Universität zu Köln

II. Physikalisches Institut

Entwicklung des M-Praktikumsversuch "Transient Grating"

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0   Introduction

Since many years laser technique gets more and more important in our life. Meanwhile it is possible to use them in the medicine to repair the eyes or to fight against cancer. It is possible to measure distances by measuring the time a laser beam needs to travel the double of a way. But lasers are not only used in those special domains, everyone who's got a PC or CD-Player profits on the laser technique. One of the reasons why lasers have such a big application area is that they are very multifaceted, e.g. by modify the wavelength or the repetition rate and by association the intensity. Hence, lasers are as that interesting for research so you are able to make a controlled modification of properties of a material.

In our case we are using laser to excite a semiconductor with a very high intensity beam, called pump. By this excitation we are adding much energy to the material or rather to the electrons. Now they are able to switch from the valence band into the conduction band. After we take off the pump the system decreases into its old state. A second laser beam with a lower intensity than the first one, called probe gets diffracted at the periodic excitation produced by the pump. This diffracted beam bears a lot of information about the properties and the processes within the material. So we excite the electrons with the pump and observe their behavior while the decrease. By using two different setups, the pump-probe and the transient grating, we are able to have a look on two different properties, the ambipolar diffusion and the relaxation of the electrons, anyway, the velocity and the lifetime of the electrons within the conduction band. Both characteristics are fundamental for the conductivity of the material. That's what we have to know to produce faster PC's or in general for the improvement of data transfer.

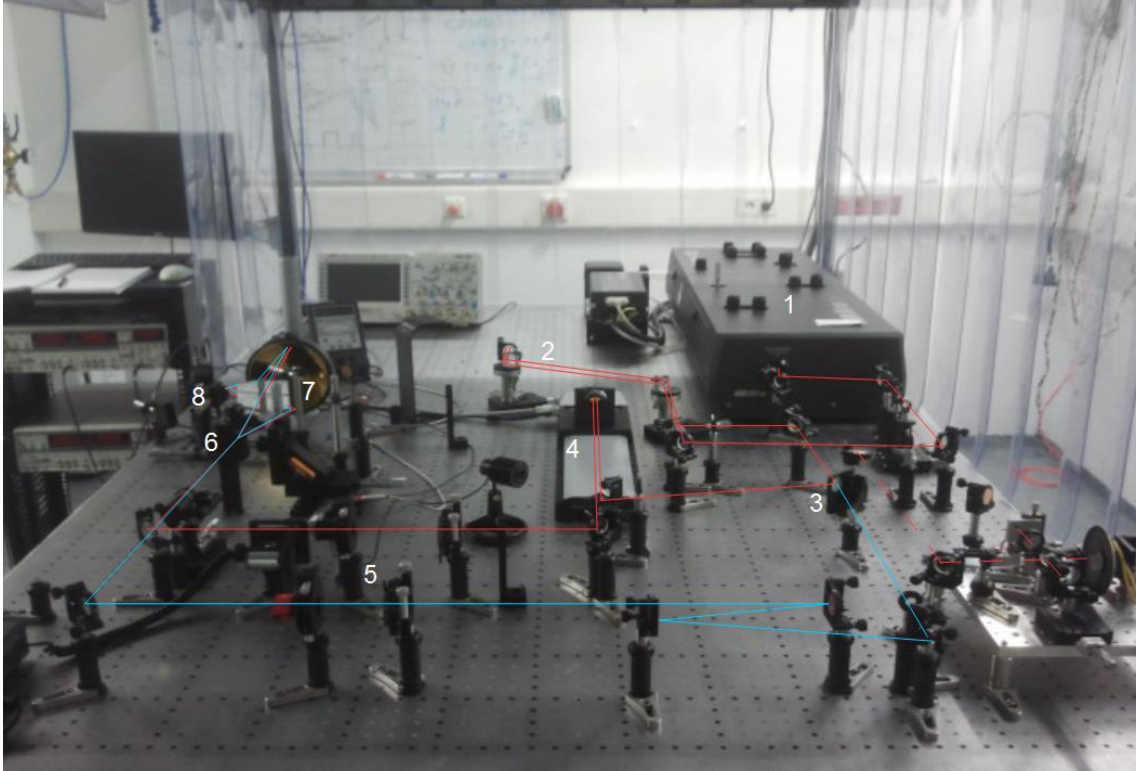This experiment tells you the story of a lonely laser beam which is about to change our life…

BE PART OF IT!

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1   Experimental setup

Picture 1: The experimental setup. Red line: Pump beam, blue line: Probe beam. 1: Laser; 2: Prism-pair; 3: Beam splitter; 4: Delay-line; 5: Wave plate and polarizer; 6: Phase mask; 7: constellation of mirrors; 8: Sample.
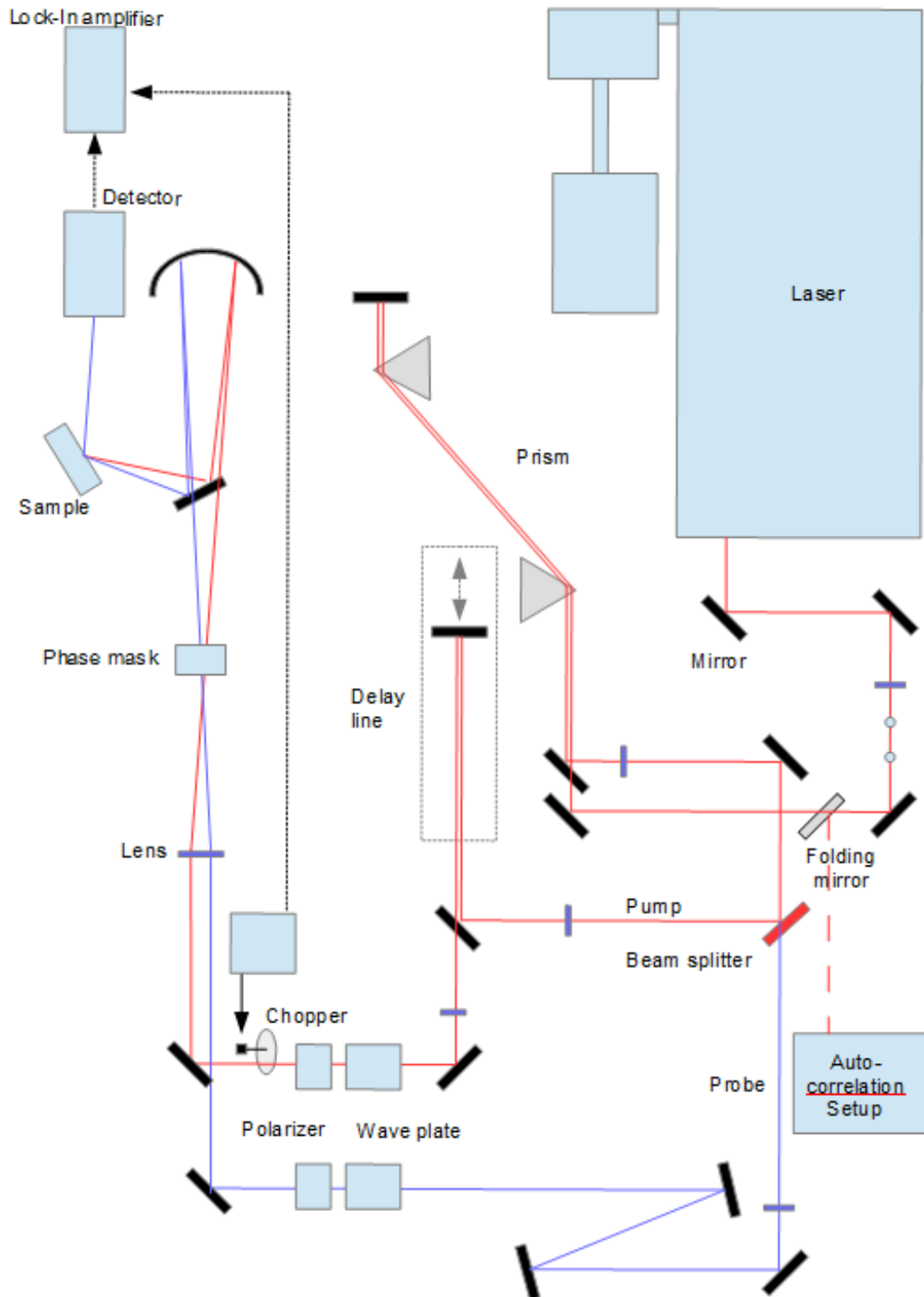
Figure 1: Schematics of the experimental setup shown in Pic.1. Red line: Pump beam, Blue line: Probe beam

1.1   The laser – ultra short pulses

The reason for using a laser which produces ultra-short laser pulses is very easy. In the year 1872 Eadweard Muybrigde wanted to answer the question whether a horse has a flying moment while galloping. Therefore he had to make photos faster than the hors gallops [1].
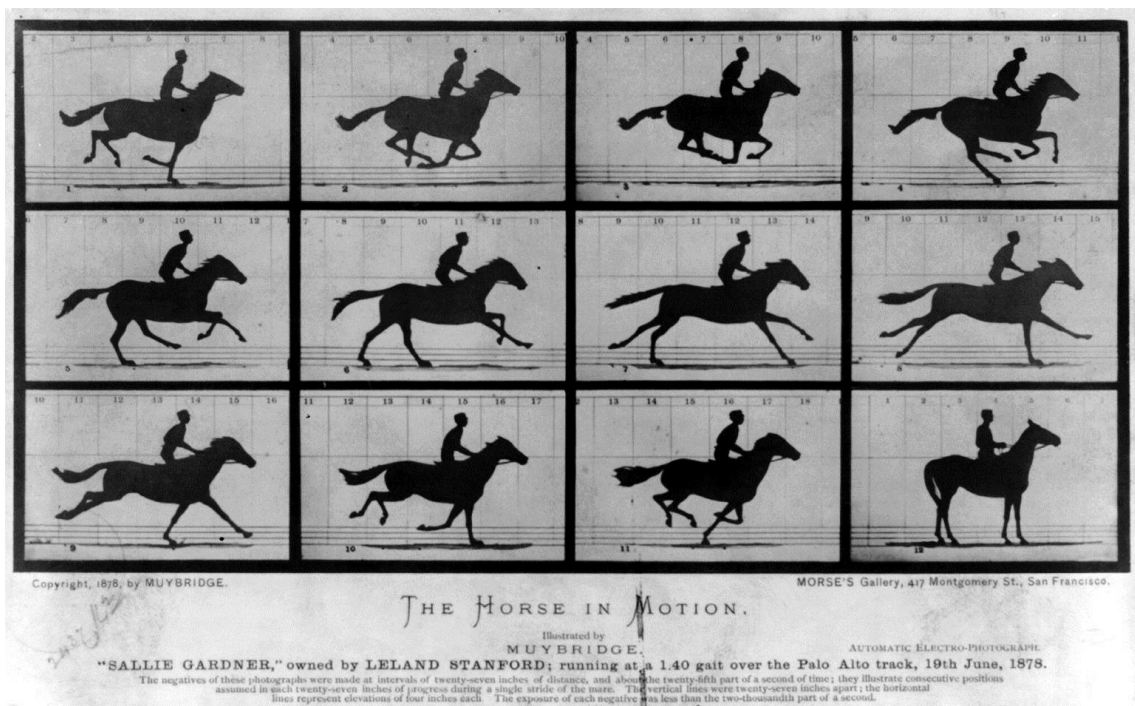
Picture 2: Horse in motion (1)

The processes we are watching are very fast, shorter than we are able to imagine. To observe them we need a measurement method which is faster than the process we want to measure. So to understand the experiment you have to understand lasers and ultra-short pulses first.

1	pump energy
2	particular mirror
3	laser beam
4	gain medium
5	mirror
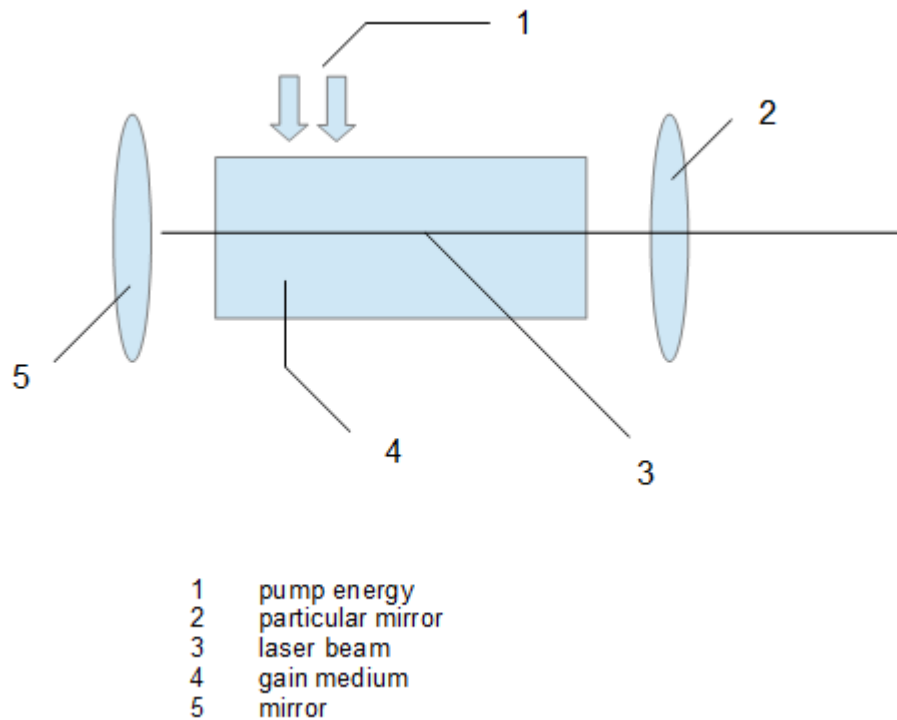
Figure 2: Basic structure of a laser (2)

Every laser is based on the same basic structure [2] (Fig. 2): an energy source to pump the gain medium, a gain medium to amplify the energy and a resonating cavity to steer the beams more times through the gain medium.

The main principle is quite easy. Einstein concluded that there exist three different types of basic interactions between light and matter [3]:

    1. Absorption

    2. Spontaneous Emission

    3. Stimulated Emission

Figure 3: Interaction between light and matter (3)

The energy source pumps the medium by raising the electrons in the atom into higher states. That concludes the excitation of electrons to higher energy levels by absorption of the photons from t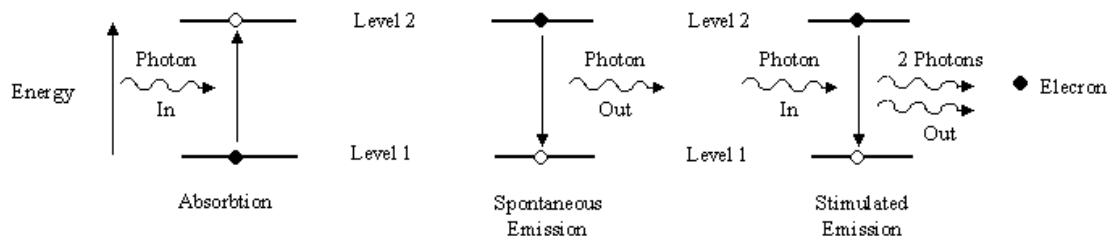he energy source. Afterwards the electrons are getting stimulated to emit photons. These emitted photons are getting reflected by a laser cavity of basically two mirrors so that they are going some more times through the medium.

1.1.1   Gain medium [4], [5]:

In this experiment we use the lasing medium Ti:Sapphire. This gain medium is made up of a crystal of sapphire (Al_2O_3) which is doped with titanium ions and has got some useful properties:

Picture 3: Gain medium Ti:Saphire (4)

1. Excellent thermal conductivity → alleviating thermal effects for high laser powers.

2. It has a very brad gain bandwidth → allows the generation of ultra-short pulses and wide wavelength tunability (650nm – 1100nm / the maximum efficiency is preserved at 800nm)

3. Wide range of possible pump wavelength → allows no necessity of powerful laser diodes → max. absorption at ~530nm

1.1.2 Energy source:

The pumping energy source has to produce light with the correct wavelength for absorption and emission in the gain medium. Usually a continuous wave laser is used to apply the right wavelength.

1.1.3 Resonating laser cavity:

The cavity is basically a pair of two mirrors which are reflecting the beams some more times through the gain medium. By doing this photons with the correct wavelength are supporting the energy source by doing the same. As a consequence of this the number of stimulated photons increases very fast.

The beam is almost ready. Only two things are left we want to change. First the energy of our beam and second like said in the beginning the pulse duration. We get there by the so called "mode-locking", a principle to lock the gain medium for special modes to amplify only wanted wavelengths. This goes with one useful effect and two little features:

1. The optical Kerr-lens effect
2. The aperture
3. A particularly reflecting mirror within the laser cavity

One of the decisive factors is the optical Kerr-lens effect [6]. This effect goes back to the influence of high energy light with matter. When the energy of the light is high enough the electric field gets larger and changes the refractive index of the matter. As a consequence of this light with a higher energy is exposed to a harder refraction than light with lower energy such as the continuous waves.
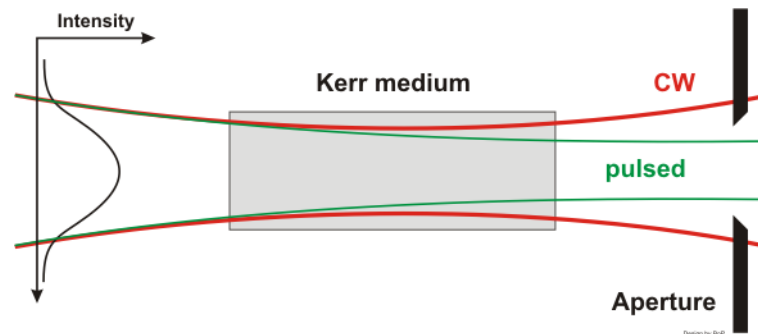

Figure 4: Optical Kerr-effect (5)

By the aperture the continuous wave gets coupled out of the cavity and doesn't reach the gain medium a second time. This self-focusing principle is also called "Kerr lens mode-locking". So by reflecting the out coming photons on the mirrors back to the medium they interact with the medium again and again on the same way. Consequently more and more identic photons with a high energy by the use of the optical Kerr-effect are produced. One little problem of this proceeded is the start-up problem [7], meaning that there is no intensive pulse in the beginning and the continuous waves are predominating. But by giving a little vibration to one of the mirrors, e.g. by banging on the table or vibrate the mirrors mechanically, the cavity length is changed and new modes will oscillate. Once the mode-locking process has begun it will continue without any help.

One of the two reflecting mirrors (Fig. 2) doesn't reflect 100% of the beam but rather 99%. Thus by every contact between beam and particular mirror a part of the light beam is coming out of the laser. By knowing this and the consideration that a standing wave is

needed inside of the laser, the length of the laser cavity has to be the half of the multiple of the wavelength, it is easy to get the period of the out coming beams:

$$L = \frac{m \cdot \lambda}{2}$$

$$\nu = \frac{m \cdot c}{2 \cdot L} \qquad \nu = \frac{c}{\lambda}$$

$$T = \frac{2 \cdot L}{c}$$

1.2   Mirrors and lenses

On its trip to the sample the laser beam has to pass many lenses and mirrors like you see in (FIG. 1). These are there to control and to focus the beam. To localize a beam you always need to fix two points in space so you need two mirrors and two irises to get a controlled beam.
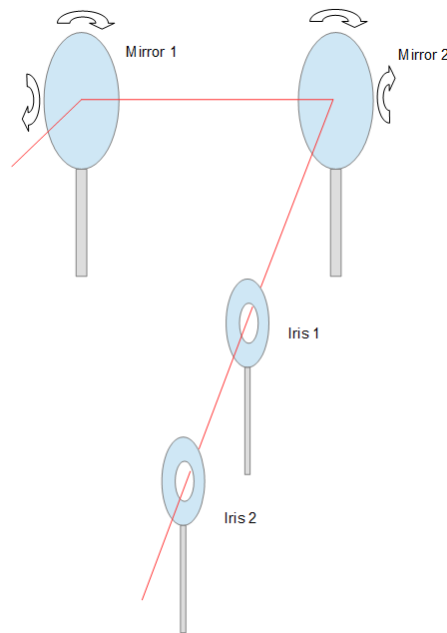

Figure 5: Exemplary constellation of mirrors and irises in the experimental setup

The experimental table would be very full of components if one would place two irises after every constellation of mirrors so one makes use of mobile irises.

But it is well known that when you reflect light with a mirror the reflected light is not as good as the originally light. So we need a kind of special mirrors which have low reflection losses, a high optical quality of the surface (that means good flatness, low microscopic roughness) and good resistance against high optical intensities to avoid damage. In this experiment we are using dielectric mirrors [8], [9]. This kind of mirror works with multiple thin layers of different transparent optical materials. Although the

reflection coefficient between two layers is very small the reflections from many interfaces can interfere. The result is a very high reflection of this mechanism.

1.3   **Prisms** [10], [11]

As mentioned beforehand, a short pulse in time has a broadband spectrum $\Delta v \sim \frac{1}{\Delta t}$. This has certain consequences, namely that a short pulse gets temporally broadened as it travels through a transparent material. In fact, when light crosses transparent materials the different frequencies of the pulse are susceptible to different refractive indexes, and the dependence of the refractive index with frequency, called dispersion. It resulted in different group velocities so that the red part of the light pulse has a higher velocity than the blue part within the transparent material. This produces a chirp in the pulse, resulting in a temporal broadening. In a laser cavity there are a lot of dispersive objects which are crossed several times by the beam. The light pulse gets some chirp every roundtrip.
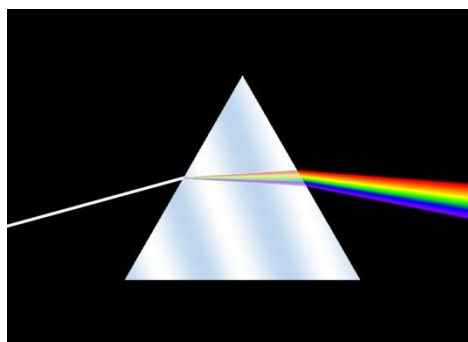
Picture 4: Dispersion of light in transparent material (6)

In order to compensate for the chirp induced by various dispersive objects in the beam path we use a so-called "prism compressor" (pc) pairs. A pc is made by a constellation of four prisms to correct the pulse by using the same dispersion to delete the dispersion.
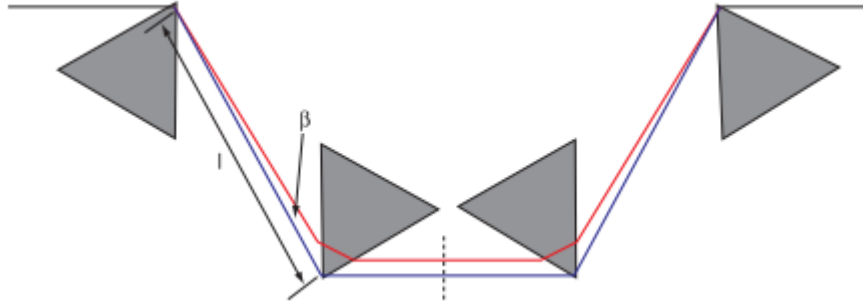

Figure 6: Prism constellation in Brewster angle to delete the dispersion (7)

In our experiment we're using only two prisms by placing a mirror at the dashed line. The prisms are placed to each other in such a way, that the beam enters and exits in the Brewster angle so that the reflection losses are minimized. It follows that the red part of the pulse has a longer way through the glass, than the blue part of the beam this compensates for of the chirp. After the laser beam passes the constellation of prisms the red and blue components are together again and the pulse is useable for our experiment. We can see how the prism compressor compensates the "positive" chirp (red part faster than blue part) by introducing an equal but opposite "negative" chirp in the system one prevent the given chirp of all the other mirrors and lenses in the beam path.

1.4   Fast Autocorrelation

But how are we able to measure the duration of an ultra-short laser pulse? With a very small calculation you can show, that you never can measure light with some electric devices:

$$m_{Ph} = 0 \qquad\qquad m_{el} = m_{el}$$

$$v = c \qquad\qquad v = v_F$$

$$v = 3 \cdot 10^8 \, m/_s \qquad v = 10^{3-5} \, m/_s$$

That means electrons in measuring instruments are always a lot more slowly than light $v_{Ph} \gg v_{el}$. So no electronic device is fast enough to measure such a short pulse therefore we have to resort to methods to measure the pulse with the pulse itself, by performing a so-called optical autocorrelation experiment. We use a Michelson interferometer for this (Pic. 4 / Fig. 7) [12]:
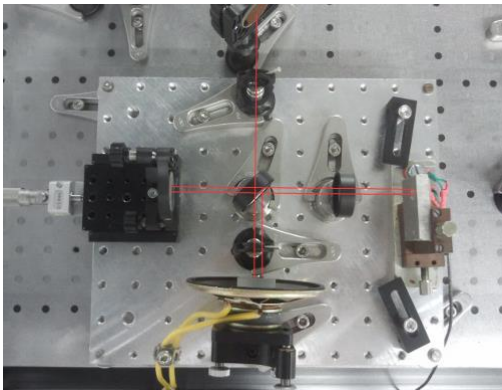
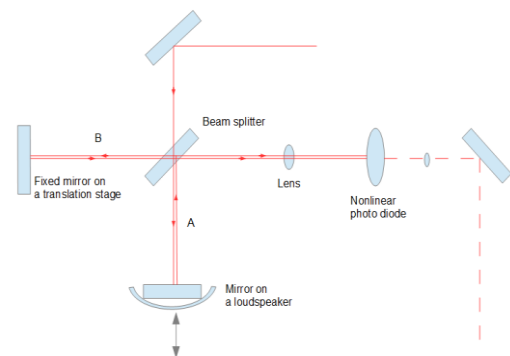**Picture 5:** Fast autocorrelation setup

Figure 7: Path of the beam within the autocorrelation setup

The main principle is to split a beam with a beam splitter to get two identical beams on the arms (A and B) and by vibrating the mirror on the loudspeaker the way the beams

have to travel is changed so that the delay between both beams is changed. We get the original pulse $x(t)$ and a time shifted identical copy $x(t - \tau)$ depending on the position of the vibrating mirror. Afterwards they are getting recombined in a second harmonic generation crystal [13] or a nonlinear photodiode, a nonlinear medium which response with a signal whose field has intensity proportional to the product of the intensities of the two pulses

$$I(t, \tau) \propto X(t) \cdot X(t - \tau). \tag{1}$$

Electric devices are too slow to measure this equation, but it is possible to measure the time integral of it, called an "intensity autocorrelation"

$$A(\tau) = \int_{-\infty}^{+\infty} I(t) \cdot I(t - \tau) \, dt. \tag{2}$$

So the value of correlation is getting big when the two signals are identical ($\tau = 0$), becoming smaller the bigger the delay between them.

Exercise 1:   Calculate the intensity autocorrelation of a Gaussian pulse
$I(t) = \exp(-t^2/(2\tau^2))$.

Exercise 2:   By using the definition (2) prove that the autocorrelation is an even function: $A(-\partial) = A(\partial)$
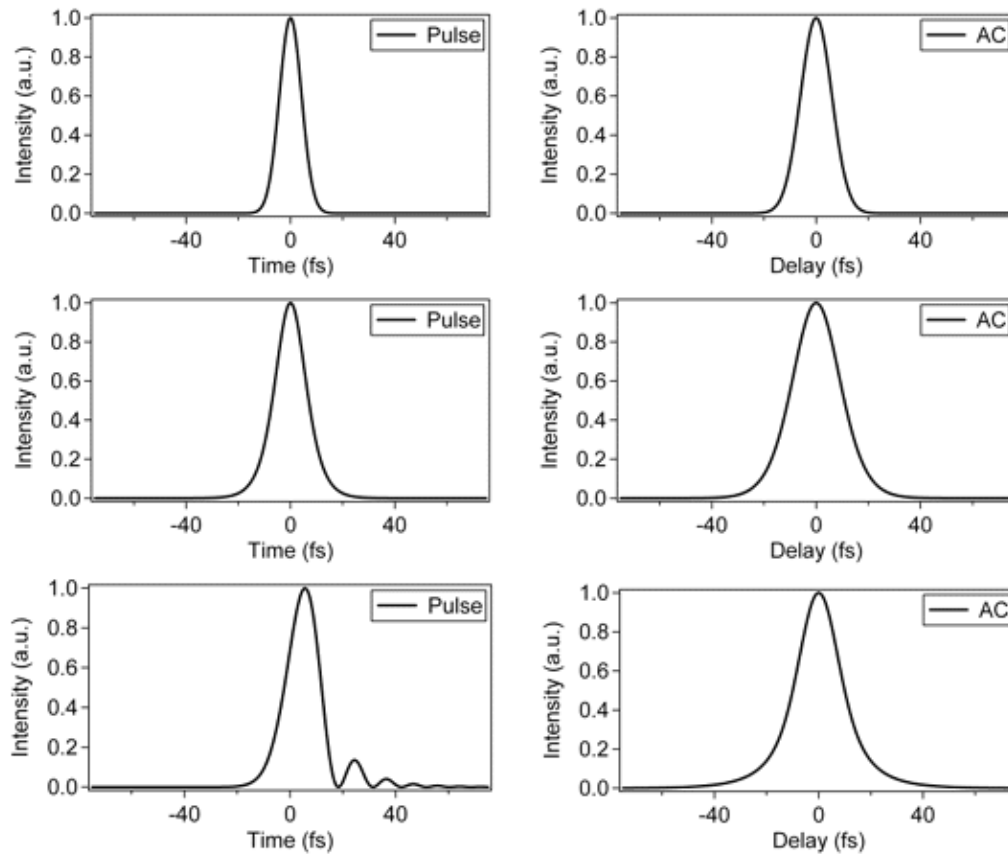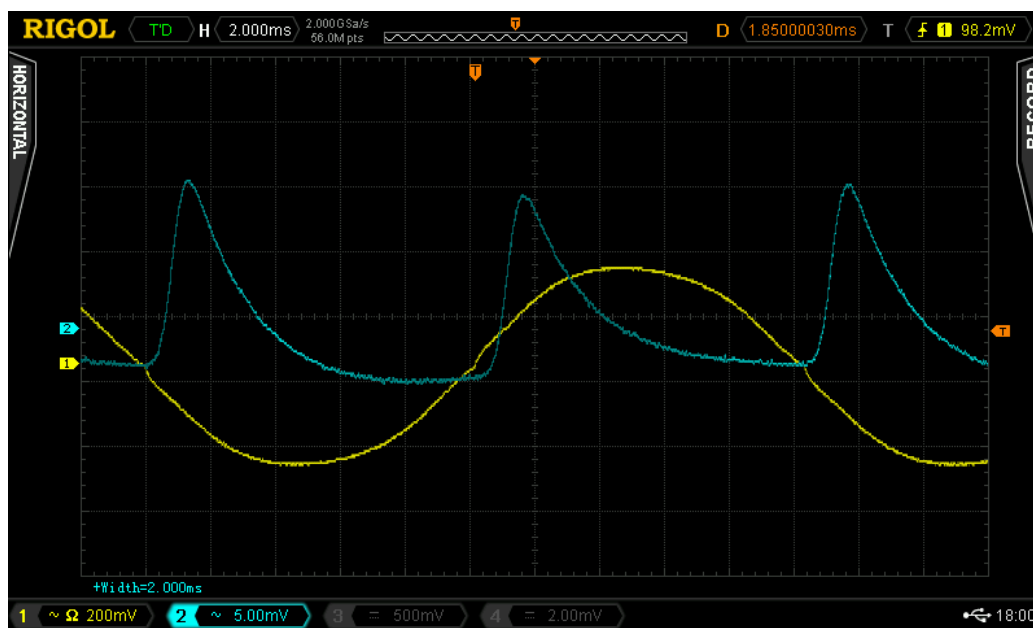
Figure 8: Examples of theoretical pulse intensies and their intensity autocorrelations. Left: Intensities vs. time. Right: The intensity autocorrelation corresponding to the pulse intensity to its left. (8)

Picture 6: Our autocorrelation signal on the oscilloscope. Yellow: Movement of the vibrating mirror, blue: Autocorrelation signal.

Now one wants to get out the pulse duration. This goes by sliding one pulse slowly past to the other while having a constant velocity. Otherwise the term is studded with too many velocities and this would make it very hard to calculate the pulse duration.

$$\Delta t_{AC} = \frac{2}{c} \cdot \frac{\Delta y}{\Delta t_{piek}} \cdot \Delta t_{FWHM} \tag{3}$$

With: $\Delta t_{AC} = Pulse\ duration\ of\ the\ autocorrelation$

              $\Delta y = Length\ you\ move\ the\ fixed\ mirror$

              $\Delta t_{piek} = Time\ shift\ between\ the\ pulses$

              $\Delta t_{FWHM} = FWHM\ of\ the\ autocorrelation\ pulse$

For Gaussian $\Delta t_{AC} = \sqrt{2} \cdot \Delta t_{pulse}$ takes effect.

Exercise 3:   Looking at picture 5. Explain why the autocorrelation showed as a blue trace indicates a misalignment of some parts in the setup.

1.5   Beam splitter

The integrated beam splitter divides the beam in two almost identical beams. The relative intensity between the two pulses is 3:1 (pump and probe). With the first beam, the pump, it is possible to excite the sample the second beam, the probe, gets diffracted at the excited states of the sample.

1.6   Delay

To perform a pump-probe measurement a more intense "pump" beam excites the sample subsequently, a second, dimmer "probe" pulse, coming at a controlled delay after the pump, is reflected by the sample. Therefore we need to control the delay between the pump and the probe with a good (~10fs) precision. So if there were no adjustable time-delay between the pump and probe beam you would always measure the same positions of the relaxing electrons. But finally you want to obtain a continuous movement of the electrons. Therefore you have to change the delay between the two pulses to measure other positions. To change the delay between them you only change the way of one of the beams. The longer the way, the more time the beam needs to travel.

$$\frac{2 \cdot \Delta s}{c} = \Delta t \qquad \frac{2 \mu m}{3 \cdot 10^8 m/s} \cong 6.6 \cdot 10^{-15} s$$

That means one micro meter change of the delay-stage brings a time-delay of 6.6 fs between the pump and the probe.

1.7   Wave plate and polarizer

After we split the beam and produce a time delay between them we have to control its intensity and polarization. Therefore we need this polarizer and wave plate.

The wave plate is a device which is able to change the phase of a electromagnetic wave running through it [14]. By two different refractive indexes along two axes it is possible to get different phase velocities of the light along the two axis inside of the medium with $n_{fast} < n_{slow}$. The component which is parallel to the fast axis passed the medium more quickly than the part parallel to the slow axis. After the medium the parts are shifted against each other and interfere again therefore the resulting beam has its

polarization state changed depending on the orientation of the plate and the incident light. In the experiment we use a wave plate that produces a shift of lambda half.

A polarizer is an optical device that filters from a beam one part with special polarization [15]. Other polarized parts get blocked or reflected. It's like trying to get a couch through a door, there are only a few positions to get it. The same principle is used in the polarizer, only special polarized waves can pass through it.

Figure 9: Polarizer (9)

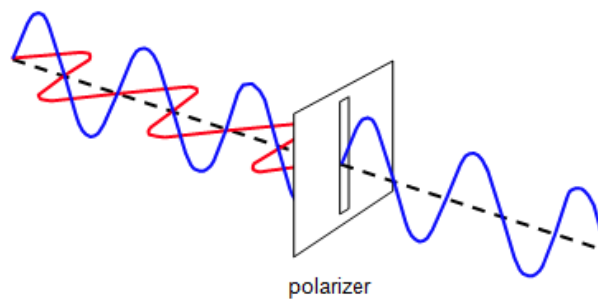## 1.8   Diffractive setup

The beam hits the home stretch to the sample. But to reach the one it has to pass some challenges, diffractive optics. A simple overview is shown in the next figure:
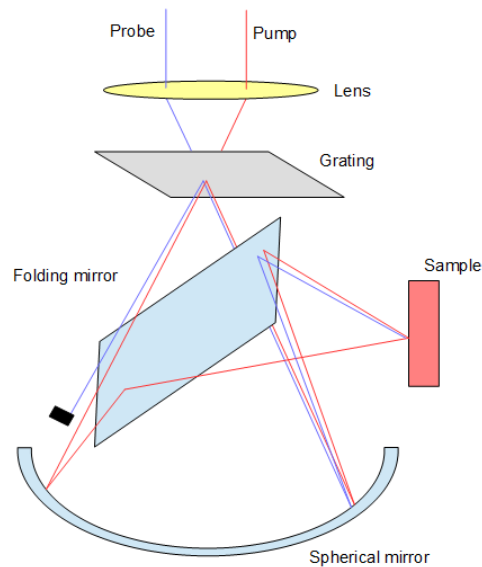
Figure 10: Diffractive setup with the main steps. Blue: Probe beam, Red: Pump beam

Let's go step by step through this special setup to the sample to see which tasks our beam has to manage to reach its aim.

1.8.1   Phase mask array

Before the beam reaches the sample it has to pass diffractive-optics in the form of a phase mask [16]. This is a special transmission grating which is made up of linear identical grooves etched on fused silica (SiO_2). It is formed like a plane gear with periodic identical grooves.
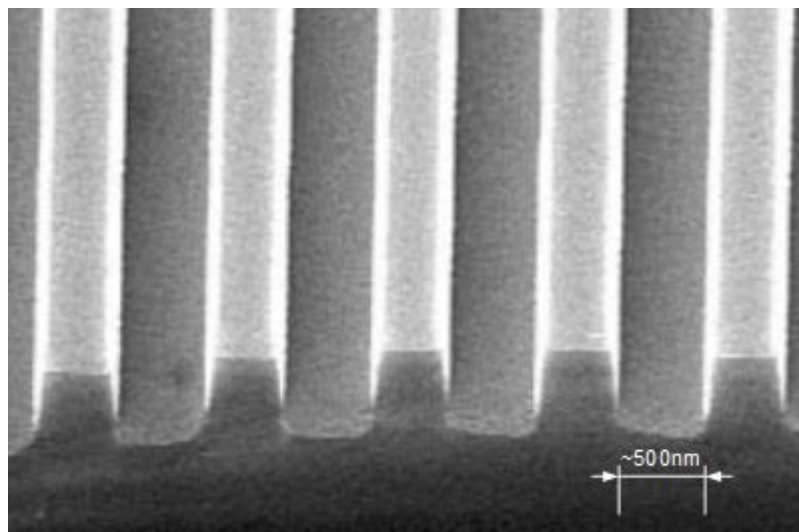
Picture 7: Enlarge phase mask profile (10)
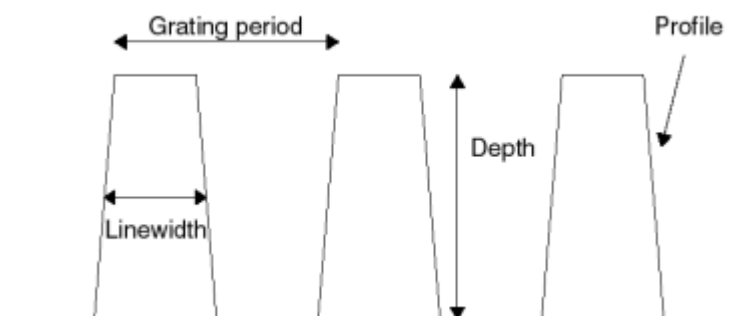
Figure 11: Phase mask (11)

As the incoming light beam hits the grating each groove acts like an emitter. The beam gets divided into several diffracted beams, along different directions (Fig. 12).
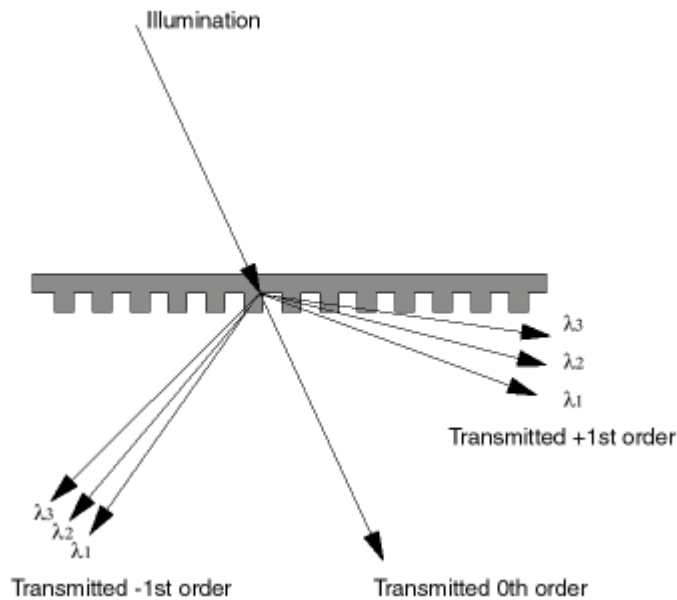
Figure 12: Phase mask divides the incoming beam into the different wavelengths and orders (12)

For each order, this equation holds:

$$m \cdot \lambda = \Lambda \cdot (\sin \theta_I + \sin \theta_D) \qquad (4)$$

Where:

m	-	m'th order of diffraction
λ	-	wavelength of the illumination
Λ	-	grating period
Θ_I	-	incidence angle
Θ_D	-	diffraction angle for the m'th diffraction order

By carefully optimizing the grating parameters (Fig. 11) for a certain wavelength it is possible to delete each beam except the two first-order-beams (+/- 1) get maximized. In the reality the first orders are amplified, more than 95% of the incident intensity while the other orders are supposed down to 1% of the incident intensity.
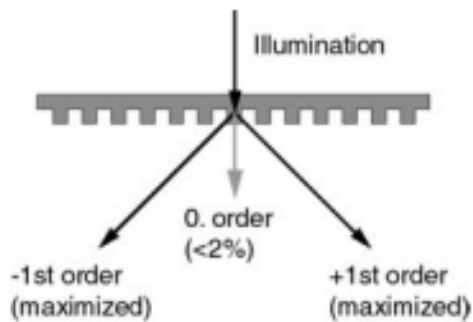
Figure 13: Phase mask which amplifies the first order (13)

Later on these two splitted pump beams, suitably focused by a large spherical mirror, will produce a grating on the surface of the sample. It is important to have the possibility to induce more than one grating periodically on the sample. This goes by changing the angle of the incoming beams. Therefore, there exist two feasible solutions. The first one would be to change the angle of the incoming beams manually, but this is the more difficult one because it is hard to re-align the beams afterwards. The second easier one is to install some more commutable phase masks of different periodicities (Fig. 14) so that you only have to move the phase mask and the system will be almost automatically aligned.

(measures in mm)
THICKNESS: 2 mm
LASER WL = 800 nm, 10 nm BW

SUPPORT: UVFS
SURFACE QUALITY 60-40 S-D
FLATNESS Lambda/10

PERIODICITIES (in micron)

1,76
2,23
2,7
3,1
3,5
3,9
4,8
5,6
6,5
8,0
12
16
24
28
32
40
48
60
68
80
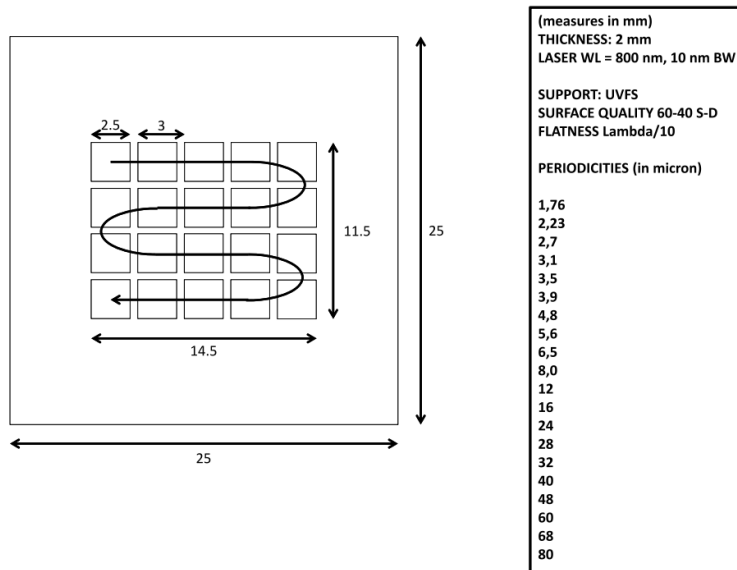
Figure 14: Phase mask array, used in the present experiment.

Like it is visible in (Fig. 13) the phase mask produces two beams from every incoming beam so that there are two pump and two probe beams afterwards. These beams will get focused on the sample surface by a combination of a spherical and a collecting mirror where the two pump beams interfere and produce the grating.

1.8.2   Intensity grating on the sample

Like mentioned the two pump beams are forming a grating on the sample. The periodicity depends on the incidence angle of the pumps [17].
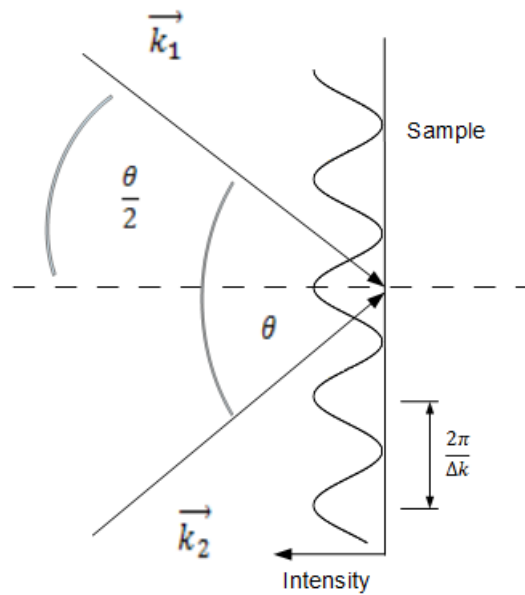
Figure 15: Pump beams interfere on the sample surface and produce the special grating

As a consequence, to change the grating period we have to change the angle of the interfering pump beams. So the more we enlarge the angle the more the grating period gets smaller.

Based on the same principle we change the intensity of the formed grating by changing the angle of the pump beams.

$$I \sim \left| e^{i\vec{k_1}\vec{r}} \cdot e^{i\vec{k_2}\vec{r}} \right|^2 \tag{4}$$

$$I \sim 2 + 2 \cdot \cos(i\vec{r} \cdot \Delta k) \tag{5}$$

With $\qquad \Delta k = \frac{4\pi}{\lambda} \cdot \sin\frac{\theta}{2}$

$$T = \frac{\lambda}{2 \cdot \sin\frac{\theta}{2}} \tag{6}$$

But what is this grating on the sample for? Why is this grating so important and what are the components of the grating?

After the phase mask two mirrors, one spherical and one plane, are used instead of a lens to steer and focus the beams on the same point on the sample. The both pump beams interfere on the sample surface and produce areas with constructive and areas with destructive interference. In other words, they are producing regions with high energy intensity and regions with low energy intensity (grating of intensity, grating of refractive index and grating of polarization). In the regions in which the energy intensity is high enough electrons of the sample are able to switch from the valence band into the conduction band. Electron-hole pairs which are neutral are arising from this and diffuse from the high density areas to the low density areas. This process is also called ambipolar diffusion [18]. Another process is the relaxation of electrons to their ground states. The change of the charge density is, of course, proportional to the change of the refractive index of the sample (see 1.10.1).

1.8.3 Four wave mixing

To describe the interaction of the induced grating and the probe bea, we use the tools of nonlinear systems. The four-wave-mixing [19] is an interaction of three incoming beams k_1, k_2, k_3 in a nonlinear medium. The interplay of the three fields yields a fourth field which wave vector is a combination of k_1, k_2, k_3. It is related to the third order nonlinear optical susceptibilities. A mathematical description the nonlinear response of the optical medium is the expansion of the induced polarization as a power series in the electric field strength:

$$P = \chi^{(1)}E + \chi^{(2)}EE + \chi^{(3)}EEE + ..$$

Where $\chi^{(1)}, \chi^{(2)}, \chi^{(3)}$ are first, second, third order nonlinear optical susceptibilities.

In our experiment all beams are coming from the same laser that means they all have the same frequency (degenerate four-wave-mixing, DFWM) [20] what makes the look on the experiment quite easier. Two pump beams build a grating on the sample which is readout by the third beam, the probe, which gets diffracted and reflected. The diffracted (fourth) beam is the construction of the interaction of all three previous beams respectively of the three interacting fields. In this case the scattered wave at $k_s = k_3 \pm (k_1 - k_2)$ is detected
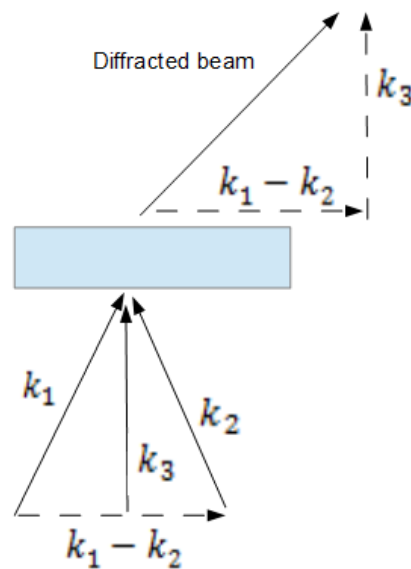

Figure 16: Four wave mixing, sketch of the direction of the diffracted beam

The probe beam penetrates the sample from a distance of $y = 1/\alpha$ known as the penetration depth [21] into the sample. So the area in which everything happened is so small that the beam sees only grating and no changing grating.

1.8.4   Boxcar geometry

The diffracted beam will be very weak, about 10^{-6}-10^{-10} of the incoming one, so two points are very important: First to maximize its intensity and second it is useful to know the direction of the wanted signal. The arrangement takes place in three dimensions, two pump beams and one probe beam to reach the conditions for the maximum intensity. A commonly used geometry used for this is the so called "boxcar" geometry (Fig.17, 18) [22]. The two pump beams $\vec{k_1}$, $\vec{k_2}$ and one probe beam $\vec{k_3}$ are parallel to each other and form three corners of a rectangle perpendicular to their path. The collecting mirror (or a lens) is used to focus the beams in on the sample in a very symmetric fashion. The two pump beams interfere on the sample surface and build the grating at which the probe beam gets diffracted. The diffracted beam quits the sample in direction $\vec{k_S}$ and satisfies the phase matching condition. This beam builds the fourth corner of the rectangle $\vec{k_s} = \vec{k_3} + \vec{k_2} - \vec{k_1}$.
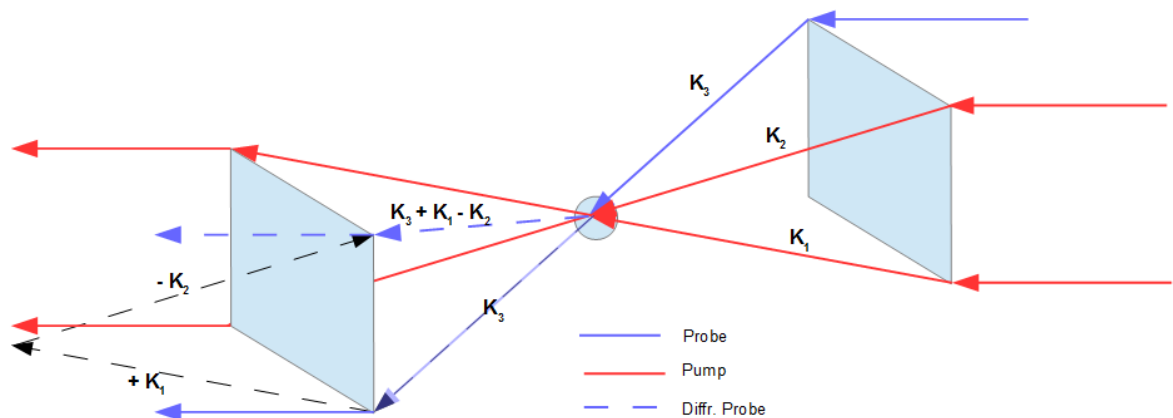
Figure 17: Boxcar geometry with the way of the pump, probe and diffracted beam

Figure 18: Boxcar geometry from another view

After the collecting mirrors which focus the four beams on the sample you can see nine light points instead of four. Those are emerged by the interference of the four basic beams so every beam is combined with each other.

Figure 19: The 9 spots. Interference of pump and probe

Picture 8: The 9 spots. Interference of pump and probe

To check and optimize the spatial and temporal superposition of the four beams at the sample position, the sample has to be in the focus, we use a non-linear crystal (α-BBO). By optimizing its orientation, we can produce the second harmonic of the four beams and the sum frequencies between them (Pic. 8). This results in a mesh of nine blue spots appearing after the crystal.
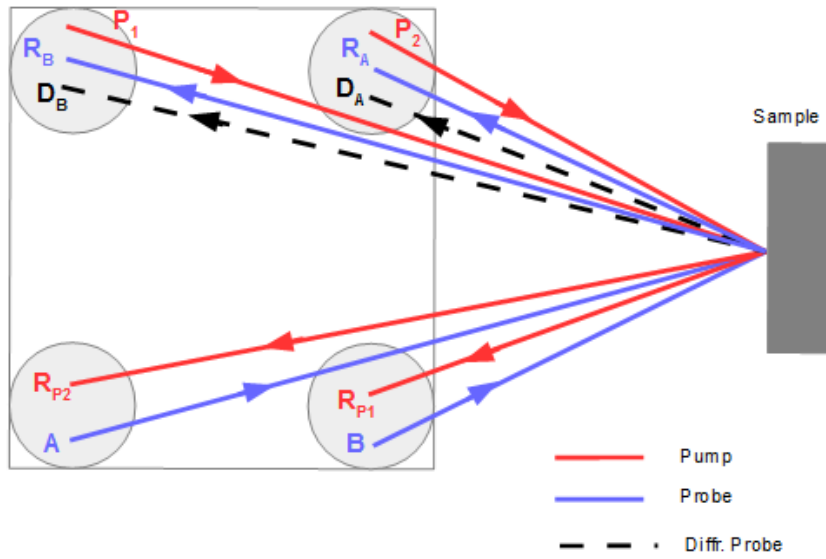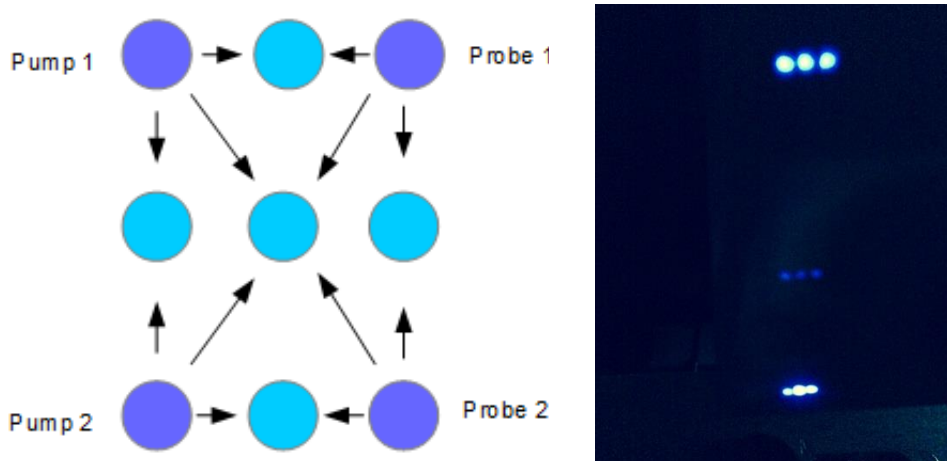
1.9   Sample

Finally the beam reaches its target the sample after a long and hard way through the setup. The sample consists of GaAs, a semiconductor called Gallium Arsenide. It's a compound of Gallium from the third main group and Arsenide from the fifth main group of the period system. The elements are building ionic bonds together so it's a polar semiconductor [23].

1.9.1   Crystal structure [24], [25]
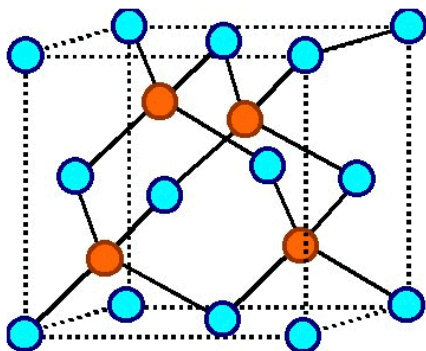
Mass density:	5.3174 g/cm³
Atom density:	4.4279×10^{22} cm^{-3}
Dielectric constant:	12.9
Lattice constant:	5,6533 Å (300K)

Figure 20: Structure of GaAs, Blue: Gallium, Red: Arsenide (14)

1.9.2   Band structure and optical properties
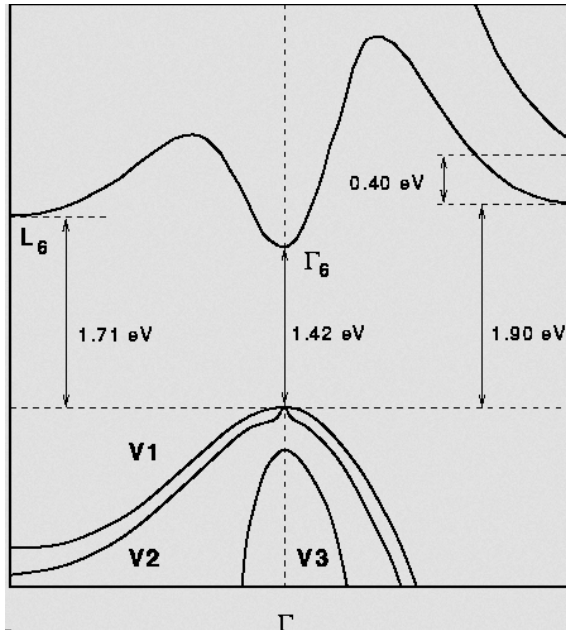
Figure 21: Band structure of GaAs (15)

Near zero doping and at room temperature (RT) the band gap between the conduction and the valance band amounts to 1.42 eV [26]. As can be seen from FIG 21 the minimum of energy in the conduction band is directly located above the maximum of energy in the valance band. That's the reason why GaAs belongs to the direct band gap semiconductor class. The structure of the conduction band defines the optical properties of the electron transport and the conductivity as well.

The band gap can be modified by doping the sample with other materials (n-doped; p-doped) which changes the size of the band gap and decreases the energy is needed to get over the gap. The general equation for the band gap energy is defined by [27]:

$$E_g(T) = E_g(0) - \frac{\alpha \cdot T^2}{T+\beta} \tag{7}$$

Where: $E_g - Energy\ of\ the\ band\ gap$

$E_g(0), \alpha, \beta - fitting\ parameter$

$T - Temperature$

For GaAs: $$E_g(T) = 1.519 - \frac{5.41 \cdot 10^{-4} \cdot T^2}{T+204} \tag{8}$$

$$E_g[eV] = \frac{1.519 - 5.41 \cdot 10^{-4} \cdot T[K]^2}{T[K]+204} \tag{9}$$

1.9.3   Optical excitation of materials

By optically excitation of a material like semiconductor many different processes take place. The first is the photoexcitation of electrons between the valence and the conduction band. In its ground state all possible states within the valence band are filled up while the conduction band is empty. But when one add energy to the material or rather excite the material, the electrons are able to switch to the conduction band and leave each a hole in the valence band, forming electron-hole pairs. After the photoexcitation the electrons relax by two different processes [28]. Firstly the electron-hole pairs diffuse from the area of excitation to areas without excitation by still staying in the conduction (electrons) and the valence (holes) band. This process is called ambipolar diffusion, and is governed by the velocity of electrons and holes. Secondly the excited electrons relax back into the valence band and refill the holes. This process is governed by the lifetime of electrons within the conduction band.
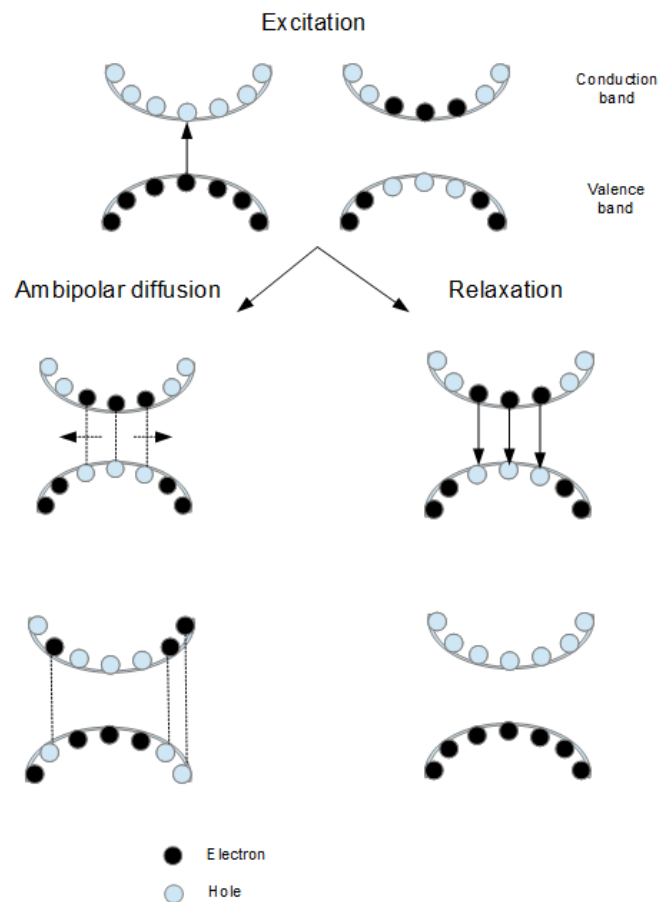
Figure 22: Different processes by optically excitation of a semiconductor

1.10   Detection

1.10.1 Diffracted beam

Thanks to the boxcar geometry we know that the diffraction of one probe is collinear with the reflection of the other so that we are able to detect it with a photodiode. There are several components which have an influence on the intensity of the diffracted beam [29], like the wavelength of the incoming beam, the refractive index change, the thickness of the sample and the excitation grating period and so on. But let's start on a simple level and getting step by step to the final goal, to the final equation.

If a material is subjected to an interference area of two pump beams the interactions between the light and the material create a spatial modulation of the material properties, e.g., exited electronic states, the density of conduction electrons, the refractive index and the absorption coefficient. The excitation grating impresses an optical grating on the material. This excitation depends on the incoming pump light intensity. Therefore the excitation amplitude has to be a function of the pump intensity and wavelength

$$\Delta X = g^{p}(\lambda_p) \cdot \Delta I(\lambda_p) \tag{10}$$

where g^p is a coupling coefficient which consists of the type of material excitation and the pump wavelength. The material excitation amplitude ΔX is proportional to the intensity amplitude ΔI. This change of material properties produces a grating-like modulation on the sample surface. These refractive index modification in time influences the interaction between the material and the probe beam: The refractive index $\Delta n(\lambda_C)$, the absorption coefficient $\Delta K(\lambda_C)$ and the excitation amplitude ΔK. Where Δn and ΔK are functions of the probe beam wavelength and depending on the excitation amplitude:

$$\Delta n = (\partial n/\partial X)\Delta X \tag{11}$$

$$\Delta K = (\partial K/\partial X)\Delta X \tag{12}$$

As opposed to make use of two optical parameters it is common to combine them in a complex refractive index:

$$\tilde{n} = n + iK/2k_C \tag{13}$$

$$\Delta\tilde{n} = \Delta n + i\Delta K/2k_C \tag{14}$$

Where k_C is the absolute value of the wave vector of the light for which the optical properties are measured.

So for an ideal plane wave interaction with constant intensity and grating amplitudes, the normalized intensity of the first order diffraction I_D/I_C is

$$\frac{I_D}{I_C} = \eta = \left|\frac{\pi\Delta\tilde{n}d}{\lambda_C}\right|^2 = \left(\frac{\pi\Delta nd}{\lambda_C}\right)^2 + \left(\frac{\Delta Kd}{4}\right)^2 \tag{15}$$

Where: $\eta - diffraction\ efficiency\ of\ a\ weak\ signal$
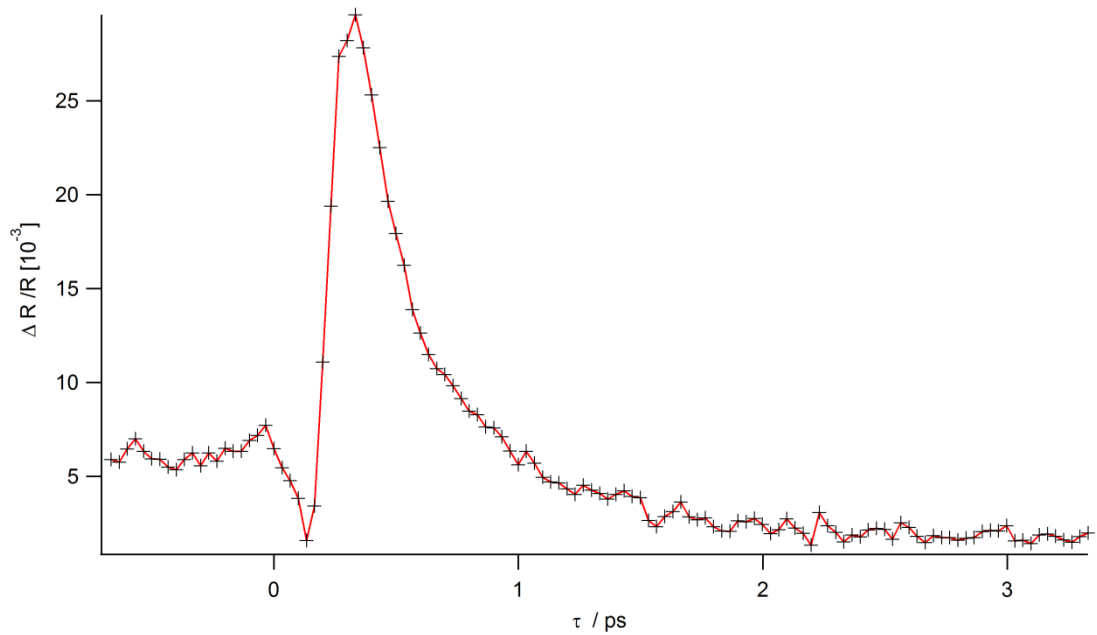
$d - thickness$
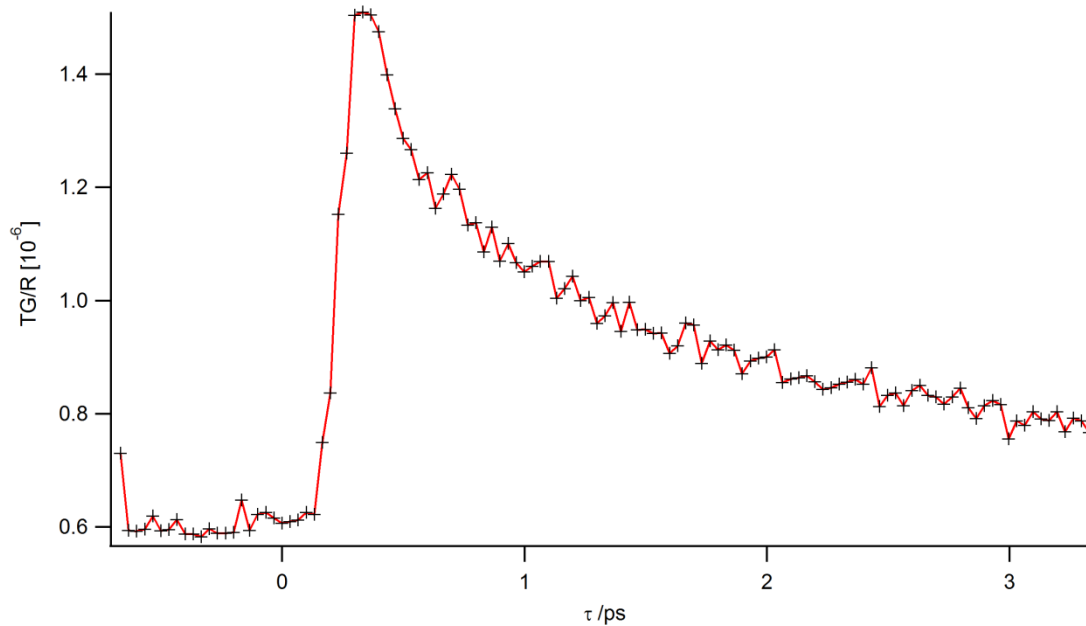

Figure 23: Pump-Probe spectrum

Figure 24: Transient grating spectrum

1.10.2 Lock-In amplifier

After our beam gets diffracted on the sample we want to measure the diffracted beam, the signal. The only problem is the overwhelming noises around which are much larger than the signal: $\frac{S}{N} \sim 10^{-2} - 10^{-10}$. Hence, we wouldn't measure anything without extract the signal from the noise. The only way to distinguish our real signal from all the rest of incoming waves is filtering the frequency spectrum of the signal [30]. The lock-in amplifier is used to measure very small signals by separating a part of the signal at a specific frequency and phase. Therefore we have to know the frequency of our signal. This goes with a trick by modulating the pump with an optical chopper. This in turn modulates the grating and the probe variation.

The lock-in amplifier uses the phase-sensitive detection (PSD). At it the signal is modulating with the controlled frequency of the grating by the chopper. The lock-in amplifies the diffracted signal and a low pass filter removes all the noise on other frequencies than the reference frequency. The PSD detects only the responses at this obtained frequency so that the signal gets separated.
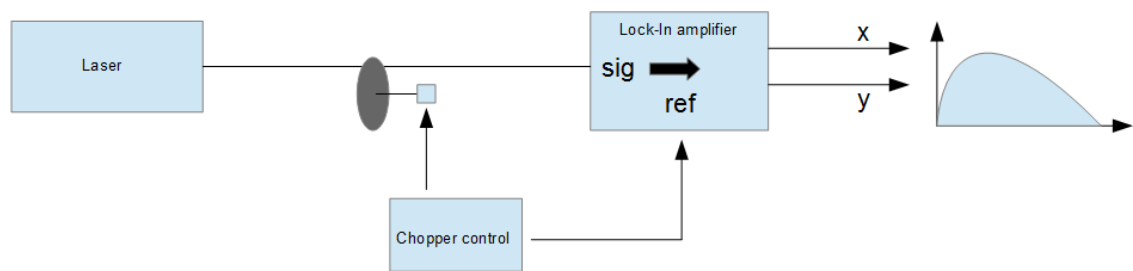
Figure 25: Lock-In amplifier within the experimental setup

At the same time, the probe signal from the photodiode is read by the lock-in, which is directed to extract the component of the signal modulated at the chopper frequency.

1.10.3 Average heating signal

When the pump pulse hits the sample a big number of electrons getting excite up to the conduction band. The sample needs some time to calm down, specially the electrons or electron-hole-pairs need a special time to decrease to their normal states. After the system calms down the next pump pulse arrives to the sample and excites it another time. But if the next pump pulse arrives at the sample before the system is in its normal states again you take the rest of the excitation with you in the next excitation period, called heating [31]. That means the higher the frequency of the pump pulse, specially the chopper, the lower is the time the system has to cool down. The lower the relaxation time the bigger the energy is taken with into the next excitation period the lower is the oscillation of the system. However, the Lock-In amplifier separates only the frequency of the chopper from the signal so that heating is better to see at a low frequency because of the bigger oscillation (see Fig. 22,23).

1.10.4 Pump-probe / Transient grating

In our experimental setup we have the opportunity to block every beam separately. This allows us to work with two different kinds of this experiment, with the 'pump-probe' and with the 'transient grating'. The difference between them is at pump-probe only one pump and one probe beam hits the sample by blocking one of the two pump beams after the phase mask. In the 'transient grating' version the experimental setup stays like discussed above with two pumps and one probe. These different setups allow us to separate and to observe different phenomena (Fig 26):

By using the 'pump-probe' setup it is possible to excite the sample at one single area. The system decreases almost exclusively by recombination of the excited electrons from the conduction band back to the valence band. Anyway, the lifetime of the electrons within the conduction band.

By using the 'transient grating' setup we are producing high excited areas flush against very low areas of charge density in turns. So the system decreases almost exclusively by diffusion of electron-hole pairs. So that it is feasible to measure the velocity of electrons within the electrons in excited states.
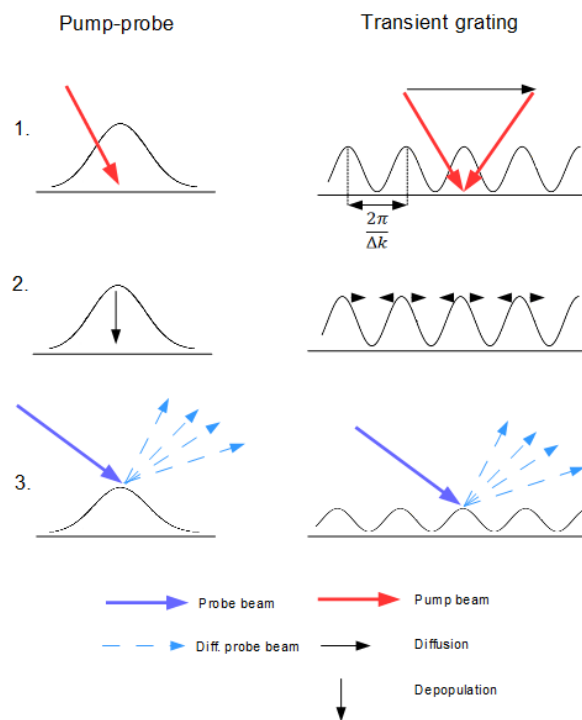

Figure 26: 'Pump-probe' and 'Transient grating'. 1. Excitation, 2. Relaxation/diffusion, 3. Diffraction

2   Practical application

First of all, this is an experiment with ultra-short laser pulses with a repetition rate about 80MHz. It is possible that some scattered beams are crossing through the room, if one of those beams finds the way into your eyes it is possible to lose your eyesight.

So please for your own safety, take care of your eyes and wear the safety googles!

Furthermore experiments with lasers, mirrors and lenses have always exacting optical paths. Moreover all the mirrors, the phase mask and lenses are exacting geared to each other. That's one of the reasons why the whole experimental table is build up on an air bearing to absorb the most of vibration. If one element gets changed every following element has to get aligned again and that takes up a lot of time.

So please would you be so kind as to touch nothing before your supervisor told you so!

2.1   Autocorrelation

At first we want you to measure the duration of one laser pulse. Therefore you have to open the folding mirror to change the path of the beam to steer it into the autocorrelation setup. Now you have to align the mirrors, hold in mind that this could take a lot of time.

1. Blocking the fixed mirror e.g. with a piece of paper
2. Take out of the setup the lens and the photodiode
3. Steer the reflected beam of the vibrating mirror almost back so that it get blocked by one of the irises on the way back to the laser (why?). It is possible to change its axes by turning the little wheels on its back.

4. Make sure that the beam in direction to the photodiode is going well through the iris. Mark the point of the beam on the wall (maybe on a piece of tape so that you can change it back)

5. From now on, do not touch the vibrating mirror again!!

6. Now block the vibrating mirror and open the fixed mirror

7. Steer the beam through the iris on the same point where you marked the first beam.

8. Mount the lens and insert the photodiode in the focus of the beams.

9. Try to get a signal by changing the distance of the fixed mirror to the beam splitter. To get a better signal average it.
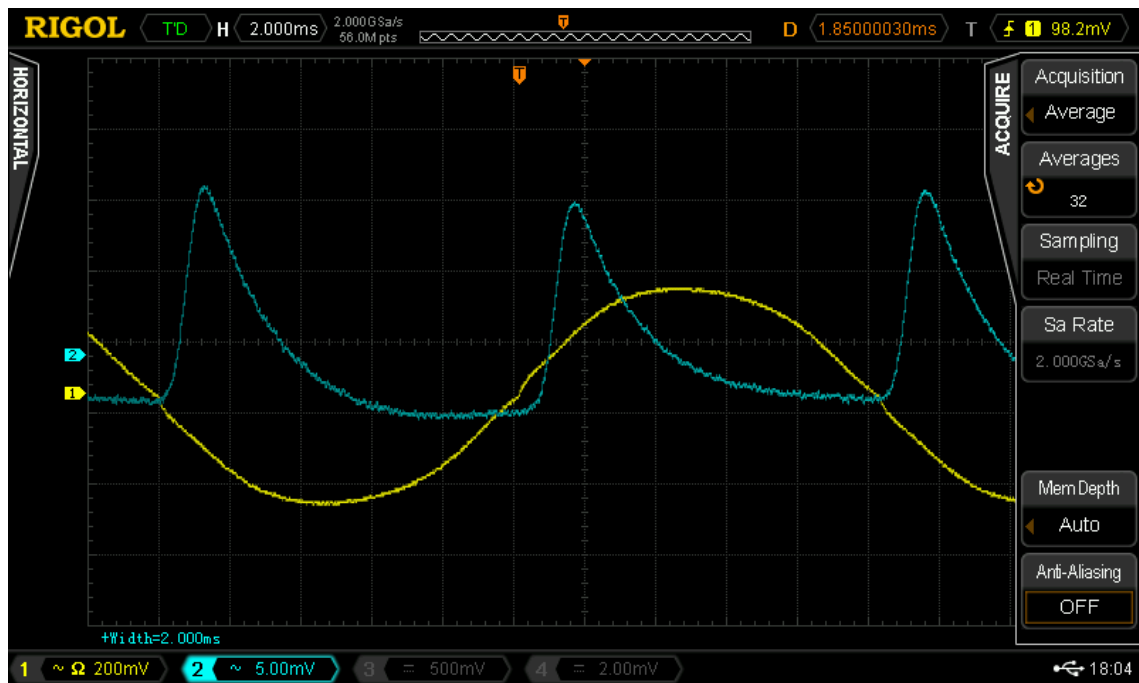
Picture 9: Autocorrelation signal; Yellow: Movement of the vibrating mirror, blue: Autocorrelation signal

10. Try to get the same distance between peaks. If so, you get the autocorrelation in areas where the vibrating mirror moves almost with the same velocity. Why is that so important?
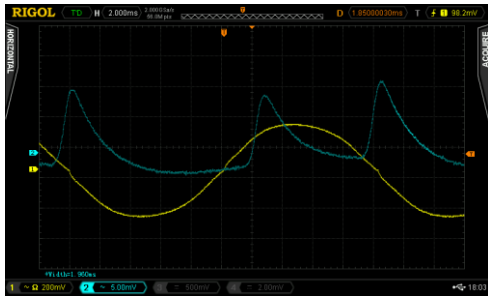
Picture 10: Autocorrelation signal without the same distance between the peaks
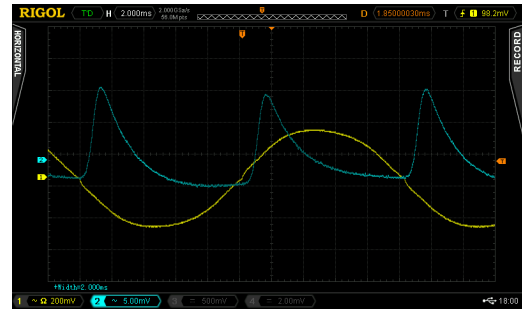
Picture 11: Autocorrelation signal with almost the same distance between the peaks.

11. Measurement: Start with one of the peaks at the beginning of the straight part. Change the distance of the fixed mirror step by step along this path. Take about 20 logic values (in around 5-10μm steps) by noting the moved distance and the delay between the two signals.

12. Note the width of your peaks.

2.2   Main experiment

You will work with Gallium Arsenide (GaAs) as sample. It's a semi-conductor with an easy band structure and widely used material since many years. So it is perfect for you to make friends with the experiment and all the new stuff.

You will do two different measurements one with pump-probe and one with transient grating.

First half:

1. Blocking one pump and one probe beam. With what a material shouldn't you block the beams? Reason: Look on the inside of the door!
2. Align the sample on the right position
3. Check and understand the paths of the beams from before the phase mask to the detection.
4. Which beam is the diffracted probe beam? Are there beams missing?
5. If yes, search in the room especially at every element for straying beams. Maybe something crosses the path of a beam at which it's getting scattered.
6. Let your supervisor check the setup
7. Align the photodiode to catch the diffracted probe beam
8. Take a measurement

Second half:

1. Open the blocked pump beam
2. Repeated aligning of the sample
3. Check and understand the paths of the beams
4. Align the photodiode
5. Repeated check by your supervisor
6. Take a measurement

If there is enough time it is possible to change some settings.

1. To change the intensity of the pump beams and taking an energy dependent measurement
2. To change the temperature of the sample by cooling down with the cryostat and taking a temperature dependent measurement

2.3   Analyzing tasks

1. Plot the values of the autocorrelation.
2. Calculate the pulse duration and compare it to a value from the literature.
3. Compare your ambipolar diffusion constant and the value for the electron lifetime with values from the literature.
4. Compare GaAs with Silicium and two other semiconductors. Which material is the better semiconductor? But which one is in use and why?
5. How can you crack the heating problems?
6. Problems and sources of error by the measurement

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