary condition that at $z = 0, I_\nu(z) = I_0$, and assuming a constant rate of change of the population levels $\dot{N}_i$ yields

$$I_\nu(z) = I_0 e^{g_\nu z} \quad , \tag{1.8}$$

where

$$g_\nu = \sigma_{21}\left[N_2 - \frac{g_2}{g_1}N_1\right]$$

is referred to as the *gain coefficient*. In order to have sustained stimulated emission, there should be more atoms occupying higher transition levels than lower ones i.e. $g_\nu > 0$ and hence,

$$N_2 - \frac{g_2}{g_1}N_1 > 0 \quad .$$

This condition is what is referred to as a *population inversion* and is a necessary condition for laser beams to be produced [1]. Using equation (1.8), the **gain**, G, of a laser which is a measure of the amplification of the input signal is

$$G = \frac{\text{Output}}{\text{Input}} = \frac{I_\nu(z)}{I_0} = e^{g_\nu z} \quad . \tag{1.9}$$

Thus, there is an exponential growth in the intensity of the laser beam produced provided that $g_\nu > 0$. In a situation where $g_\nu < 0$, there is an exponential decay in the intensity of the beam produced i.e. it is absorbed.

1.2 Lasers

What then is a laser? The mention of the word 'laser' brings to mind images of science-fiction movies where individuals employ lasers for anything from cutting through metals to vapourising enemies! Real lasers, however, are not always that dramatic. The word *laser* is actually an acronym which stands for **l**ight **a**mplification by the **s**timulated **e**mission of **r**adiation [2]. The laser employs processes that amplify light signals that have been generated by other means. A laser takes light that would be emitted in several directions and concentrates them into a single beam [4].

In the early 20th century, most scientists were not enthusiastic about having sustained amplification by stimulated emission of radiation since, at thermal equilibrium, more atoms occupy lower energy levels than higher ones. Thus, sustained emissions would require a 'population inversion' where there would be more atoms in higher energy states than in lower ones. The first to succeed in creating the required levels of population inversion for amplification of radiation by stimulated emission, was Charles H. Townes in the 1950s [2]. He did not work with light but with microwaves and he built a device called the **maser** which is a similar acronym to the laser but the word 'light' is replaced with 'microwave.' It was not until May 16, 1960 that Theodore H. Maiman built the first laser which was a ruby laser [1]. This served as the launch pad for the development of even more sophisticated laser systems.

A laser consists of three main components [5]:

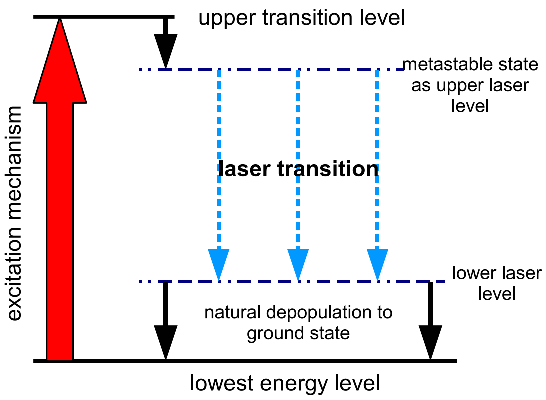
- an active medium which amplifies the incident electromagnetic wave by producing the required population inversion,
- an energy pump which causes excitation of the active medium, resulting in the filling up of the required energy levels which leads to population inversion and
- an optical resonator/cavity that stores the portion of the stimulated emission that is concentrated within a number of *resonator modes*[2].

The active medium is a material that is highly susceptible to excitation when stimulated by an external field and produces an amplification in the intensity of the field. The energy pump which could be a flashlight, gas discharge or another laser, causes the atoms in the lowest energy level to absorb the required energy to move to a high probability upper transition level. When this occurs, the atoms in the upper level then drop to a highly stable state referred to as a 'metastable state'. This metastable state serves as the upper laser level. In order for stimulated emission to

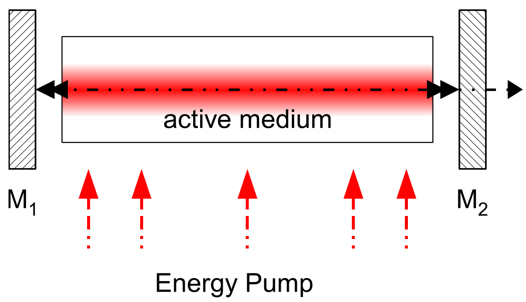
[2] See section (2.1)

dominate, there have to be more atoms in this metastable state than in the lower energy level (for population inversion to continue).

If the lower laser level is also the lowest energy level of the atom, then more energy is required to excite enough atoms to move to the higher transition level [2]. This is what is referred to as the 'three level laser.' However, most laser systems are 'four level' i.e. the lower laser level is an energy transition state which is *higher* than the lowest energy level but *lower* than the metastable state. Thus, the atoms in the lower laser level eventually lose their energy and spontaneously drop to the lowest energy state, allowing the required population inversion to be maintained and emission to continue (see fig. 1.3(a)).



(a) A four level laser scheme.



(b) Schematic set up of a laser.

Figure 1.3: Fig. 1.3(a) shows how the principle of population inversion results in stimulated emission while fig. 1.3(b) shows the schematic set up of a laser. M_1 and M_2 are the coupling mirrors within the resonator.

As shown in fig (1.3(b)) the resonator can consist, for example, of two coupling mirrors, M_1 and M_2. In this case, M_1 is a totally reflective mirror which allows photons to be reflected back into the medium. On the other hand, M_2 is not totally reflective but allows a part of the photons to be transmitted through it (which constitute the output laser beam) while reflecting back a significant proportion of the photons. This results in more photons causing excitation within the medium leading to more stimulated emission (in essence, amplification) until the photons within the resonator reach a threshold at which this feedback[3] (which is positive) converts the laser amplifier into a laser oscillator and the power levels inside and outside the laser are constant [2, 5].

[3]Feedback occurs when all or part of the output signal of a system is coupled back into the system. If this 'feedback' signal is in phase with the input signal, it is regarded as positive and negative if otherwise. When feedback is negative, it results in a reduced gain in the output signal. However, when positive, it leads to a greater amplification of the output signal. This continuous amplification leads to oscillation within amplifiers [6].

Threshold Gain

As described above, to produce a laser beam, a population inversion must be produced in the active medium and the gain within the resonator must be greater than or equal to the losses within it. Consider the laser setup shown in fig (1.2) with two beams of intensities $I_\nu^+(v)$ and $I_\nu^-(v)$ travelling between two mirrors M_1 and M_2.

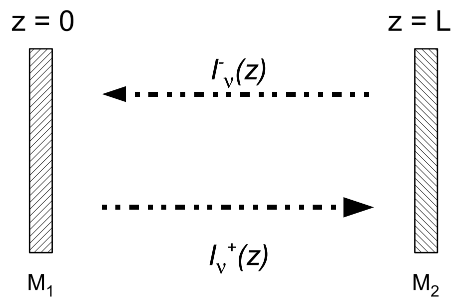


Figure 1.4: Two beams propagating in different directions within a laser resonator

At the mirror $M_1,\ z = 0$ and the intensity of the beam is [1]

$$I_\nu^+(0) = r_1 I_\nu^-(0) \quad , \tag{1.10}$$

where r_1 is the reflectivity of the mirror. Similarly, at $M_2,\ z = L$ and hence,

$$I_\nu^-(L) = r_2 I_\nu^+(L) \quad , \tag{1.11}$$

where r_2 is the reflectivity of the mirror M_2. As the laser oscillation approaches the threshold, the intercavity intensity is small [1] and thus for the beam propagating in the positive z-direction, using equation (1.7),

$$\begin{aligned} \frac{dI_\nu^+(z)}{dz} &= g_\nu I_\nu^+(z) \\ \Rightarrow \quad \ln[I_\nu^+(z)] &= \int_0^L g_\nu dz \\ \therefore \quad I_\nu^+(L) &= I_\nu^+(0) e^{g_\nu L} \quad . \end{aligned} \tag{1.12}$$

Similarly, for the beam travelling in the negative z-direction,

$$\begin{aligned} \frac{dI_\nu^-(z)}{dz} &= -g_\nu I_\nu^-(z) \\ \Rightarrow \quad I_\nu^-(0) &= I_\nu^-(L) e^{g_\nu L} \quad . \end{aligned} \tag{1.13}$$

Using equation (1.11) and the expressions in equations (1.12) and (1.13),

$$\begin{aligned} I_\nu^-(L) = r_2 I_\nu^+(L) &= r_2[I_\nu^+(0)e^{g_\nu L}] \\ &= r_1 r_2[I_\nu^-(0)e^{g_\nu L}] \\ \therefore \quad I_\nu^-(L) &= r_1 r_2 e^{2g_\nu L} I_\nu^-(L) \quad . \end{aligned} \tag{1.14}$$

If $I_\nu^-(L)$ is non-zero, then at steady state, $g_\nu = g_t$ and

$$\begin{aligned} r_1 r_2 e^{2g_t L} &= 1 \\ \Rightarrow \quad g_t &= \frac{1}{2L}\ln\left(\frac{1}{r_1 r_2}\right) \quad . \end{aligned} \tag{1.15}$$

The value of the gain g_t that satisfies equation (1.15) is called the threshold gain of the laser and is the minimum gain required for a laser beam to be emitted within the laser [1, 3]. By setting $r_1 r_2 = 1 - x$ and using the Taylor expansion for $\ln(1-x)$, a useful approximation is obtained when $r_1 r_2 \ll 1$ i.e. equation (1.15) becomes

$$g_t \approx \frac{1}{2L}(1 - r_1 r_2) \quad . \tag{1.16}$$

Taking into account not only losses at the mirrors but the distributed loss per unit length, a, within the laser cavity itself, the general expression for the threshold gain is [1]

$$g_t \approx \frac{1}{2L}(1 - r_1 r_2) + a \quad . \tag{1.17}$$

Uses of Lasers

Lasers are used for a variety of applications. They are applied in different fields from telecommunications to, not surprisingly, military technology. Below is a sampling of the various uses of lasers [2]:

- **Information Technology**: fibre optic communications, laser printing, playing various types of digital media (e.g. compact discs and DVDs) and bar code readers.
- **Medicine**: various forms of laser surgery, treatment of kidney stones, tattoo removal and other forms of cosmetic surgery.
- **Military Technology**: Antipersonnel weapons, ‘war games’ and battle simulation, and target locking for bombs and missiles.
- **Other Uses**: holography, isotope separation, illuminating cells for biomedical measurements, measuring chemical concentrations and, various distance and velocity measurements.

1.3 Characteristics of Laser Beams

Laser beams generally have unique properties such as much narrower frequency distributions and higher intensities than other more common light sources like candles, lamps and flashlamps. There are different types of lasers for a number of applications. Though the underlying principle of generating them remains the same, there exists some differences in their actual operation. However, there are some properties which are common to all lasers and are important in describing their behaviour and are crucial for their generation. We will discuss them briefly.

Coherence

Light waves are said to be coherent when they are in phase with one another i.e. the 'crests' and 'troughs' of the wave line up at the same points. Consider two waves of angular frequencies ω_1, ω_2 with wave numbers, k_1 and k_2 travelling in the z-direction; then their electric field amplitudes will be given by

$$E_1 = E_{1_0} e^{-i[k_1 z - \omega_1 t]} \quad \text{and} \quad E_2 = E_{2_0} e^{-i[k_2 z - \omega_2 t]}, \tag{1.18}$$

with E_{1_0} and E_{2_0} representing the peak values of the amplitudes. For these two waves located closely in space, the combined wave E, will be given by

$$\begin{aligned} E &= E_{1_0} e^{-i[k_1 z - \omega_1 t]} + E_{2_0} e^{-i[k_2 z - \omega_2 t]} \\ \Rightarrow \quad E &= E_{1_0} e^{-i\phi_1} + E_{2_0} e^{-i\phi_2} \quad , \end{aligned}$$

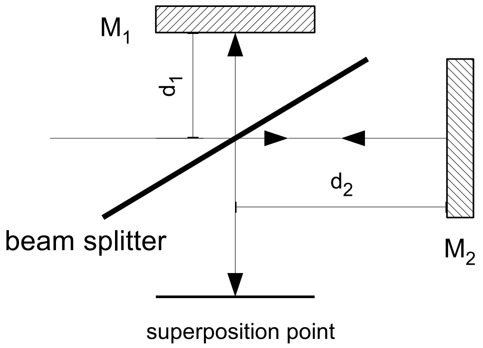
with $\phi_1 = k_1 z - \omega_1 t$ and $\phi_2 = k_2 z - \omega_2 t$. The intensity of the wave will be

$$\begin{aligned} I \sim |E^2| &= (E_{1_0})^2 + (E_{2_0})^2 + E_{1_0} E_{2_0} (e^{-i[\phi_1 - \phi_2]} + e^{i[\phi_1 - \phi_2]}) \\ \Rightarrow \quad I &= I_1 + I_2 + 2E_{1_0} E_{2_0} \cos \Phi, \end{aligned} \tag{1.19}$$

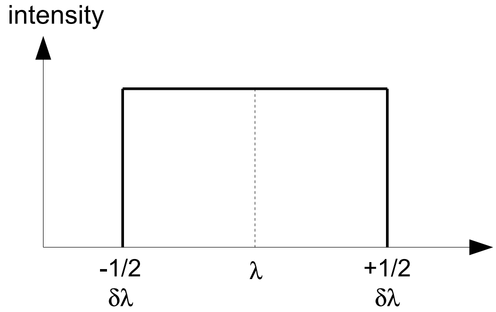
where $\Phi = \phi_1 - \phi_2$. Examining equation (1.19), we notice that when $\Phi \approx 0$ (i.e. both waves are in phase), the intensity of the new wave is equal to the sum of the intensities plus twice the product of the electric fields of the individual waves forming it. For totally incoherent waves, the total intensity would be

$$I = I_1 + I_2 \quad .$$

We could either have *spatial* or *temporal* coherence. **Spatial** coherence measures the area over which light waves are coherent while **temporal** coherence measures the duration over which the waves are coherent [2]. Thus, in generating lasers, the photons excited must be in phase with each other in order to achieve amplification. If they are not, there will be destructive interference within the laser cavity leading to 'cancelling' out of photon energies and thus, no laser beam will be generated for a long duration. However, 'perfect' coherence does not exist and so, for lasers, we measure their *coherence length* which determines the distance over which the laser beam is



(a) Setup of a Michelson interferometer. M_1 is a fixed mirror while M_2 is an adjustable mirror.



(b) Intensity distribution as a function of wavelength.

Figure 1.5: A Michelson Interferometer and an illustration of the intensity distribution of the beam within the interferometer as a function of wavelength.

coherent. To explain this, consider a laser beam of spectral width $\delta\lambda$ incident upon a Michelson interferometer. This leads to the formation of bright and dark 'spots' as the distance $|d_1 - d_2|$ is varied (see figure (1.5(a)) below).

Assuming that the intensity is constant for wavelengths in the range $\lambda \pm \frac{1}{2}\delta\lambda$, let the maximum intensity occur at [1]

$$|d_1 - d_2| = n(\lambda + \frac{1}{2}\delta\lambda) \quad . \tag{1.20}$$

And let minimum intensity occur at

$$|d_1 - d_2| = (n + \frac{1}{2})(\lambda - \frac{1}{2}\delta\lambda) \quad . \tag{1.21}$$

Subtracting equation (1.20) from (1.21) yields

$$\begin{aligned} |d_1 - d_2| \left(\frac{1}{\lambda - \frac{1}{2}\delta\lambda} - \frac{1}{\lambda + \frac{1}{2}\delta\lambda} \right) &= \frac{1}{2} \\ |d_1 - d_2|\delta\lambda &= \frac{1}{2}\left[\lambda^2 - \left(\frac{1}{2}\delta\lambda\right)^2\right] \\ \Rightarrow \quad |d_1 - d_2| &\approx \frac{\lambda^2}{2\delta\lambda} \end{aligned} \tag{1.22}$$

if $\delta\lambda \ll \lambda$. The quantity $|d_1 - d_2| = c\tau$ is the *coherence length* of the beam where c is the speed of light and τ is the time taken for the beam to traverse the distance $|d_1 - d_2|$. Using the relationship $\lambda = c/\nu$ where ν is the frequency of the beam, then equation (1.22) can be expressed as

$$\begin{aligned} |d_1 - d_2| &= \frac{c}{2|\delta\nu|} \\ \Rightarrow |d_1 - d_2| &= \frac{c}{\Delta\nu} \end{aligned} \tag{1.23}$$

where $\Delta\nu = 2\delta\nu$ is the frequency bandwidth of the laser.

Monochromaticity

Monochromaticity implies that the laser beam generated is of a single wavelength. However, lasers are not fully monochromatic but have a range of wavelengths $\Delta\lambda$ that are transmitted [1]. This is especially true for broadband lasers with a large frequency bandwidth which implies a narrow range of wavelengths over which the laser beam propagates [2].

Output Power and Brightness

The output power of a laser is defined as the energy radiated by the laser per unit time. It is given by [2]

$$P = \frac{\Delta E}{\Delta t} \quad , \tag{1.24}$$

where ΔE is the energy of the beam over the duration Δt. On the other hand, the brightness of a laser measures the power emitted per unit area of incidence per unit solid angle. It is given by the expression [3]

$$B = \frac{4P}{(\pi D\theta)^2} \quad ,$$

where P is the power of the laser beam, D is the diameter of the cross section of the laser and θ is the beam divergence.

Directionality

The directionality of a laser beam is a measure of its beam divergence. By *divergence*, we refer to how the laser beam 'spreads out' with increasing distance from the source from which it is emitted; it is measured at half the angle at which the beam spreads and is given by [3]

$$\theta_d = \frac{\beta\lambda}{D} \quad ,$$

where β is a numerical coefficient of the order of unity which depends on the variation of intensity across the beam, λ is the beam wavelength and D its diameter at waist. The distance over which the light rays of the laser beam remain parallel is called the *Rayleigh range,* z_R and is given by [2]

$$z_R = \frac{D^2}{\lambda}$$

with D and λ being the diameter at waist and wavelength of the beam respectively.

2. Generation of Laser Pulses

Lasers can emit light either in a **continuous wave** mode or in a series of **pulses** depending on the physics and design of the laser. Lasers can have continuous or *steady state* emissions when the set of energy levels and transition states of the active medium allow for continuous operation. However, a large number of lasers emit light in pulses; short 'bursts' of light (ranging from milli- to femtoseconds) with various repetition rates and high peak intensities. To the naked eye, these might appear to be continuous due to the short duration between bursts, but this is not the case. We will be discussing in detail the generation of these ultrashort pulses but suffice it to say at the moment that there are three primary ways of generating these pulses: Q switching, cavity dumping and mode-locking [2].

It is important to note that population inversion is not the only criterion necessary to achieve the generation of laser beams. In order to have a laser beam, the light produced through stimulated emission has to be *concentrated* in the desired direction in order to be of any meaningful use. Extracting the energy from the active medium and generating a laser beam requires a resonant cavity which amplifies the stimulated emission by positive feedback [2].

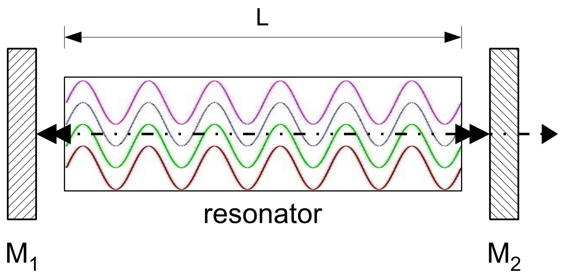
The idea of positive feedback is important. From basic amplifier design, it is known that the build up of positive feedback leads to oscillation in an amplifier i.e. the generation of a continuous wave signal within the amplifier. This is critical in the design of laser systems because most laser materials have very low gain and hence the light would have to travel a long distance to produce much amplification. In order to achieve the gain desired within the resonator, the photons travel back and forth within the laser which increases the distance over which they travel thus leading to a greater amplification and eventually, oscillation.

In order for this oscillation to take place, there must be *resonance* in the laser activity [2] i.e. the frequency of the photons *driving* the oscillation must equal that of the *driven* photons. This is actually a consequence of the coherence property of laser beams as discussed in section 1.3. The boundary conditions of the resonator require that the electric field be zero at the mirrors [2]. Thus, standing waves are formed within the laser cavity and these satisfy the relation [2]

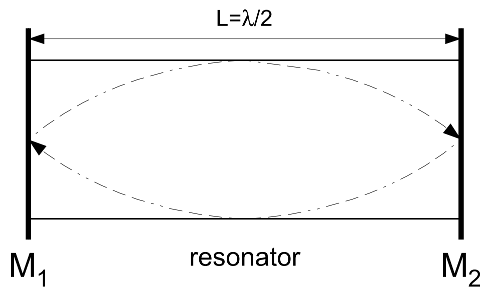
$$\begin{aligned} \frac{N\lambda_N}{2\eta} &= L \\ \Rightarrow N\lambda_N &= 2\eta L \quad . \end{aligned} \tag{2.1}$$

N is an integer number, λ_N the wavelength of the laser beam, η the refractive index of the active medium that constitutes the cavity and L is the length of the cavity.

This criterion for resonance causes the number of wavelengths that can resonate in the cavity to be spread over a range of wavelengths referred to as the **gain bandwidth** of the laser; and a number of different wavelengths can fit within this bandwidth as long as they satisfy equation (2.1). Each resonant value N is referred to as a **longitudinal mode** of the laser [2].



(a) A schematic diagram of a laser resonator.



(b) Example of a standing wave pattern within laser resonator.

Figure 2.1: A schematic diagram of a laser resonator and a corresponding example of the standing wave pattern within the resonator. Notice that the waves 'built' up within the resonator are all coherent. M_1, M_2 are mirrors.

2.1 Laser Modes

From equation (2.1) we can write the resonance criterion in terms of the frequency ν of the laser beam using the relation, $c = \lambda\nu$ as

$$\nu_N = \frac{Nc}{2\eta L} \quad , \tag{2.2}$$

where c is the speed of light in a vacuum. Thus, the frequencies that satisfy this criterion are referred to as the **modes** of the laser beam. For two adjacent modes, ν_N and ν_{N+1} we derive the frequency separation as follows:

$$\begin{aligned} \Delta\nu &= \nu_{N+1} - \nu_N \\ &= \frac{(N+1)c}{2\eta L} - \frac{Nc}{2\eta L} \\ \Rightarrow \quad \Delta\nu &= \frac{c}{2\eta L} \quad , \end{aligned} \tag{2.3}$$

where $\Delta\nu$ is referred to as the **frequency difference** between two consecutive modes [3]. It is observed that the frequency spacing is independent of the frequency ν of the laser beam but depends on the refractive index, η of the active medium and the length, L of the resonator. A mode is usually specified by three numbers, l, m, n. This is because the wave varies in two different ways viz: transverse modes and longitudinal modes.

Transverse Modes

Transverse modes of a laser determine the pattern of the distribution of intensity across the width of a laser beam and are usually referred to as **TEM**$_{l,m}$ modes, which stands for Transverse Electric and Magnetic modes. The indices l, m refer to the direction of the wave in the electric and magnetic field directions respectively. The modes are called transverse because they oscillate in directions mutually perpendicular to the direction of propagation of the wave. There are a large number of different transverse modes as specified by the indices l, m [2, 3]. Shown below is the intensity profile for a number of different TEM modes.

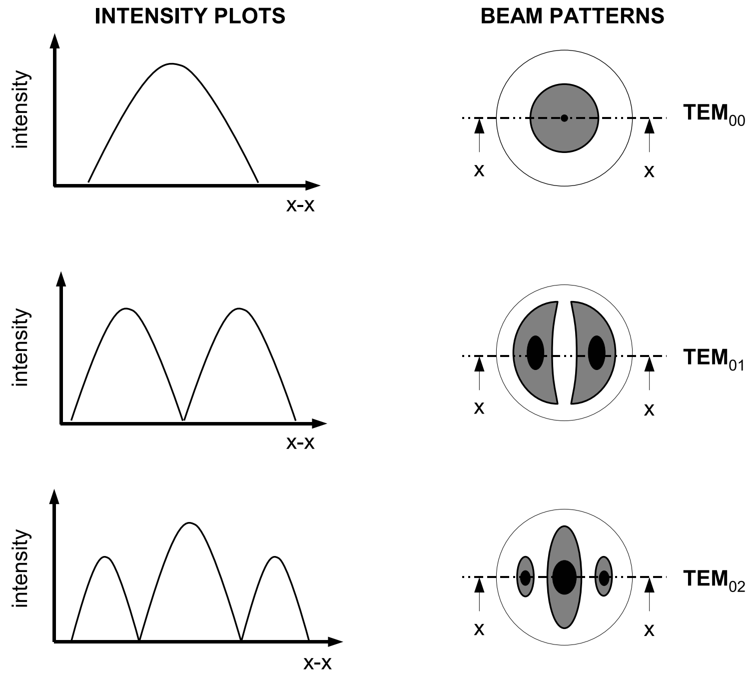


Figure 2.2: Examples of various transverse beam modes

Longitudinal Modes

In order for equation (2.2) to hold, a necessary boundary condition is for the value of the electric field at the mirrors of the resonator to be zero [4]. This results in a standing wave pattern to be formed within the laser cavity. These standing waves (or modes) which vary in the *axial* or *longitudinal* direction are called the longitudinal modes of the laser. It refers to the number of half wavelengths of the mode along the laser resonator. As stated previously, these modes are a result of equation (2.1). In the resonator, it is observed that the gain profile has a maximum value at a resonant frequency ν_0. Hence, in the resonator, the mode which provides the highest gain is normally selected when designing the laser. This is what is referred to as *single mode oscillation*.

In order for these longitudinal modes to develop,

- the gain within the laser must be much greater than the losses and
- the frequency of the mode must satisfy equation (2.2) [4].

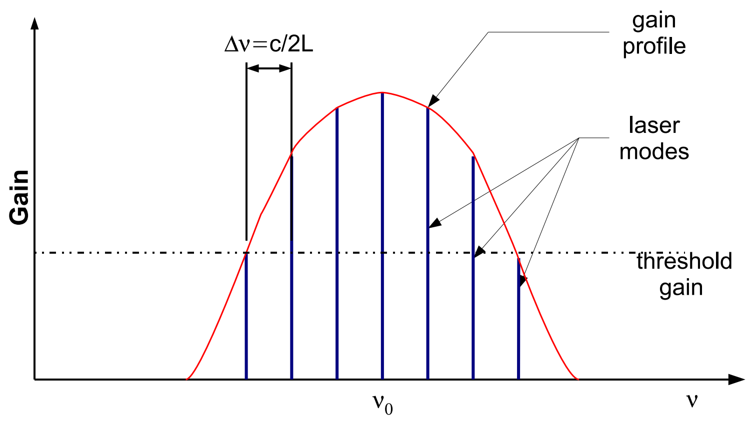


Figure 2.3: Gain bandwidth of a laser showing the various modes within the laser cavity (in thick lines). ν_0 is the resonant mode with the highest gain.

2.2 Ultrafast Pulses

As discussed earlier, laser beams could be emitted in either a continuous or pulsed mode. Lasers operate over a range of wavelengths (in the order of nanometres). It is important to be able to control the characteristics of laser pulses depending on what they are to be used for. One of the characteristics over which control is desired is the length of the pulse. This is because it is often useful to concentrate the laser energy into short time intervals as this boosts the output power (intensity) of the laser beam. Most lasers emit pulses of durations ranging from milli- to femtoseconds. This is especially useful when we intend to employ the laser in measuring various ultrafast processes in molecules and semi-conductors.

Some lasers may be able to generate relatively fast pulses in the order of nanoseconds. However, these pulses, could be 'compressed' to make them 'shorter' (and by implication faster) by means of **self modulation** and **pulse compression** [4]. *Self modulation* (also referred to as pulse or frequency chirping) involves the *spectral broadening* of the pulse. In essence an 'extra' bandwidth is added to the pulse. This is achieved by passing the pulse through a non-linear medium (i.e. a medium whose refractive index is dependent on the electric field passing through it) which results in the required increase in pulse frequency bandwidth. It is this increase in bandwidth that results in the pulse being narrowed in time [1, 4]. *Pulse compression* involves the 'compacting' of a pulse that has already undergone self modulation. This pulse is then made to interact with dispersive elements (which could be prisms or diffraction gratings) or to pass through media that have anomalous dispersive properties. This results in a 'squeezing' of the pulse width [4].

For most time-resolved methods, their spectral resolution, $\Delta\omega$ is usually determined by a Fourier limit

$$\Delta\omega = \frac{K}{\Delta\tau} \quad ;$$

where $\Delta\tau$ is the pulse duration and K is a numerical constant which depends on the pulse shape [5, 7]. These pulses are said to be *Fourier limited*. For a pulse that is Gaussian, we determine its 'transform limit' as follows:

For a normalised Gaussian distribution,

$$f(\tau) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(-\frac{\tau^2}{2\sigma^2}\right) \quad , \tag{2.4}$$

the Fourier transform is given by

$$f(\omega) = \frac{1}{\sqrt{2\pi}} \exp\left(-\frac{\omega^2\sigma^2}{2}\right) \quad , \tag{2.5}$$

where σ is the root mean square deviation of the function (equal to $\Delta\omega$ or $\Delta\tau$ depending on the function we are working with) [8]. At half-maximum, using equations (2.4) and (2.5) we derive

$$\Delta\tau_{1/2} = 2\sigma\sqrt{\ln 2} \quad \text{and} \quad \Delta\omega_{1/2} = \frac{2\sqrt{\ln 2}}{\sigma}. \tag{2.6}$$

This implies that the length between pulses $\Delta\tau$ and the frequency bandwidth, $\Delta\omega$ are related by

$$\begin{aligned} \Delta\tau_{1/2}\Delta\omega_{1/2} &= 4\ln 2 \\ \Rightarrow \quad \Delta\tau_{1/2} &= \frac{0.441}{\Delta\nu_{1/2}} \quad , \end{aligned} \tag{2.7}$$

where $\Delta\nu_{1/2}$ is the bandwidth of the laser. Equation (2.7) is referred to as the transform limit of a laser [2] and the pulse is said to be *transform limited* [4]. This determines how short we can make a laser pulse of a given frequency spectrum. A *bandwidth limited* pulse is governed by the relation [4]

$$\Delta\tau \sim \frac{1}{\Delta\nu} \quad , \tag{2.8}$$

where $\Delta\tau$ is the time duration between successive pulses and $\Delta\nu$ frequency separation between successive modes. Thus, it is observed that in order to have short laser pulses, a laser beam with a broad bandwidth i.e. a 'broadband' laser is required. The duration of these pulses range from milli- to femtoseconds and the means by which they are generated depends on the duration required. For lasers operating in the pulsed mode, ultrashort (or ultrafast) pulses are usually generated using three techniques: cavity dumping, Q-switching and mode-locking which are discussed below.

2.2.1 Cavity Dumping

The quality factor Q of a laser is a measure of the sharpness of the frequency transmission in the laser cavity. It is defined as the ratio between the resonant frequency ν_0 and the linewidth $\Delta\nu$ of a given laser mode i.e. [2, 3, 5]

$$Q = \frac{\nu_0}{\Delta\nu} \quad .$$

The process of cavity dumping works essentially by releasing the energy stored within the laser cavity. Unlike the resonator described earlier where the reflectivity of the output mirror M_2 is less than 1, in the cavity dumped resonator, both mirrors at the end of the resonator are totally reflective. This is in order to keep the losses within the resonator high and the Q value low [2, 3, 5]. As a result, there is a steady build up of power within this cavity. In order to have this power coupled out of the resonator, a third mirror is placed within the resonator and is oriented in such a way as to aim the laser beam out of the cavity. This mirror, M_3 is usually held at a reflectivity of zero until the power within the resonator builds up to the desired level. When this is achieved, the reflectivity is then switched to 1, upon which all the energy within the laser is 'dumped' out in a single pulse. On the other hand, this coupling mirror could have a reflectivity between 0 and 1 and the energy dumped lasts only as long as the light makes one trip around the cavity [2, 3]. A schematic diagram of a cavity dumped laser is as shown below.

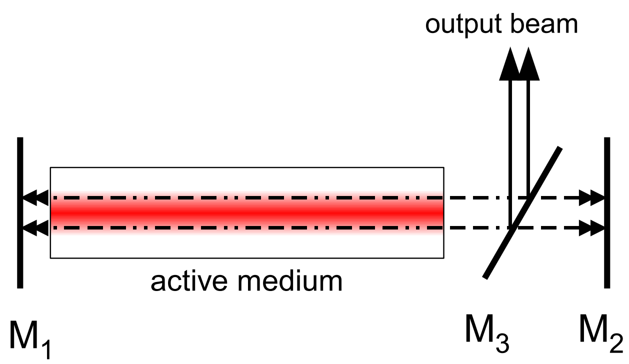


Figure 2.4: Schematic diagram of a cavity dumped laser resonator.

The length of the cavity dumped pulse is given by [2]

$$\Delta t = \frac{2L\eta}{c} \quad ,$$

where L is the cavity length, η the refractive index of the active medium and c the speed of light. Cavity dumping however, yields lower energy pulses than those generated using Q switching.

2.2.2 Q Switching

As mentioned in section 2.2.1, the Q factor of a laser beam measures its 'sharpness'. In essence, the Q factor measures the loss within a resonator. Thus, the higher the Q factor, the lower the loss within the resonator. Ultrafast pulses can be generated by selectively altering the Q factor of the resonator; a process referred to as 'Q switching.'

In this case, the cavity losses are kept high (implying a low Q) at the start of exciting the laser medium by means of an optical switch. As a result, the oscillation threshold of the laser cannot be reached and a very large population inversion is produced within the laser cavity. When the build up of photons reaches the desired level, the Q factor of the resonator is suddenly 'switched' (implying a high Q) and a quickly rising intense laser pulse is developed which quickly drains the energy that has been built up within the resonator [2, 5].

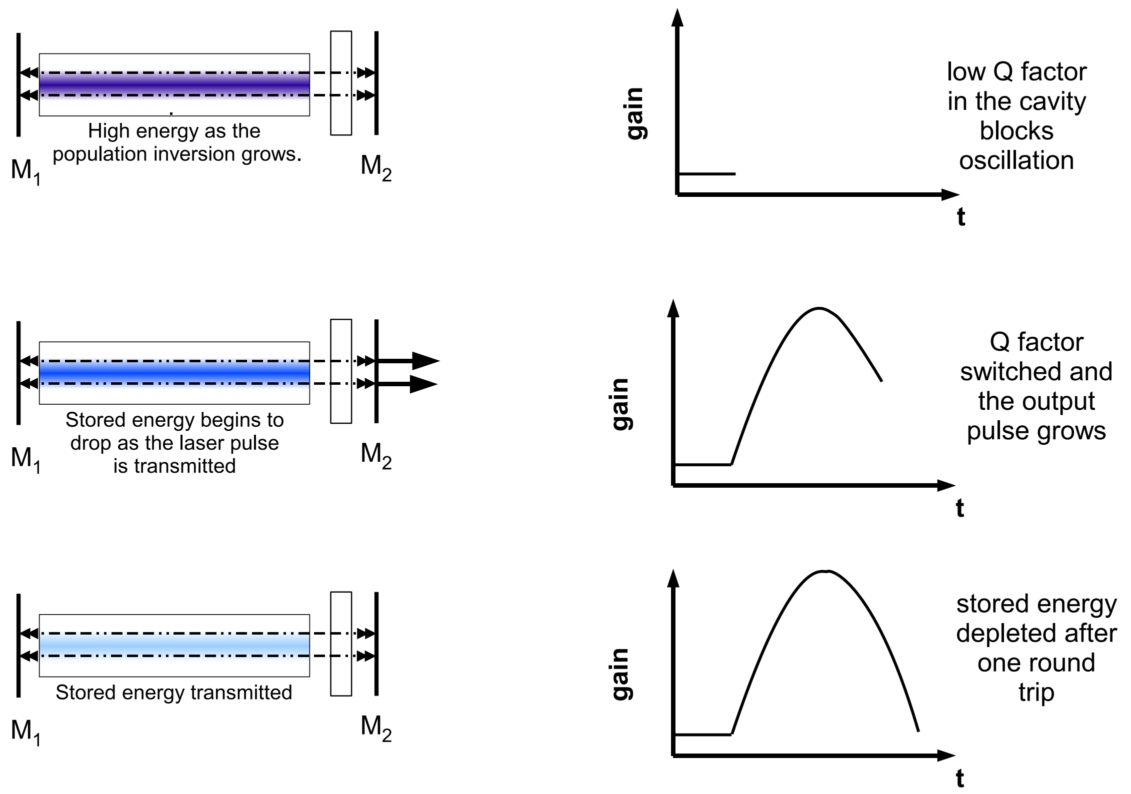


Figure 2.5: Schematic diagram showing the process of Q switching within a cavity resonator. Initially, the laser beam is not transmitted until the required population inversion has taken place and then the beam is transmitted in an intense short pulse.

A resonant cavity can be Q switched to produce a single 'giant' pulse or repeatedly to produce a series of pulses of varying intensities. Q switching can be achieved within a laser cavity by means of [4, 5]

- **Rotating Mirrors**: In this method, the optical switching within the resonator is achieved by mounting one of the mirrors on a rapidly spinning motor shaft. Thus, the laser beams are reflected back into the resonator only when the rotating mirror is aligned with the second mirror in the cavity.

- **Electro-Optic Shutters**: Electro-optic shutters can be used as fast optical devices for switching the Q factor within the cavity. The electro-optic device is usually a crystal such as a Pockels or Kerr cell that undergoes birefringence[1] when under the influence of an applied electric field [4]. This results in the polarization of the beam to be changed as it passes through it, allowing for the reflected beam to return into the laser cavity with a polarization of $90°$ with respect to the polarization it left the cavity with, thereby inducing

[1] Birefringence refers to the phenomenon when, under an applied electric field, a material has different refractive indices depending on the polarization of the field passing through it [1].

a low Q within the cavity [4]. As the wave continuously travels back and forth within the cavity, this 'polarization switch' results in the switching of the Q factor within the laser.

- **Acousto-Optic Shutters**: Similar to electro-optic devices, acousto-optic devices respond to changes in a radio frequency signal applied to it. The laser resonator is fitted with a transducer that converts the electric signal into an acoustic one. A typical acousto-optic device is a quartz crystal.

- **Saturable Absorbers**: Usually liquid dye materials, saturable absorbers can be used as a passive Q switching element within the resonator. When the active medium is pumped, spontaneous emission increases in intensity as the population of the upper laser level increases. A point is reached, however, when the spontaneous emission within the laser cavity results in the saturation of the absorber causing the a low loss to be developed within the absorber [4].

2.3 Mode-Locking

Another technique employed in generating short pulses is *mode-locking*. It is achieved by combining a number of unique modes of a laser of differing frequencies, but which are all coherent (i.e. they are in phase). The result is a wave characterised by a series of 'spikes' or pulses in the profile of the resulting wave amplitude which are separated in time by[2] $\Delta t_s = 2\eta L/c$. When the modes are coherent, these 'spikes' tend to be evenly distributed or repetitive, forming a train of pulses. On the other hand, combining together incoherent modes of a laser results in a random distribution of these spikes. The means by which mode-locking is achieved is derived below.

2.3.1 Superposition of Modes

As shown in section 2.1, the longitudinal mode separation is $\Delta\nu = c/2\eta L$, where c/η is the velocity of the beam in the resonant cavity of refractive index η and length L. Let the amplitude of the nth mode be

$$E(t) = E_n e^{i(\omega_n t + \phi_n)}, \tag{2.9}$$

with ω_n the frequency and ϕ_n the phase of the mode. If we assume that N modes of equal amplitude are oscillating simultaneously in the resonant cavity, then the combined total amplitude of the modes is

$$E(t) = E_0 \sum_{n=0}^{N-1} e^{i(\omega_n t + \phi_n)} \quad . \tag{2.10}$$

[2] See equation 2.17

Since these different modes are oscillating in a random fashion, the total intensity if the laser beam will be given by

$$\begin{aligned} I(t) \sim |\langle E(t)\rangle|^2 &= E_0^2 \sum_{m,n=0}^{N-1} e^{i[(\omega_n-\omega_m)t+(\phi_n-\phi_m)]} \\ &= E_0^2 \sum_{n=0}^{N-1} e^{i(\omega_n t+\phi_n)} e^{-i(\omega_n t+\phi_n)} \\ \Rightarrow \quad I &\sim NE_0^2 \end{aligned} \tag{2.11}$$

as expected. If all the modes are oscillating in phase, then we set $\phi_n = \phi_0$ for all n and equation (2.10) becomes

$$E(t) = E_0 e^{i\phi_0} \sum_{n=0}^{N-1} e^{i\omega_n t} \quad . \tag{2.12}$$

Setting $\omega_n = \omega_{N-1} - n\Delta\omega$, equation (2.12) becomes

$$\begin{aligned} E(t) &= E_0 e^{i\phi_0} \sum_{n=0}^{N-1} e^{i(\omega_{N-1}-n\Delta\omega)t} \\ &= E_0 e^{i(\phi_0+\omega_{N-1})t} \sum_{n=0}^{N-1} e^{-in\Delta\omega t} \\ &= E_0 e^{i(\phi_0+\omega_{N-1})t} \left[1 + e^{-i\Delta\omega t} + e^{-2i\Delta\omega t} + \cdots + e^{-i(N-1)\Delta\omega t}\right] \quad . \end{aligned} \tag{2.13}$$

We have that

$$\sum_{n=0}^{N-1} e^{-in\Delta\omega t} = \frac{1-e^{-iN\Delta\omega t}}{1-e^{-i\Delta\omega t}} \quad .$$

Thus, equation (2.13) becomes

$$E(t) = E_0 e^{i(\phi_0+\omega_{N-1})t} \left[\frac{1-e^{-iN\Delta\omega t}}{1-e^{-i\Delta\omega t}}\right] . \tag{2.14}$$

The total intensity of the beam is

$$\begin{aligned} I(t) &\sim E_0^2 \left|\left[\frac{1-e^{-iN\Delta\omega t}}{1-e^{-i\Delta\omega t}}\right]\right|^2 \\ &= E_0^2 \left[\frac{1-\cos N\Delta\omega t}{1-\cos \Delta\omega t}\right] \\ \therefore \quad I(t) &\sim E_0^2 \frac{\sin^2(N\Delta\omega t/2)}{\sin^2(\Delta\omega t/2)} \quad . \end{aligned} \tag{2.15}$$

The intensity varies with t but has a maximum at times t_n given by

$$\begin{aligned} \frac{\Delta\omega t_n}{2} &= 0, \pi, 2\pi, \cdots, n\pi \\ \Rightarrow \quad t_n &= \frac{2n\pi}{\Delta\omega} \quad . \end{aligned} \tag{2.16}$$

Using equation (2.16), the separation between two successive maxima of the pulses, referred to as the pulse repetition rate, is

$$\begin{aligned}\Delta t_s &= t_n - t_{n-1} \\ &= \frac{2n\pi}{\Delta\omega} - \frac{2(n-1)\pi}{\Delta\omega} \\ \Rightarrow \quad \Delta t_s &= \frac{2\pi}{\Delta\omega} = \frac{2\eta L}{c} \quad ; \end{aligned} \tag{2.17}$$

and the pulse duration $\Delta t = \frac{2\eta L}{Nc}$ where N is the number of modes. In order to evaluate the maximum (peak) value of the intensity, we evaluate equation (2.15) as $\Delta\omega/2$ approaches zero:

$$\begin{aligned} I(t)_{max} \sim \lim_{\Delta\omega/2 \to 0} E_0^2 \frac{\sin^2(N\Delta\omega t/2)}{\sin^2(\Delta\omega t/2)} &= E_0^2 \lim_{\Delta\omega/2 \to 0} \frac{(\sin N\Delta\omega t/2)^2}{(\sin \Delta\omega t/2)^2} \\ &= E_0^2 \frac{(N\Delta\omega t/2)^2}{(\Delta\omega t/2)^2} \\ \Rightarrow \quad I(t)_{max} &\sim E_0^2 N^2 \quad . \end{aligned} \tag{2.18}$$

Figure (2.6) below shows the resultant intensity profiles after a simulation of mode-locking of different modes of a laser was carried out.

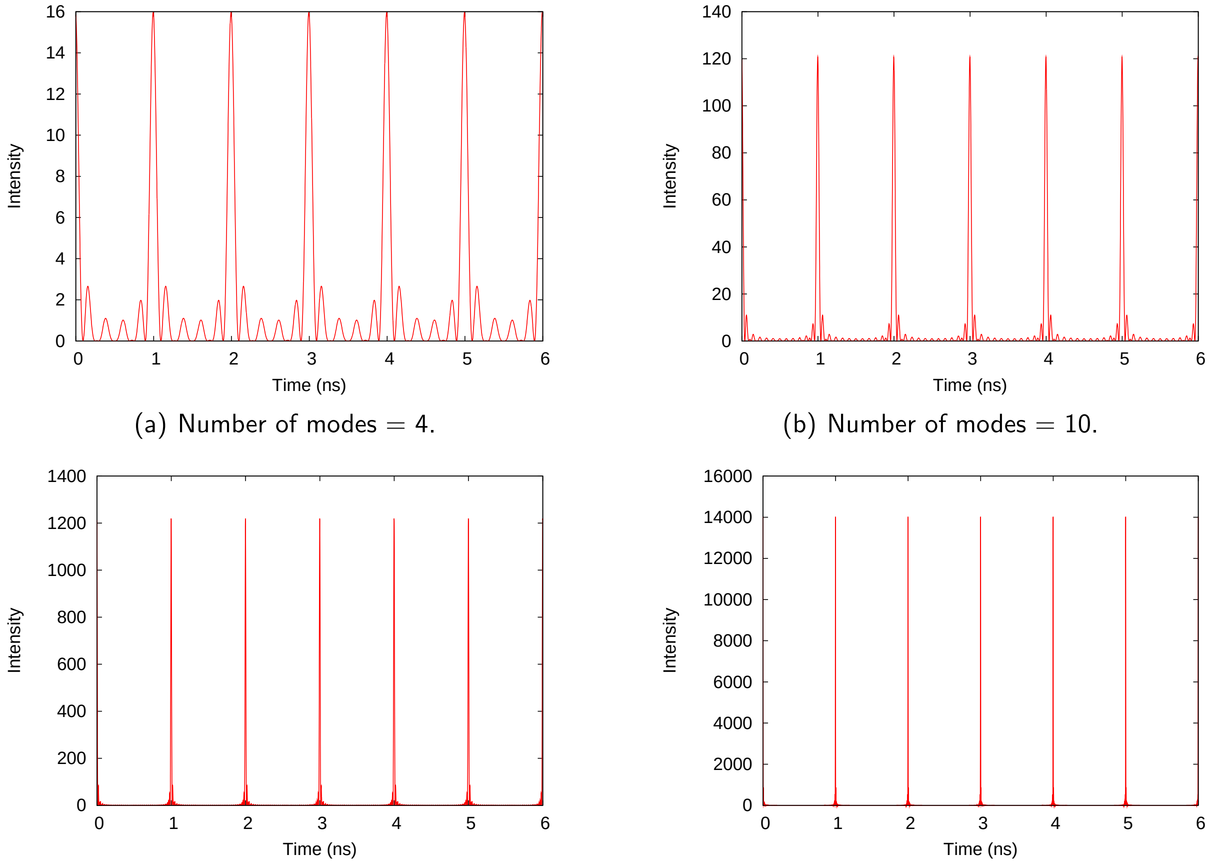
2.3.2 Mode-Locking Techniques

Mode-Locking techniques are generally classified as either *active* or *passive*. **Active mode-locking** involves the active element (or modulator) interacting with an external source. In other words, the 'behaviour' of the active element is dependent on the nature of the signal it receives. On the other hand, in **passive mode-locking** the modulating element within the laser is not driven by an external force but makes use of the non-linear properties of a material to produce mode-locking. Its behaviour, in essence, is independent of the light signal acting on it [2, 3].

There are three major ways in which active mode-locking is achieved. These are as follows [1, 3, 5]:

- **Amplitude Modulation (AM) Mode-Locking:** *Modulation* is a process by which the properties of a wave signal are varied in order for it to carry a desired signal [6]. In AM mode-locking, the amplitudes of the various modes of the laser are periodically modulated in order to achieve mode-locking. Unlike the illustration in section 2.3.1, where all the laser modes had equal amplitudes, the amplitudes of the modes being modulated are different and the final amplitude of the mode-locked pulse is a superposition of the different amplitudes of the modes [5].

- **Frequency Modulation (FM) Mode-Locking:** In this case, the *phases* of the laser modes are periodically modulated [1].



(a) Number of modes = 4. (b) Number of modes = 10.

(c) Number of modes = 30. (d) Number of modes = 100.

Figure 2.6: The figures show the result of adding together, *in phase*, different modes of a laser. Notice the repetitive nature of the ‘peak pulse’, the narrowing of the pulse bandwidth and the increasing intensity of the laser beam as the number of modes increases.

- **Synchronous Pumping:** This involves pumping the laser to be mode-locked with a long-pulse mode-locked laser. This causes the gain in the laser medium to be ‘switched on’ for a very short time as a result of the synchronisation with the mode-locked ‘pumping’ laser. In essence, the frequency of the mode-locked pulse that is to be generated is chosen in such a way as to be coincidental in time with the pumping pulse [3, 5].

Passive mode-locking can be achieved by:

- **Kerr Lens Mode-Locking (KLM):** Kerr lens mode-locking makes use of the intensity-dependent change in the refractive index of a material as a result of the non-linear polarisation caused by the electric field of an optical wave [4, 5]. For a laser beam with a Gaussian beam profile, the refractive index of the material will be much higher at the centre than at the edges; this results in the material acting as a lens that focuses the beam as it traverses the material. An example of the sort of material used to achieve this optical Kerr effect is a titanium-doped sapphire. Thus, for KLM, the self-focusing effect is used to select the preferred modes of the laser beam that are of the required ultrashort duration instead of allowing a steady continuous wave to be propagated [4]. The figure below is a schematic diagram of the KLM set-up.

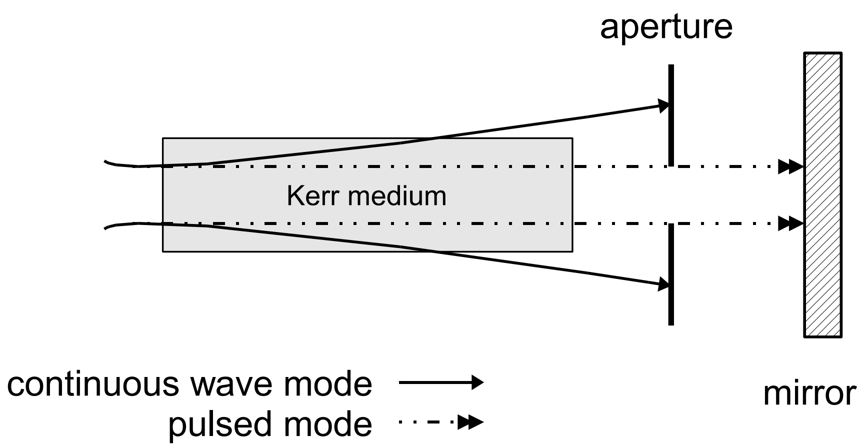


Figure 2.7: Schematic diagram of a KLM set-up. The aperture (or shutter) prevents the continuous laser mode from being propagated allowing only the pulsed mode, which is generated as a result of the Kerr effect, to be propagated.

- **Additive Pulse Mode-Locking (APM):** As the name implies, APM involves the addition of pulses. However, what occurs is that a portion of the laser beam generated is made to travel through an optical fibre and then coupled back into the laser cavity. This pulse is self modulated within the optical fibre and if added back into the cavity at the appropriate time adds on to the main pulse in such a manner as to reduce the effective length of the pulse and increase its intensity. This process is continuously repeated and results in the generation of a highly intense and fast pulse [4].
- **Colliding Pulse Mode-Locking (CPM):** CPM is obtained by the interaction of two counter-propagating waves within a saturable absorber in a laser cavity. As these two waves collide within the saturable absorber, they interfere in such a way as to create a highly intense standing wave [4].

3. Measuring Ultrafast Pulses

Many processes that occur at the molecular level do so on time scales that are relatively much shorter than what we can observe in everyday life. Processes such as molecular vibrations, photosynthesis, phonon dynamics and band gap excitation, all occur at times scales of the order of a few femtoseconds. As such, there is a limitation with respect to the kind of apparatus available to measure these processes because, in order to be able to determine how 'fast' these processes occur, it is necessary to 'compare' them with a much shorter event. This is where ultrafast pulses play a major role in the field of ultrafast sciences. With their use, it is possible to temporally resolve these events and obtain some idea of how fast they occur. However, this births another problem as to how we measure these pulses as, practically speaking, we have no shorter events to resolve them to. Thus, the most common means by which these pulses are measured is by comparing them with themselves—a technique referred to as **autocorrelation**.

3.1 Autocorrelation

The cross-correlation of two functions f and g is defined as [8]

$$C(x) = [f \otimes g](x) = \int_{-\infty}^{\infty} f^*(t)g(x+t)dt \tag{3.1}$$

where f^* is the complex conjugate of the function f. The cross-correlation function, $C(x)$ measures the degree of similarity between the two functions f and g. For functions that are completely *uncorrelated*, $C(x) = 0$. However, in a situation when $f = g$, then the cross-correlation function becomes

$$\begin{aligned} A_f(x) &= \int_{-\infty}^{\infty} f^*(t)f(x+t)dt \\ &= \int_{-\infty}^{\infty} f^*(t-x)f(t)dt \end{aligned} \tag{3.2}$$

and $A_f(x)$ is referred to as the *autocorrelation* of f [8]. It is used to describe how similar a function is to 'another version' of itself that has undergone some transformation of the variable t.

3.1.1 Optical Correlators

Using autocorrelation, it is possible to determine the duration of a light pulse. An optical correlator is a device that splits a light pulse into two beams, causes them to travel through different path lengths and superimposes them again. For a light pulse of intensity $I(t) = c\epsilon_0|E(t)|^2$ that has been split into two pulse beams of intensity $I_1(t)$ and $I_2(t)$, the coherent superposition of both pulses with a relative time delay τ yields a total intensity [5]

$$I(t,\tau) = c\epsilon_0[E_1(t) + E_2(t-\tau)]^2 \quad ; \tag{3.3}$$

where $E_1(t)$ and $E_2(t-\tau)$ are the electric fields of the two pulses. Using equation (3.2), we can write the autocorrelation of the electric field as

$$A_E(\tau) = \int_{-\infty}^{\infty} E(t)E^*(t-\tau)dt \quad , \tag{3.4}$$

and of the intensity as

$$A_I(\tau) = \int_{-\infty}^{\infty} I(t)I^*(t-\tau)dt \quad . \tag{3.5}$$

Thus, our optical correlator consists of a device that is able to detect the required autocorrelation term. For a linear detector, the output signal $S_L(\tau) \sim I(t)$. Thus, using equation (3.3) the signal measured over a time interval ΔT will be given by

$$\begin{aligned} S_L(\tau) \sim \langle I(t,\tau)\rangle &= \frac{1}{\Delta T}\int_{-\Delta T/2}^{\Delta T/2} I(t,\tau)dt \\ \Rightarrow \quad S_L(\tau) &\sim \int_{-\Delta T/2}^{\Delta T/2} |E_1(t)+E_2(t-\tau)|^2 dt \\ \therefore \quad S_L(\tau) &\sim \int_{-\infty}^{\infty} |E_1(t)+E_2(t-\tau)|^2 dt, \end{aligned} \tag{3.6}$$

since for a detector which has a response time $\Delta T \gg \Delta t$, (where Δt is the pulse duration), $\Delta T \to \infty$. Setting $E_1 = E_2 = E$, we have

$$\begin{aligned} S_L(\tau) &\sim \int_{-\infty}^{\infty} [|E(t)|^2 + |E(t-\tau)|^2 + E(t)E^*(t-\tau) + E^*(t)E(t-\tau)]dt \\ &\sim 2\int_{-\infty}^{\infty} |E(t)|^2 dt + 2\int_{-\infty}^{\infty} \Re[E(t)E^*(t-\tau)]dt \\ \Rightarrow \quad S_L(\tau) &\sim 1 + A_E(\tau) \quad , \end{aligned} \tag{3.7}$$

with

$$A_E(\tau) = \frac{\int_{-\infty}^{\infty} \Re[E(t)E^*(t-\tau)]dt}{\int_{-\infty}^{\infty} |E(t)|^2 dt}$$

if the amplitude of the electric field remains constant. From equation (3.7) above, we observe that the signal intensity measured by the linear detector is proportional to the autocorrelation, $A_E(\tau)$ of the electric field plus a background signal. The intensity measured by a linear detector is independent of the delay τ if the response time of the detector $\Delta T > \tau$ and hence, does not provide the required information regarding the time profile of the pulse. However, if the pulses are made to travel through a non-linear material that doubles the frequency of the signal, it generates an intensity signal proportional to the square of the intensities [5] i.e. $I(2\omega, t, \tau) \sim [I(t,\tau)]^2$ and

$$\begin{aligned} S_{NL}(2\omega,\tau) \sim I(2\omega,t,\tau) &\sim \int_{-\infty}^{\infty} |[E(t)+E(t-\tau)]^2|^2 dt \\ &\sim \int_{-\infty}^{\infty} |[E(t)+E(t-\tau)]^2|^2 dt \\ &= \int_{-\infty}^{\infty} |E^2(t)+E^2(t-\tau)+2E(t)E(t-\tau)|^2 dt \\ &= \int_{-\infty}^{\infty} [E^2(t)+E^2(t-\tau)+2E(t)E(t-\tau)] \\ &\quad \times \quad [E^{*2}(t)+E^{*2}(t-\tau)+2E^*(t)E^*(t-\tau)]dt \end{aligned}$$

$$\begin{aligned} \Rightarrow \quad S_{NL}(2\omega,\tau) &\sim 2\int_{-\infty}^{\infty} I^2(t)dt + 4\int_{-\infty}^{\infty} I(t)I(t-\tau)dt \\ &\quad + \quad 2\int_{-\infty}^{\infty} \Re[E^2(t)E^{2*}(t-\tau)]dt \\ &\quad + \quad 4\int_{-\infty}^{\infty} [I(t)+I(t-\tau)]\Re[E(t)E^*(t-\tau)]dt \ . \end{aligned} \tag{3.8}$$

Hence, the non-linear detector measures the product of the intensities of the pulses which is dependent on τ. Using equation (3.8), the peak to background ratio for interferometric autocorrelation is 8:1. This can be deduced by evaluating the equation at $\tau = 0$ and $\tau = \infty$. If the last two terms of equation (3.8) are removed, by making $E(t)$ and $E^*(t)$ orthogonal (as is the case in intensity autocorrelation) i.e.

$$\int_{-\infty}^{\infty} \Re[E(t)E^*(t-\tau)]dt = 0,$$

then equation (3.8) reduces to

$$\begin{aligned} S_{NL}(2\omega,\tau) &= 2\int_{-\infty}^{\infty} I^2(t)dt + 4\int_{-\infty}^{\infty} I(t)I(t-\tau)dt \\ \Rightarrow S_{NL}(2\omega,\tau) &\sim 1 + 2A_I(\tau) \quad . \end{aligned} \tag{3.9}$$

which is similar to equation (3.7) and

$$A_I(\tau) = \frac{2\int_{-\infty}^{\infty}[I(t)I(t-\tau)]dt}{\int_{-\infty}^{\infty}|I(t)|^2 dt}$$

. The peak to background ratio in this case is 3:1. A measurement of the signal $S_{NL}(2\omega,\tau)$ against the delay time τ then allows the time profile of the intensity $I(2\omega,t,\tau)$ to be obtained. In general, optical correlators measure the correlation between either the intensity $I(t)$ or field $E(t)$ of a light pulse at a time t, and a later time $t+\tau$. The two most common means of carrying this out is either by noncollinear intensity correlation or interferometric autocorrelation which are discussed below.

3.1.2 Noncollinear Intensity Autocorrelation

Noncollinear intensity autocorrelation involves measuring a pulse duration by using just one pulse. In 'single shot' measurements, the input beam is split into two beams that are made to interfere at an angle 2Θ, in a non-linear crystal. It involves overlapping the pulse at different angles so as to obtain a different time delay at different points in the pulse. If the beam waist[1] of the region of overlap is much larger than the length of the pulse, the distribution of the intensity of the second harmonic beam is proportional to the intensity autocorrelation function [9, 10]. This is sometimes referred to as the 'background free' measurement. In general, when determining the time profile of a light pulse, a pulse shape is initially assumed and the calculated correlation functions are compared with the experimental results. The duration of the autocorrelation signal, Δt, is related to the diameter of the second harmonic beam Δx, by the relationship [9, 10]

$$\Delta t = \frac{\Delta x \sin \Theta}{c_\eta} \quad , \tag{3.10}$$

where Θ is the half angle between the two beams and c_η is the speed of the beam. Shown below is a schematic setup of noncollinear correlation pulse measurements.

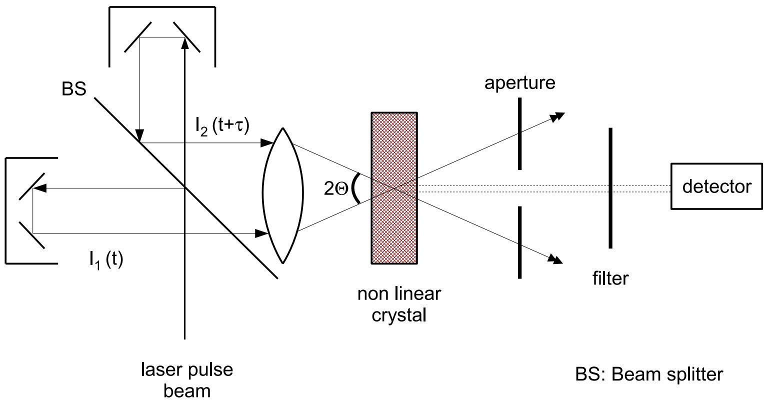


Figure 3.1: Setup for 'single shot' measurements of the noncollinear intensity correlation.

3.1.3 Interferometric Autocorrelation

Unlike in the noncollinear method, interferometric autocorrelation involves measuring the duration of a pulse by the *collinear* superposition of two pulses. The incoming laser pulse passes through a beam splitter which splits it into two parts which are made to travel through different pathlengths. They are then superimposed collinearly and focused into a non-linear crystal, by means of a lens, which then generates the desired output signal (as shown in equation (3.9) above). One of the key differences between interferometric and intensity autocorrelations is the presence of fringes in

[1]The beam waist of a laser pulse is defined as the smallest beam radius within a confocal resonator [5]

the output signal. Information regarding the number of fringes above the FWHM[2] is then used in determining the pulse duration.

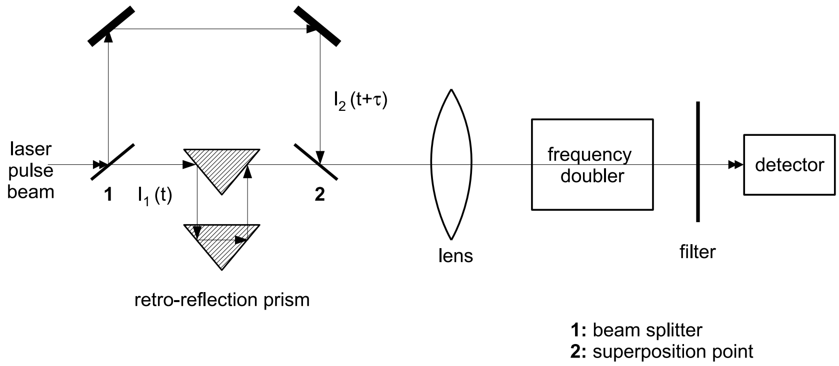


Figure 3.2: Optical correlator using a retroreflecting prism and second harmonic generation.

3.2 Other Measurement Techniques

Although the most commonly used, autocorrelation techniques are not the only means through which ultrafast pulses are measured. The development of fast photodetectors has made it possible to explore and develop other means of measurement. We will proceed to briefly discuss two other means of measuring ultrafast pulses, namely, streak cameras and the method of frequency resolved optical grating (FROG).

Streak Cameras

The streak camera is a device that measures the variation of the intensity of the pulse as a function of time. It operates by transforming the time profile of the beam into a spatial distribution which is then used to deduce the duration of the light pulse. This is achieved by focusing the beam onto a photocathode which generates a photoelectron pulse, which is then deflected by a pair of deflection plates and then imaged unto a luminescent screen as a light 'streak' [5].

Frequency Resolved Optical Gating (FROG)

The FROG method allows measurements of the third-order autocorrelation which might give information with regards to possible asymmetries in the ultrafast pulse and also the ability to make phase measurements [5]. Here, a Kerr cell replaces the second harmonic generator of

[2]Full width at half maximum (FWHM) is defined as the interval between the values of the wavelength that correspond to half of the maximum intensity of the laser beam [5].

autocorrelation and the pulse is represented as a two dimensional function of frequency and time. The signal measured at the Kerr gate is [5, 9]

$$E_s(t,\tau) \sim E(t) \cdot g(t-\tau) \quad , \tag{3.11}$$

with the gate function, $g(t-\tau) \sim I_2(t-\tau)$. Thus, the two dimensional intensity function measured by the detector (usually a CCD camera) is [5, 9]

$$I_t(\Omega,\tau) = \left| \int_{-\infty}^{\infty} E(t) \cdot g(t-\tau) e^{-i\Omega t} dt \right|^2 \quad . \tag{3.12}$$

Allowing the gate time to be much larger than the required delay time yields a time profile [5]

$$E(t) = \int_{-\infty}^{\infty} E_s(t,\tau) d\tau \quad . \tag{3.13}$$

The functions $E_s(t,\Omega)$ and $E_s(t,\tau)$ are related by the Fourier transform [9]

$$E_s(t,\Omega_\tau) = \frac{1}{2\pi} \int_{-\infty}^{\infty} E_s(t,\tau) e^{-i\Omega_\tau \cdot \tau} d\tau \quad . \tag{3.14}$$

Hence, using equations (3.13) and (3.14) the measured signal $S_E(\Omega,\tau)$, given by equation (3.12) becomes

$$I_E(\Omega,\tau) = \left| \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} E_s(t,\Omega_\tau) e^{-i\Omega t + i\Omega_\tau \cdot \tau} d\Omega_\tau dt \right|^2 \quad ; \tag{3.15}$$

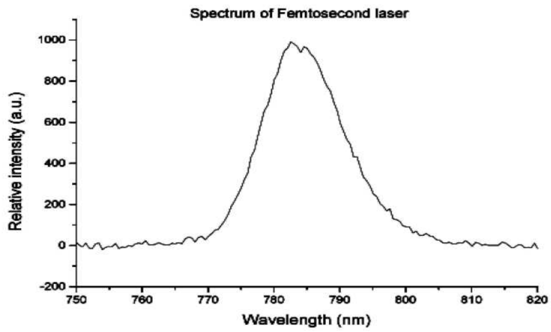
the desired signal $E(t,\Omega)$ is deduced by a two dimensional phase retrieval [5].

4. Experimental Results and Conclusion

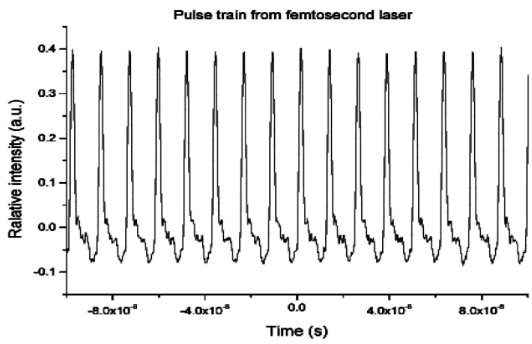
In order to illustrate how the duration of ultrafast pulses is determined experimentally, some real results will be used. Fig (4.1) below shows the output from a femtosecond mode-locked laser with a central wavelength of 785 nm and the corresponding pulse train as a function of time. Using the relationship $\nu_N = c/\lambda_N$ implies that the frequency bandwidth of the laser is

$$\Delta\nu_L \;=\; 3\times 10^8\left(\frac{1}{770\times 10^{-9}} - \frac{1}{800\times 10^{-9}}\right) \sim 15 \text{ THz.}$$

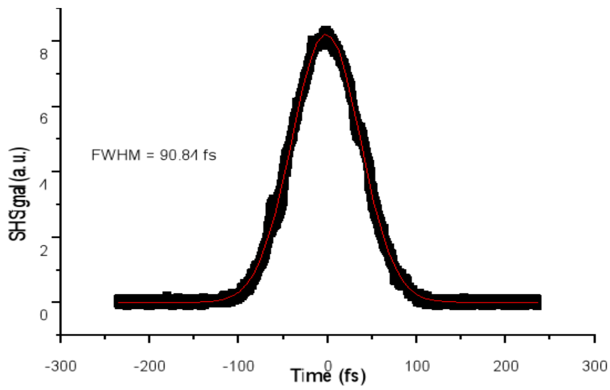
From fig (4.1(b)), the pulse repetition rate T was measured to be approximately 12 ns, hence the frequency spacing of the modes $\Delta\nu = 1/T$ is approximately 80 MHz.



(a) Results from a spectrometer showing the intensity spectrum as a function of wavelength for a broadband femtosecond laser.



(b) Results from a spectrometer showing the intensity spectrum as a function of time showing the repetitive nature of the pulses from a broadband femtosecond laser.



(c) Intensity profile obtained from a background-free autocorrelator.

Figure 4.1: Some experimental results.

Using equation (2.3), the resonator length $L = 1.8$ m. Hence, the number of modes oscillating is determined by

$$N \;=\; \frac{\Delta\nu_L}{\Delta\nu} \;=\; \frac{15\times 10^{12}}{80\times 10^{6}} \;=\; 187,500 \text{ modes.}$$

The average power was measured to be approximately 1 W and hence the energy $E = 10$ nJ. Using a background free intensity correlator, the pulse duration of the autocorrelated signal (see fig (4.1(c))) was determined to be 90.81 fs. Assuming a Gaussian pulse shape, the FHWM of the correlation signal is related to that of the 'real' signal by a factor of $\sqrt{2}$. Thus, the pulse duration of the laser beam is

$$\Delta t_m = \frac{90.81}{\sqrt{2}} = 64.2 \text{ fs} .$$

Using equation (2.8), the expected duration of the pulse is calculated to be

$$\Delta t \approx \frac{1}{\Delta \nu} = \frac{1}{15 \times 10^{12}} \approx 66.67 \text{ fs} \quad .$$

This result implies that the experimental results agree with the theoretical prediction of the bandwidth limit discussed in section 2.2.

4.1 Conclusion

This essay has discussed the basic principles governing the generation of laser beams and has focused on the means by which ultrafast laser pulses are generated. Population inversion within the active medium was shown to be a necessary condition for laser light amplification. This is because the intensity of the laser beam is dependent on the gain coefficient of the laser. When the gain coefficient is positive, there is an exponential growth in the intensity of the laser beam and a decay if it is negative.

Different means of generating these short pulses were highlighted with particular attention paid to the technique of mode-locking. By means of a simulation, it was shown how the pulse duration of a mode-locked femtosecond pulse becomes increasingly shorter as the number of locked modes are increased. This also results in an increase in the peak intensity of the pulse.

The measurement technique of autocorrelation was discussed and the duration of a mode-locked femtosecond pulse was determined experimentally. Interferometric autocorrelation produces a higher peak to background ratio than noncollinear intensity correlation (8:1 as compared to 3:1). By assuming a pulse shape and measuring the autocorrelation signal, the duration of an ultrafast pulse can be deduced. Other measurement techniques were also discussed.

Current research shows that even shorter pulses of duration in the order of a few femto- to atto- (10^{-18}) seconds and novel ways of measuring them are also being developed. These ultrafast pulses are used in telecommunications for faster signal transmission and as waveguides. Apart from being used in temporally resolving the duration of ultrafast phenomena, ultrafast pulses are also being employed in medicine for various surgical procedures. As further developments and new applications emerge, what the future portends for the field of ultrafast processes is one that will doubtless be of great interest to different fields of research.

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