Dynamics of Femtosecond Laser Modification Inside Fused Silica

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Abstract

This paper discusses the dynamics of fs-laser modification in fused silica by tightly focusing (NA = 0.25) a 200 fs, 800 nm pump probe. Transmission spectroscopy is performed during a pump-probe experiment with the 800 nm (near infrared) laser and generated supercontinuum light filtered around roughly 660 nm (visible) and 1100 nm (infrared) as probe. In the visible and near infrared spectrum a consistent drop of 40-70% in transmission is seen when pump and probe are temporally overlapped. Within 1 ns after modification the transmission is recovered to its value before modification. In the infrared spectrum a drop in transmission is seen but the transmission plots show unexplainable behavior, probably due to the instability of the generated infrared light. Finally, a difference in time of arrival of different wavelengths is shown to be in the order of a few picoseconds which is confirmed by a ray-tracking model.

Contents

1 Introduction

In this paper the dynamics of fs-laser modification in fused silica are discussed which continues on the work of Dr. J.J. Witcher (1).

During the modification of glass (fused silica) by a focused femtosecond laser electrons in the glass are excited and essentially form a plasma. This plasma can absorb and reflect incoming laser light and the manner in which it does this is mainly dependent on the electron density and the electron collision time. With the use of a delayed pump-probe experiment the transmission of incident broad band light (the probe beam) is measured with a certain delay after the modification (caused by the pump beam). This will give information about the electron density (and thus the plasma frequency) and the electron collision time right after the modification within the range of a nanosecond.

Witcher stated that during the modification the electron density changes such that "[...] wavelengths below 750 nm are unaffected by the presence of the fs-laser generated plasma". This was partly confirmed by theoretical models.

The main goal of this research is to confirm or reject the results found by Dr. J.J. Witcher about the transmission of supercontinuum light through a laser created plasma.

Section 2 will briefly explain the theory behind the absorption and reflection of plasmas and will look at the models and results of Witcher.

Section 3 will first give a brief description of the pump probe experiment and then describe the experimental setup and routine. It will also explain the reason behind certain methods.

Section 4 will then show the results of the experiment where distinction is made between the 800 nm and the supercontinuum experiment. It also shows the effect of the difference in arrival of various wavelengths, confirmed by a theoretical model.

Finally, section 5 will give a summary of the results and compare those to Witcher's experiment.

2 Theory

By focusing a femtosecond laser inside a piece of glass electrons are excited due to multiple photon ionization (MPI) and avalanche ionization and essentially form a plasma in the glass (a "glassma" so you will). For more information about this process I would refer to the PhD thesis of Witcher. The plasma has a frequency of:

$$\omega_p^2 = \frac{N_e e^2}{\epsilon_0 m} \tag{2.1}$$

Here ω_p is the plasma frequency, N_e is the electron density of the excited electrons, e is the charge of an electron, ϵ is the permittivity of the free space, and m is the mass of an electron. The plasma frequency has a direct effect on the refractive index of the plasma through the following relation:

$$n_p^2 = n_g^2 - \frac{\omega_p^2}{\omega^2 - i\frac{\omega}{\tau}} \tag{2.2}$$

$$n_p^2 = (n_R + i n_I)^2 \tag{2.3}$$

Here n_p is the refractive index of the plasma and n_g that of the glass, ω_p the previous mentioned plasma frequency, ω the frequency of the light incident on the plasma and τ the electron collision time that is responsible for damping. The refractive index is used to calculate the reflection and absorption of light by the plasma. One can see that the refractive index has a real and an imaginary part. The imaginary part causes absorption while both parts contribute to the reflection (see equation 2.5). Here n_1 and n_2 are the refractive indices of medium 1 and medium 2, in this experiment glass and the plasma.

$$\alpha = \frac{2 n_I \omega}{c} \tag{2.4}$$

$$I(z) = I_0 e^{-\alpha z}$$

$$R = \frac{|n_1 - n_2|^2}{|n_1 + n_2|^2} \tag{2.5}$$

$$R = \frac{(n_g - n_R)^2 + n_I^2}{(n_g + n_R)^2 + n_I^2}$$

For measuring the transmission equation 2.4 and 2.5 can be used to establish a term for the transmission T (assuming a homogeneous plasma density):

$$T = (1 - R_1)(1 - R_2)e^{-\alpha L} \tag{2.6}$$

Here, R_1 and R_2 are the reflection coefficient for the front and back interface of the plasma with the glass, respectively and L is the thickness of the plasma. Witcher used this expression to model the effects on the transmission for various damping times, plasma thicknesses and plasma densities (figures 2.1 and 2.2).

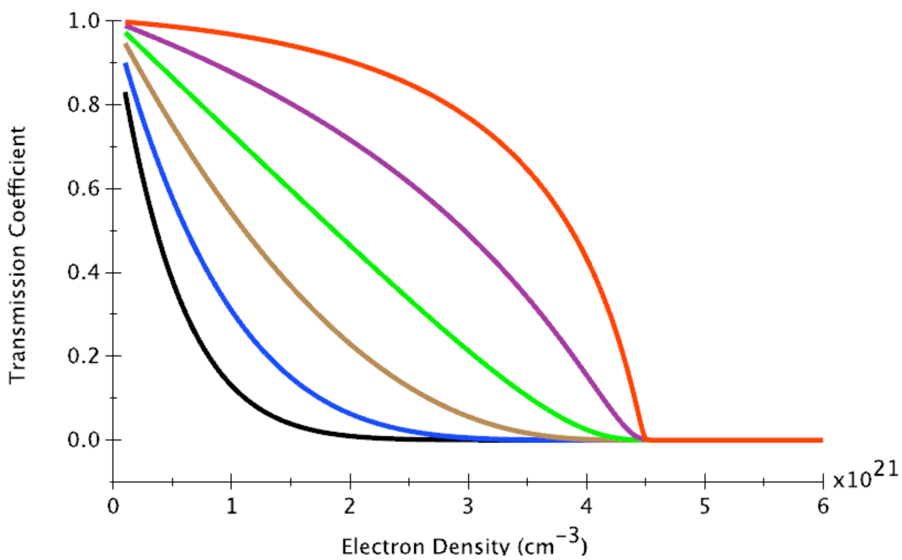

Figure 2.1: Transmission at 800 nm through a 2 μm thick, uniform density plasma as a function of electron density, Curves given for electron collision times of 1, 2, 4, 8, 20 and 100 fs shown in black, blue, brown, green, purple and red respectively ([1], figure 4.5)

The model suggests that, dependent on the electron density and collision time, certain wavelengths are unaffected by the plasma and will have a 100% transmission. Witcher confirmed this suggestion with a supercontinuum pump-probe experiment (see section 3) where he found that the plasma reaches a density of $4.0 \cdot 10^{21} cm^{-3}$ and doesn't affect wavelengths shorter than 750 nm (see figure 2.3). Unfortunately, he was unable the reproduce his result. In this paper the experiment to reproduce Witcher's results will be discussed.

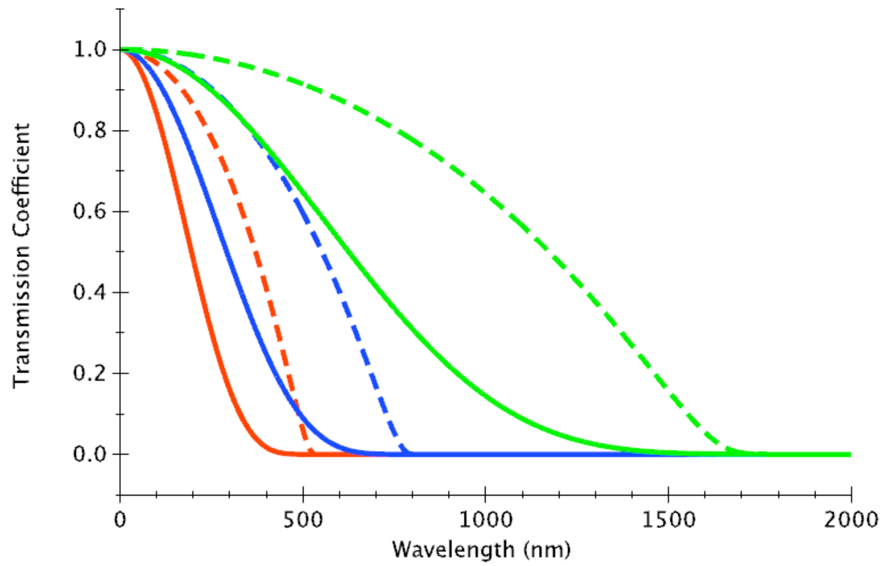

Figure 2.2: Transmission spectra for plasma densities of $1.0 \cdot 10^{21} cm^{-3}$ (green), $4.5 \cdot 10^{21} cm^{-3}$ (blue), and $10.0 \cdot 10^{21} cm^{-3}$ (red), Transmission calculated for electron collision time of 2 fs (solid curves) and 10 fs (dashed curves), All plasmas are 2 μm thick at uniform plasma densities ([1], figure 4.8)

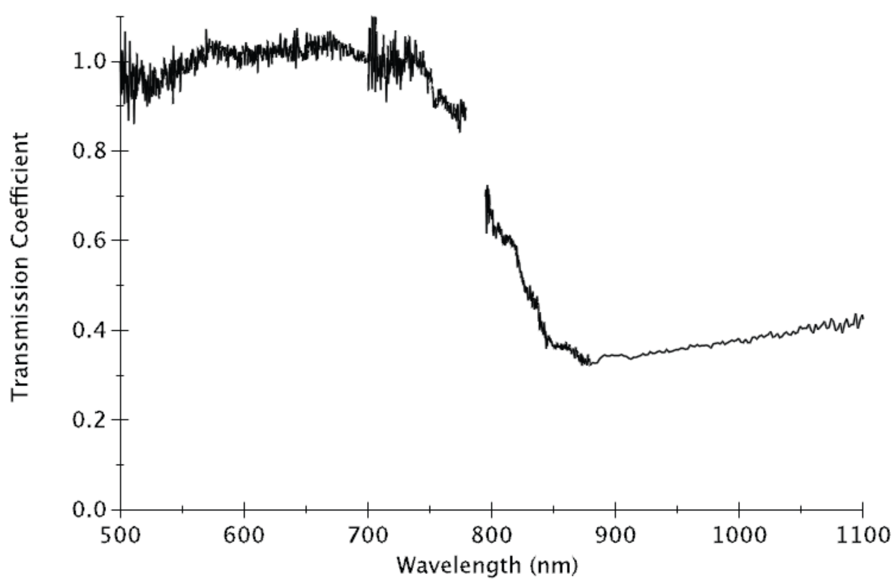

Figure 2.3: SC-probe transmission spectra through a plasma generated by a 4.0μJ, 200 fs, 800 nm pump pulse inside SiO_2, 118.0 ps after the pump pulse ([1], figure 4.8)

3 Experimental setup

In a pump probe experiment you make use of two different pulses arriving within a known delay from each other. The pump pulse is responsible for exciting (and in this experiment case even modifying) the sample and the probe pulse, arriving at the same time or with a certain delay, is used for spectroscopy to extract information about the sample at a known time after modification. In this experiment two different probe lights are used, namely 800 nm light (similar as the source) and supercontinuum light (spectrum of light from visible to infrared), which is generated by focusing the 800 nm probe through a sapphire crystal [(2)]. The pump is always 800 nm light. When in this paper the "800 nm" experiment or the "supercontinuum" experiment is mentioned, it alludes to the different probes used in the experiment.

The main difference between this setup and that of Witcher is the collinear style. In Witcher's setup the pump and probe arm are going into the sample orthogonally to each other, in this setup the arms are overlapped before they're sent collinearly into the sample. The big advantage of this method is that once the arms are overlapped both arms will, depending on the focusing objective, focus to the same spot. In Witcher's setup the focal spots of the different arms had to be aligned which required a large accuracy from the operator. However, even in the collinear setup the alignment requires careful alignment as minor deviations will become larger when focused inside the sample. Furthermore, though a achromatic doublet is used to focus the different wavelengths in the supercontinuum light, there will still be a difference in arrival time between the different rays of wavelengths (see Section 4.3).

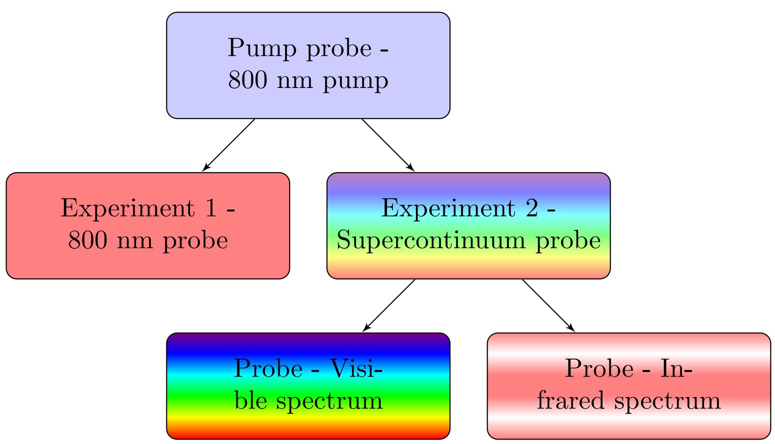

Figure 3.1: Structure of the different experiments

As there are two fairly distinguished experiments, their setups are thus somewhat different. Furthermore, within the supercontinuum experiment two different spectral ranges will be looked at, namely the visible and the infrared (see fig 3.1). This made it necessary to slightly change the setup for these experiments as well. However, a major part of the setup is used in both experiments and is therefore explained first, after which the specific differences will be discussed in more detail.

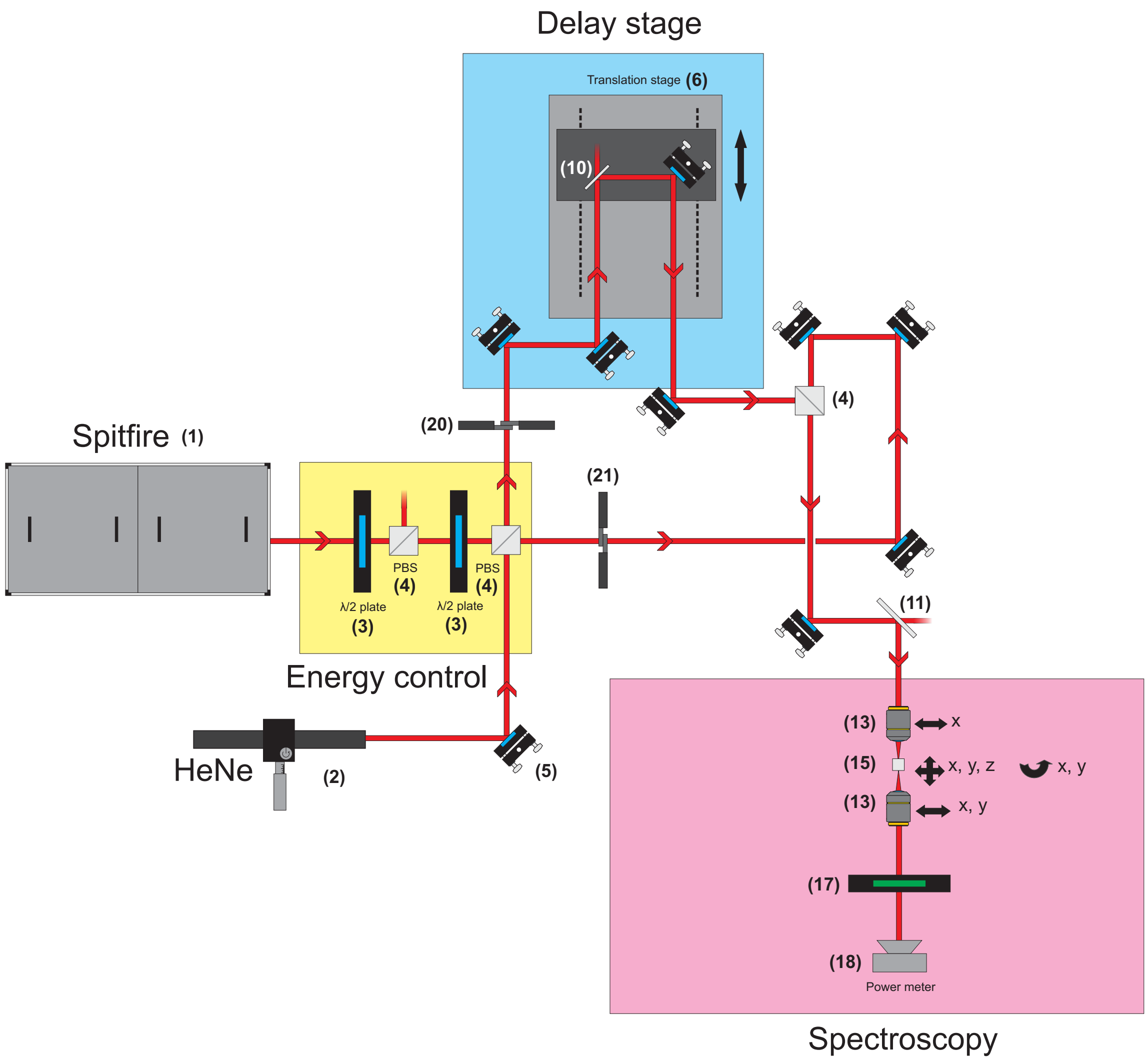

Figure 3.2: Setup for the 800 nm experiment. Numbered components are identified in list 3.1.

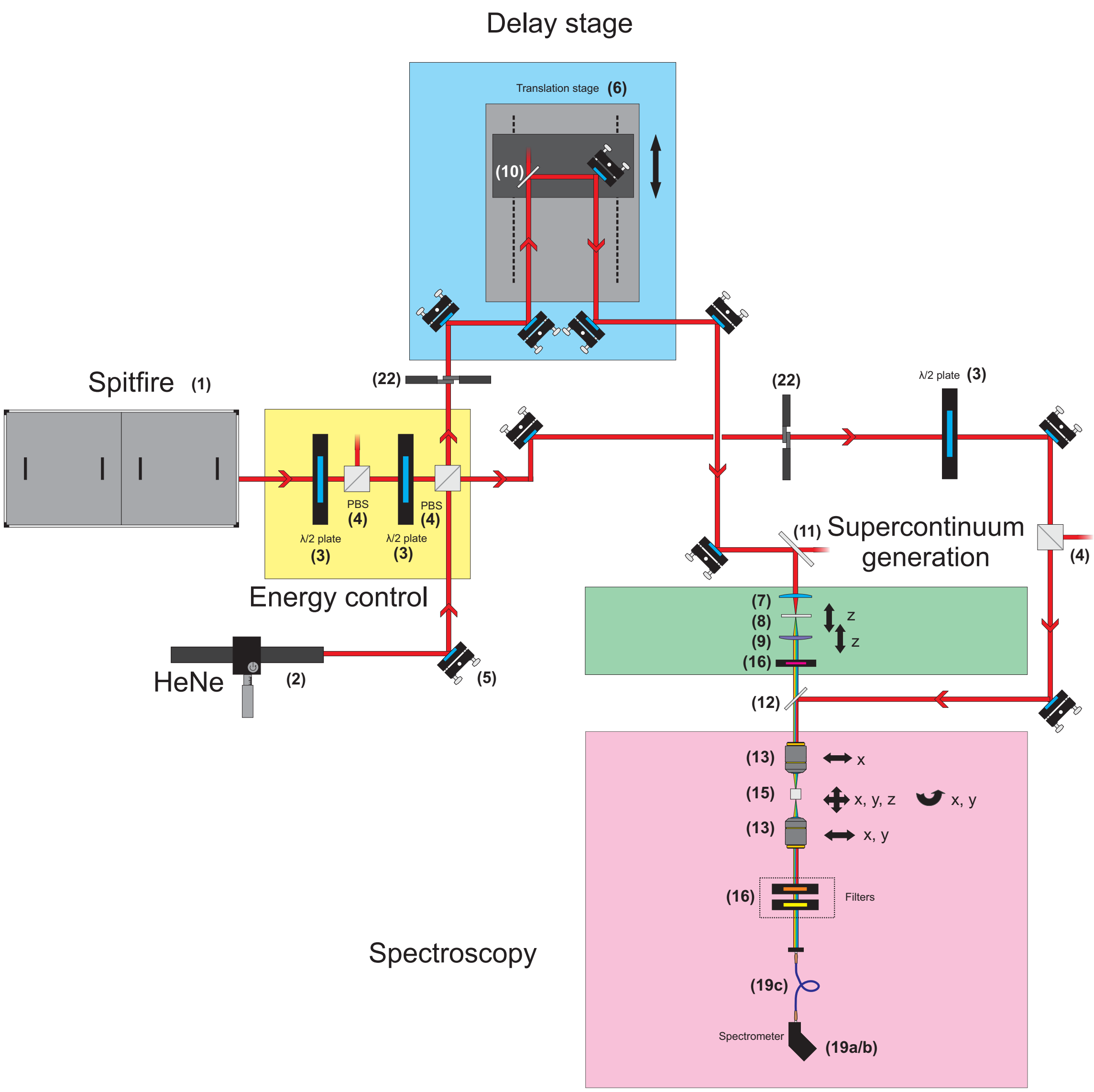

Figure 3.3: Setup for the supercontinuum experiment. Numbered components are identified in list 3.1.

List 3.1: Items used in the setup

1. Spectra-Physics Titanium:Sapphire regeneratively amplified laser system. System generates a 200 fs, 800 nm pulse with an output energy of 500 μJ and a repetition rate of 1 kHz. The entire system consists of 4 Spectra-Physics laser components: Millenia, Tsunami, Merlin, and Spitfire LCX.
2. Spectra Physics 633 nm Helium Neon (HeNe) CW laser. Used to align the setup with visibile light. The laser is overlapped with Spitfire by sending it through the same of irises as Spitfire.
3. 2x Newport 10RP02-46 800 nm zero-order half-wave plates to rotate the polarization of the fs-laser.
4. 3x COHERENT 44-4521-000 12.7 mm polarizing cube beamsplitters. This is used to split Spitfire into a probe and pump arm and in the 800 nm experiment also used to combine both arms again. It is also used in conjunction with the half-wave plate to modulate energy by transmitting P polarized light and reflecting S polarized light at 45 $^{\circ}$.
5. Several Newport 10D20ER2 broadband silver mirrors. Mirrors are mounted in a Newport flip-mount to precisely move mirrors into and out of the beam path.
6. Aerotech PRO-115 150 mm translation stage with SoloistCP controller to vary the probe pulse path length (to delay with respect to the pump).
7. Plano-convexlens (f = 100 mm), LA1509-B. Used to focus probe pulse into the sapphire plate. Lens is mounted on a translation stage allowing for adjustment of focal position.
8. Sapphire window from Edmund Optics. 25.4 mm diameter and 0.5 mm thickness. Used as a medium to create a broadband pulse via SCG. The sapphire plate is mounted to a translation stage in the z-direction that allows the sapphire plate to be moved relative to lenses 7 and 9.
9. Thorlabs AC254-030-B achromatic doublet (f = 30mm). Used to collimate the SC broadband probe. The lens is mounted to a translation stage to allow for the optimal collimation the incident beam.
10. Thorlabs BSX11, 10:90 Beamsplitter, 25.4 mm diameter, 5 mm thickness. Used to split some of the light of the HeNe laser to check for alignment in a CCD camera on the translation stage.
11. Thorlabs BSW17, 50:50 Beamsplitter, 50.8 mm diameter, 5 mm thickness. Used to split some of the light of the HeNe laser to check for alignment in a CCD camera.
12. Thorlabs EBS1, 50:50 Beamsplitter, 25.4 mm. Used to combine the probe and pump arm in the supercontinuum experiment.

13. Tasco 10x (0.25 NA) microscope objectives (f = 20 mm). Used to focus both the 800 nm pump beam and the 800 nm/SC probe beam into the sample. A second objective is used to collect both laser beams from the sample.
14. Newport translation stages. Several stages are used to accurately place the optics, especially the sample and the objectives around the sample. The focus objective (13) can thus be moved in the x-direction, the collecting objective (13)in both the x- and z-direction and the sample (15) itself can move in x-, y-, and z- direction and can also tilt around the x- and y-axis (Newport 562 5-axis). In addition, the stage was placed on two stacked 850G Newport stepper motor stages attached with Newport 850G motors and controlled by a Newport 3 axis controller. Furthermore, during the supercontinuum experiment the achromatic doublet (9) is placed on a stage in the x-direction and the optical fiber (19c) connected to the spectrometer in the x- and y-direction.
15. Glass sample. 10 mm x 10 mm x 5 mm 7980 Corning standard grade fused silica. All six surfaces polished.
16. Filters used in the supercontinuum experiment to control the intensities of both the beams and filter out unnecessary light:
    (a) Semrock NF03-808E-25, 808 nm Stopline single-notch filter. Placed after collecting the new generated supercontinuum light and used to filter out the intense 800 nm probe beam. The filter is tilted, changing the incident angle of the beam, to tune the rejection wavelengths and rejection bandwidth of the filter to the desired values.
    (b) Thorlabs FES750, 750 nm short pass filter. Placed after the sample and used to filter out the intense 800 pump light, keeping the probe beam with a spectrum range of 750 nm and shorter for visible spectroscopy.
    (c) Thorlabs FEL1050, 1050 nm long pass filter. Placed after collecting the new generated supercontinuum light and used to filter out the intense 800 nm probe beam, keeping a spectrum range of 1050 nm and longer for infrared spectroscopy.
    (d) Thorlabs FEL850, 850 nm long pass filter. Placed after the sample and used to filter out the intense 800 pump light, keeping the probe beam with a spectrum range of 850 nm and longer for infrared spectroscopy.
    (e) Thorslabs Neutral Density filters with optical densities of 0.1, 0.2, 0.3, 0.6, 1.0. Used to control the intensity of both beams.
17. 2x Newport polarizers: 740-860 nm, 12.7 mm diameter. Used after the sample to filter out the pump beam in the 800 nm experiment.
18. OPHIR Nova II power meter (0.2s response time). Used to measure the power of both the pump and probe beam, which can be calculated

to the energy of the pulse with a frequency of 1 kHz. Also used for the power transmission through the sample in the 800 nm experiment.

19. Spectrometer:
    (a) Hamamatsu TG-NIR (near-infrared) USB spectrometer to collect the near-infrared portion broadband probe spectrum.
    (b) Ocean Optics USB2000 visible spectrometer to measure the Spitfire spectrum as well as the visible portion of the broadband probe spectrum.
    (c) Ocean Optics P-600-2-VIS-NIR optical fiber, 2 m long with 600 11m fiber core and 0.22 NA, for the coupling of light into each spectrometer.
20. 3x Sony XC-77 CCD camera (black and white image collection)
21. Thorlabs SH05 shutter (12.7 mm) with SC10 controller to control exposure of the probe pulse.
22. Uniblitz shutter (25.4 mm) with a VMM-Dl driver to control exposure of the pump pulse.
23. Beam block

The setup (figure 3.2 and 3.3 and table 3.1) obviously contains the 800 nm femtosecond (fs) laser (1) which immediately is separated by a polarizing beam splitting cube (4) into two pulses, the pump arm and the probe arm. The pump arm is (before it rejoins with the probe arm) only affected by a second polarizing beam splitter, which in combination with the half-wave plate (3) is used to regulate the energy of the arm.

The probe arm however is first guided over a translation stage (6) to be able to vary the path length of the probe arm which is used to delay the probe with respect to the pump. After that the beam either directly rejoins with the pump (800 nm experiment) or is first focused on and collected from a sapphire plate (8) with two lenses (7 and 9) to create the supercontinuum light (supercontinuum experiment). The latter will also go through a set of filters (16) to control the energy of the probe in order to prevent it from modifying the glass. Both arms and their beams will be discussed in more detail in the subsections below.

When one works with with a collinear setup the probe and pump arm have to be combined again before going into the glass. Both in the 800 nm experiment as in the supercontinuum experiment this is done with a 50:50 beamsplitter, though slightly different splitters are used in each experiment (4 and 12 respectively). When combined the beams will be focused into the glass with a 10x microscope objective (13) with an NA of 0.25 mounted on a translation stage with movement in the x-direction.

The glass (10 x 10 x 5 mm) itself is double-taped on a bar which is roughly 20 cm long, 10 mm wide and 5 mm thick and can thus fully support the sample.

The bar is in turn bolted on an elaborate platform (14) that has a variety in the directions of movement. It can move in the x-, y- and z-directions and tilled around the x- and y-axis. Furthermore, it has two translation stage on the bottom in the y- and z-direction which can be automatically operated. The manual stages are used to get the sample properly in place and the automatic stages are used to get fresh material during the experiment.

Both focusing and collecting the light into and from the glass is done with the same type of microscope objective (13). The mount of the objective can be tilted over the x- and y-axis and is also placed on two translation stages, giving it movement in the xy-plane. From the collecting objective the light is then either send to a polarizer and power meter (800 nm experiment, 18) or several filters and an optical fiber (19c) connected to a spectrometer (supercontinuum experiment) for visible (19b) and infrared light (19a).

The problem with the Spitfire laser is that it has a wavelength of 800 nm and is therefore not visible for the naked eye. Furthermore, the energy of the pulses of Spitfire can irreversible damage the eye when it has direct contact with it. To make the alignment somewhat easier a Helium-Neon laser (HeNe, (2)) is overlapped with Spitfire to be able to align the setup with visible and much less energetic light. The HeNe light is merged into the experiment with a polarizing cube beamsplitter and is aligned to two irises the Spitfire goes through as well by adjusting two mirrors *before* the beamsplitter, otherwise changing the path of Spitfire as well.

3.1 800 nm - 800 nm

In the 800 nm experiment the pump and probe beams are almost identical, except for their energy and polarization. As only the probe beam is desired for spectroscopy after collecting it from the sample, the pump arm is then filtered out by using a 800 nm polarizer (6). Though the polarizer will not filter out the pump arm completely a second polarizer has little more effect but does lower the absolute power of the probe significantly (see table 3.1). Therefore the use of only one polarizer is adequate.

After the polarizers the light is collected by the power meter (18) which can be set to automatically measure the power over a range of time, ranging from a few seconds to a few minutes. Its response time is 0.2 seconds.

3.2 Supercontinuum

The setup for the supercontinuum experiment is slightly more complex. This is mainly due to the generation of the supercontinuum light and the filtering to make it suitable for the sample and the spectrometers. First, let's focus on the generation of supercontinuum light which is done by focusing on a sapphire

Table 3.1: Overview of the beam powers with different number of polarizers. Note that the powers with a † are measured with a filter on the power meter. The percentage is calculated relative to the pump and probe beam combined.

Beams present	Power with # of polarizers					
	0 (μW)		1 (μW)		2 (μW)	
Probe	68	(3%)	42	(93%)	39	(95%)
Pump	$1.87 \cdot 10^3$†	(97%)	2.7	(7%)	2.0	(5%)
Both	$1.93 \cdot 10^3$†	(100%)	45	(100%)	41	(100%)

A second polarizer has little more effect on diminishing the pump arm with respect to the probe arm but still reduces the power of the probe.

plate. Previous research showed that the best results can be achieved without damaging the plate by using a lens with a numerical aperture (NA) of around 0.05 [(2)]. The numerical aperture is defined by:

$$NA = n \sin\theta = \approx n\frac{D}{2f} \approx \frac{D}{2f} \tag{3.1}$$

Here, n is the refractive index of air, θ the angle of focusing, D the diameter of the laser beam at the lens and f the focal length of the lens. The last approximation is made due to the fact that the lens is used in air ($n = 1$). As the laser beam is roughly 10 mm wide, one would want a lens with a focal length of 100 cm (7). Secondly, the supercontinuum light has to be collected and be made completely collinear. However, the refractive index of a lens is different for each wavelength, which means that every color is bent differently. To make this difference as small as possible, an achromatic doublet is used to collect the light which minimizes the effect of different bending. In section 4.3 the differences in angle and final position are calculated when using an achromatic doublet in the experiment.

Even though supercontinuum light is created with the use of the sapphire plate, there is still a significant amount of 800 nm light left, enough to modify the glass sample. This light is minimized by using an 808nm filter (visible spectrum, 16a)) or an 1050 long pass filter (infrared spectrum, (16c)). After the sample the pump light is obsolete and is filtered out by a 750 short pass filter (16b) or an 850 long pass filter (16d), respectively for the visible and infrared spectrum. Finally, the light is focused into a fiber (19c) connected to a spectrometer for either visible (19b) or infrared (19a) light.

3.3 Alignment

Perhaps the most difficult aspect of the pump probe experiment is the alignment of both beams. As both beams have to focus on the same spot which is in the micrometer scale, the beams have to be precisely overlapped before going through the microscope objectives. Furthermore, the use of a delay stage requires the probe beam to come in and go out parallel to the stage.

The main tool that is used for the alignment is a 633 nm Helium-Neon (HeNe) laser as this gives a visible light beam in contrast to the 800 nm (infrared) Spitfire. The HeNe is aligned to Spitfire by sending it through a same pair of irises early in the setup. Important in this process is that this is done by adjusting the mirrors that only affect HeNe. Once the HeNe is overlapped with Spitfire it can be used it to make sure it's properly aligned in the setup.

Firstly, it is essential that the probe beam is going parallel into and out of the stage. Therefore a CCD camera is placed on or after the stage and see if the incoming or outgoing, respectively, beam changes position if one moves the delay stage. The beam is parallel when the beam on the CCD camera is stationary when moving the delay stage.

Secondly, when the pump and probe beam are overlapped as close as visibly possible, a test measurement is done to see if the required drop in transmission is seen when moving the delay stage. If this spot is found, the last mirrors of the pump beam right before the sample are tuned to maximize the drop. The reason why this is done with the pump beam and not the probe is that the probe beam is more difficult to adjust due to the several optical elements in the beam. Once the alignment is set accordingly the different measurements can be acquired.

3.4 Measurements

To calculate the transmission of the probe beam affected by the pump beam one needs to know at least four datasets:

A Background

B Probe transmission only

C Pump transmission only

D Pump + probe transmission

With the above sets the transmission can be calculated as:

$$
\begin{aligned}
T =& \frac{(D-A)-(C-A)}{B-A} \qquad (3.2)\\
=& \frac{D-C}{B-A} \text{ (If the background is constant throughout the experiment)}
\end{aligned}
$$

Ideally, one would want to require A, B, C and D for every time delay before moving to the next spot. However, in the 800 nm experiment the computer automation for the beam blocks had not yet been implemented which made it necessary to measure a certain data set for the entire time delay. To make sure the data sets didn't change during the experiments, the following routine was used:

1. **Background:** Transmission with both pump and probe turned off
2. **Probe while traveling:** Transmission of only the probe while moving the delay stage (to see if the beam is still parallel to the stage)
3. **Probe while stationary:** Transmission of only the probe without moving the delay stage.
4. **Pump while stationary:** Transmission of only the pump without moving the delay stage.
5. **Pump + probe while traveling:** Transmission of pump and probe while moving the delay stage (the actual experiment!)
6. **Repeat 3-5:** Repeat step 3-5 twice to get **three** independent experiments
7. **Repeat 3-4:** Repeat step 3-4 to check if the pump and probe transmission is still stable
8. **Repeat 2:** Repeat step 2 to check if the probe beam is till parallel to the stage

For the supercontinuum experiment the beam blocks were computer automated the beam blocks so all the data could be required for each time delay. To optimize the number of time delay steps (and thus the speed of the experiment) the step size on the delay was lower around the drop. This results in a higher time resolution around the drop in comparison to the approach and the recovery after the drop.

Finally, to make sure that the sample is not damaged, the sample is translated in the x-direction, perpendicular to the laser beam, with a speed of 10 μm/s. With a spot size in the range of $\sim$10 μm, this means that each spot receives about 1000 pulses.

4 Results

As with the rest of the paper the results can be divided into those for the 800 nm experiment and the supercontinuum experiment. Both experiments will show pump-probe transmission plots as a function of the delay time. The transmission is plotted relative to the transmission right before the drop. Furthermore, during the experiment the power of the pulses are measured with a power meter (18). In order to convert this to fluence the following relations are used:

$$E = P * t = \frac{P}{f} \tag{4.1}$$

$$d = \frac{2.44\lambda}{2NA} \tag{4.2}$$

$$F = I/(\pi(d/2)^2) \tag{4.3}$$

Here, E is the energy of a pulse in Joule, P the power in Watt, f the frequency in Hertz, d the diameter of the focal spot and λ the wavelength.

4.1 800 nm experiment

As described in the previous sections this experiment only uses one wavelength for probing (800 nm). Figures 4.1, 4.2 and 4.3 show the transmission with a pump power of 5.0, 8.4 and 12.5 J/cm^2, respectively. Each experiment has been done thrice.

It can clearly be seen that each experiment shows a clear drop in transmission of 20-45%. After the drop the transmission gradually increases again until it's to back to 100%. The recovery takes about 800 ps. Furthermore, for every pump fluence, it is shown that the rate of recovery decreases around 400 ps and increases again at 600 ps. It's uncertain if this is caused by a process inside the plasma or if it's caused by the setup. Also, in some experiment the recovery goes above the 100%. This is probably caused by an absolute increase of the probe fluence. One could argue that the changes in slope described before could also be caused by this increase in probe fluence, but the slope change can also be seen in the experiment with 8.4 J/cm^2 pump fluence. In that experiment however, the recovery stays at 100% and thus the increase of the probe fluence is unlikely. Furthermore, an increase in the absolute probe fluence would not explain the *decrease* in the rate of the recovery at 400 ps.

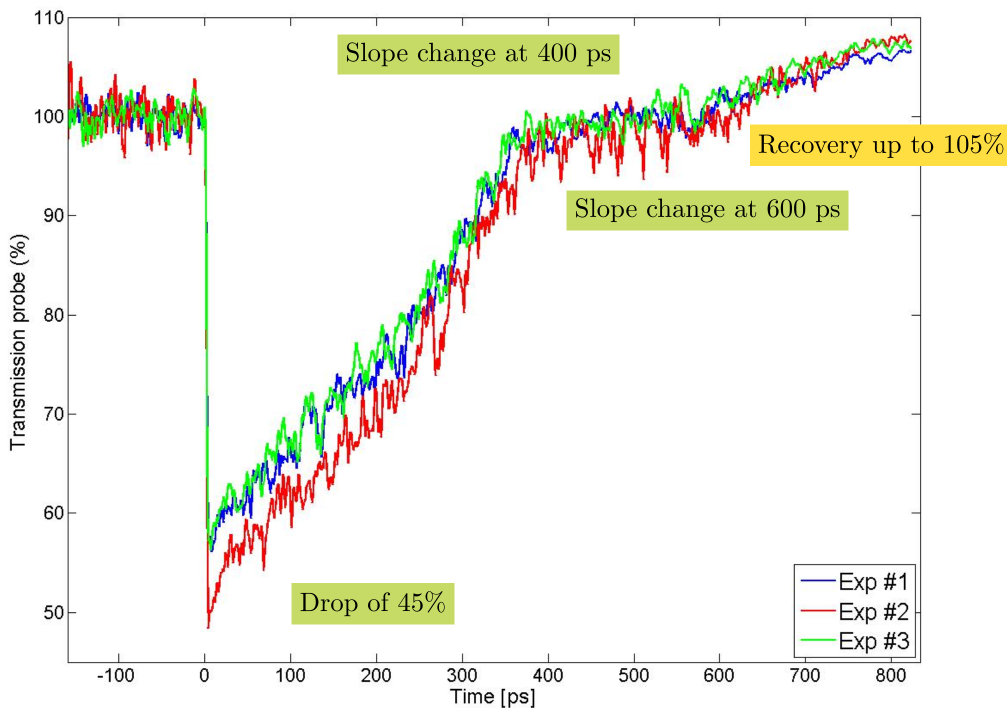

Figure 4.1: Three independent experiments of transmission spectroscopy of a pulse probe of 0.75 J/cm^2 through a plasma in fused silica generated with a pump pulse of 5.0 J/cm^2. Both probe and pump are 799 nm, 200 fs pulses focused with an NA of 0.25.

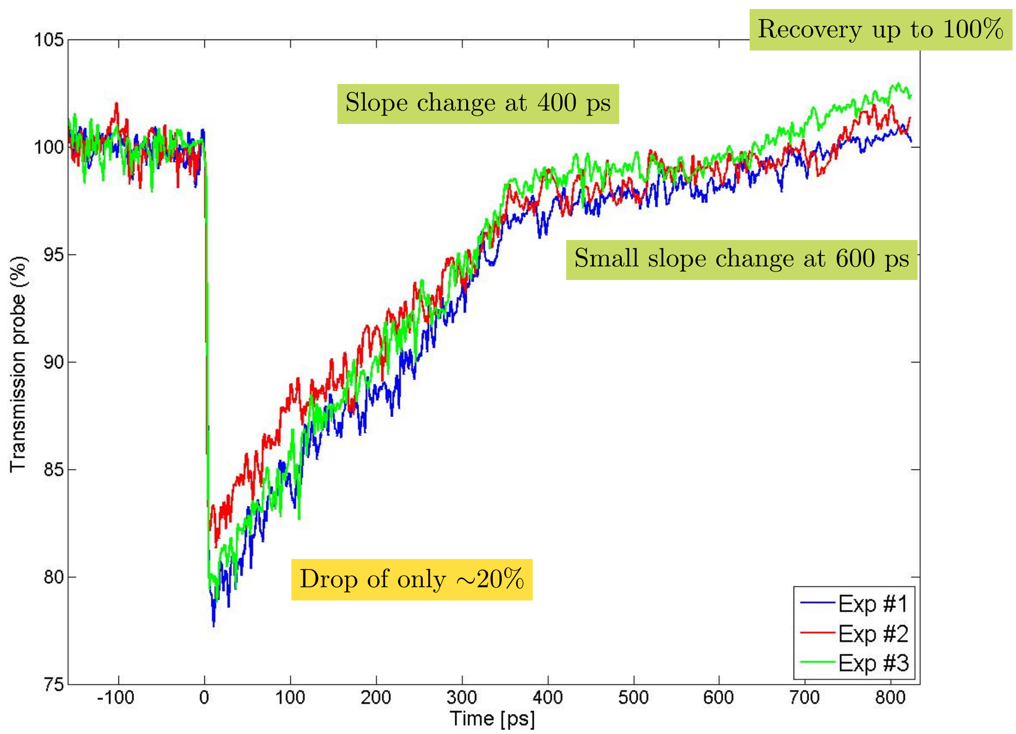

Figure 4.2: Three independent experiments of transmission spectroscopy of a pulse probe of 0.79 J/cm^2 through a plasma in fused silica generated with a pump pulse of 8.3 J/cm^2. Both probe and pump are 799 nm, 200 fs pulses focused with an NA of 0.25.

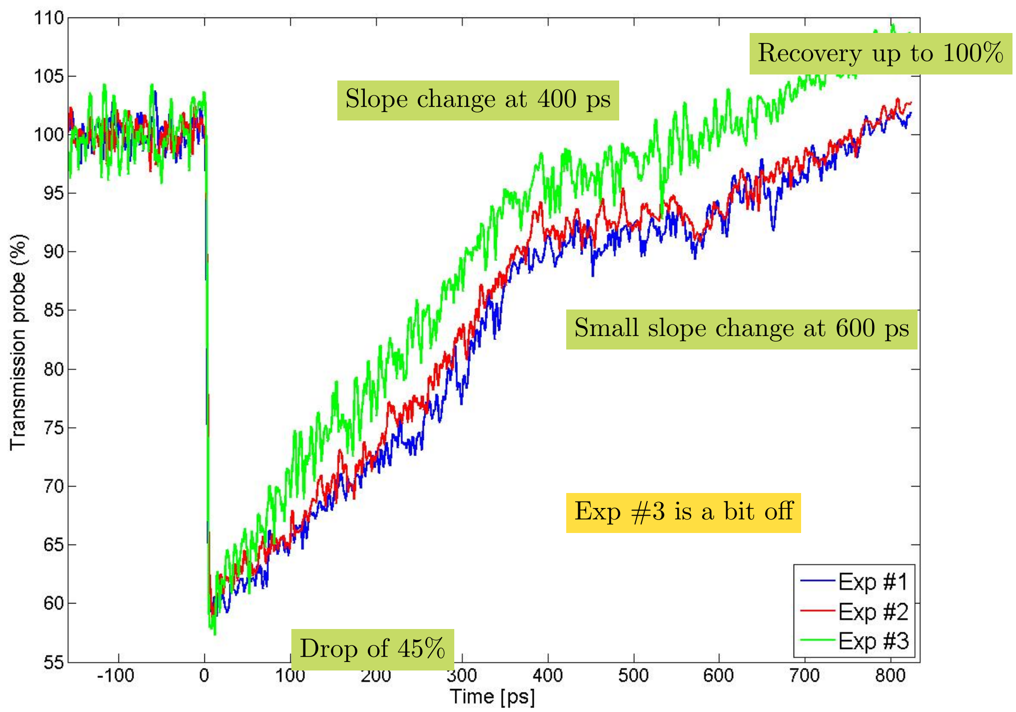

Figure 4.3: Three independent experiments of transmission spectroscopy of a pulse probe of 0.75 J/cm^2 through a plasma in fused silica generated with a pump pulse of 16.7 J/cm^2. Both probe and pump are 799 nm, 200 fs pulses focused with an NA of 0.25.

4.2 Supercontinuum

In this experiment supercontinuum light was used created by focusing 800 nm light in a sapphire plate as described in section 3. The results are divided in two subsections: visible light and infrared light. This has two reasons: first, the experiment requires a spectrometer which is either a visible or an IR one. Second, as the infrared light is not as strong as the visible light, for the infrared light the integration time and probe power has to be increased to get a strong enough signal and thus the visible light has to be blocked to prevent saturation of the spectrometer.

Another important notice is the fact that the created supercontinuum light is instable in the infrared. This means that the results are harder to analyse and that it's hard to contribute certain effects to actual processes in the plasma or just fluctuations in the supercontinuum light.

Finally, the results below show independent experiments where a small band of wavelengths are measured. The graphs show both the mean of this band as the extremes. In section 4.3 it will be shown that different colors have a slightly different time of arrival but this is in the order of a picosecond. This is smaller than the resolution of the experiment which means that taking the mean is acceptable.

4.2.1 Visible light

Experiments are done with the visible light for different pump fluences. Figures 4.4, 4.5, 4.6 and 4.7 show the transmission with a pump fluence of 8.4, 12.5, 15.9 and 20.9 J/cm^2, respectively. The supercontinuum light was created with a probe power of 2.0 mW and filtered for the range of around 660 nm. As the 800 nm light from the probe is filtered and the power of the unfiltered supercontinuum light is quite low, the probe will not modify the sample. The different color plots show the transmission for different wavelengths. The legend shows the wavelengths in nm.

Though the supercontinuum light might be unstable a significant drop in transmission of 55-75% can be seen in each experiment. Furthermore, as in the 800 nm experiment a clear recovery within 800 ps is shown. Though the resolution in time delay is less than in the 800 nm experiment, it's clear that the recovery process is somewhat different than with the 800 nm probe. For the duration of $\pm$200 ps the recovery rate is quite low after which it suddenly sharply increases. Around 350-400 ps this sharp slope ends and the recovery then gradually increases to 100% transmission. It can further be noticed that the recovery is a bit different for different wavelengths, though the wavelength difference is only 20 nm.

The experiment with 8.4 J/cm^2 (fig. 4.4) shows a large, unknown slope before

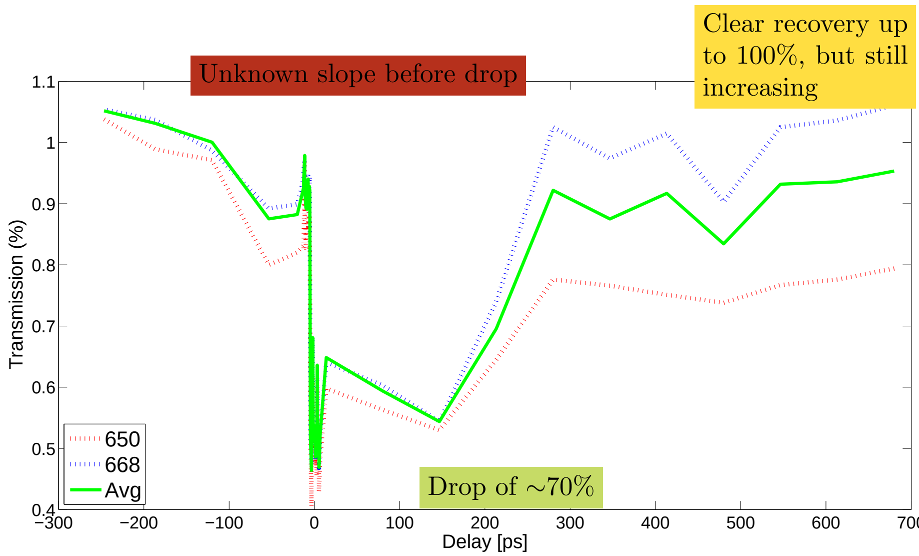

Figure 4.4: Experiment of transmission spectroscopy of a 660 nm pulse probe through a plasma in fused silica generated with a 800 nm pump pulse of 8.3 J/cm^2. Both probe and pump are 200 fs pulses focused with an NA of 0.25. Integration time of spectrometer is 250 ms.

the drop where one would expect a constant transmission. After the drop we see a similar effect where the transmission increases above the 100%. These effects don't coincide at all with the nice, proper drop of 70% in transmission and the, to the eye, gradual increase in transmission.

12.5 J/cm^2 pump fluence (fig. 4.5) also show a slope before the drop (of ±60%), though this one is small. The recovery on the other hand is rather different than the rest of the experiments in visible light. Except for the strange peak right after the drop (which can be a measuring error) all the wavelengths have a recovery within 300 ps after which they hardly increase at all. The shorter wavelengths don't even reach the 100%.

The 15.9 J/cm^2 pump fluence experiment (fig. 4.6) shows a nice transmission profile. It has again a slope before the drop but this is rather small. There is a drop of 55% which is very good and like the rest it has a 200 ps period where the recovery hasn't started yet. In the end however, it still has a significant slope when it reaches 100% and it's interesting to know if it would increase even further. Unfortunately, the delay stage didn't allow it to create an even larger time delay.

Finally, the 20.9 J/cm^2 pump experiment (4.7) shows similarities with the 12.5 J/cm^2 experiment. It also has s small slope before the drop but moreover, it also recovers within 300 ps after which the transmission hardly increases. Shorter wavelengths don't reach the 100% transmission.

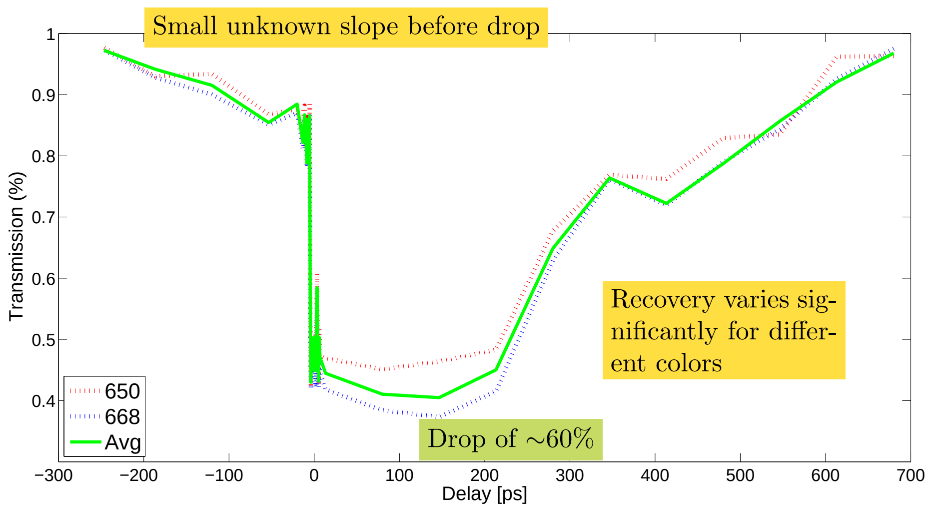

Figure 4.5: Experiment of transmission spectroscopy of a 660 nm pulse probe through a plasma in fused silica generated with a 800 nm pump pulse of 12.5 J/cm^2. Both probe and pump are 200 fs pulses focused with an NA of 0.25. Integration time of spectrometer is 250 ms.

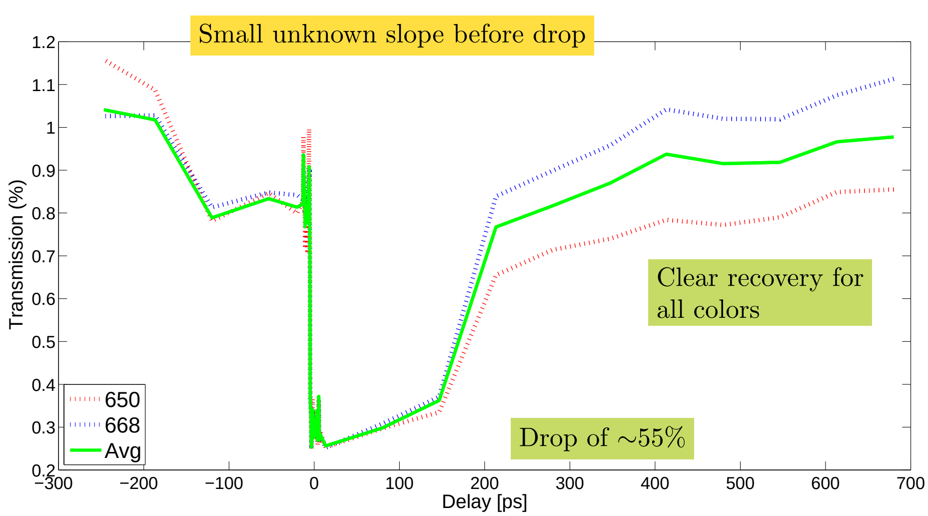

Figure 4.6: Experiment of transmission spectroscopy of a 660 nm pulse probe through a plasma in fused silica generated with a 800 nm pump pulse of 15.8 J/cm^2. Both probe and pump are 200 fs pulses focused with an NA of 0.25. Integration time of spectrometer is 250 ms.

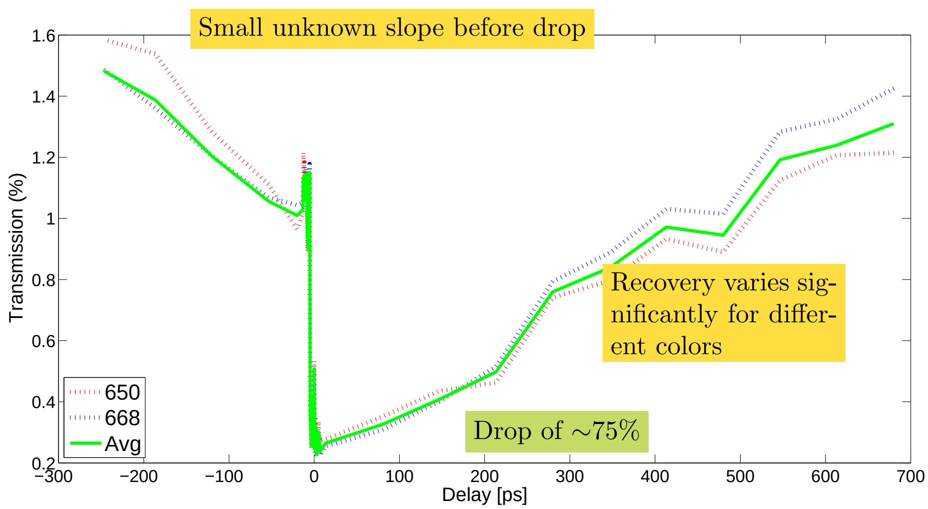

Figure 4.7: Experiment of transmission spectroscopy of a 660 nm pulse probe through a plasma in fused silica generated with a 800 nm pump pulse of 20.9 J/cm^2. Both probe and pump are 200 fs pulses focused with an NA of 0.25. Integration time of spectrometer is 250 ms.

4.2.2 IR light

Basically, the experiments in the infrared light show no proper result to confirm the absorption or reflection of the probe light. A few of the graphs below show a drop, but hardly any of them show a gradual recovery. Moreover, all the graphs show an unexplainable peak right before the drop (going above 100% transmission) and even more important, the supercontinuum light is so unstable that it's difficult to distinguish any true changes in transmission between the fluctuations in the transmission. When look at the raw data there is no clear explanation for the shown behavior. The pump and probe beam combined sometimes show a higher intensity than the sum of the separate pump and probe beam (causing the +100% transmission). This effect can be due to a nonlinear effect during the combination of the beams or simply to errors during the measurements.

Pump fluence: 36.8 J/cm^2

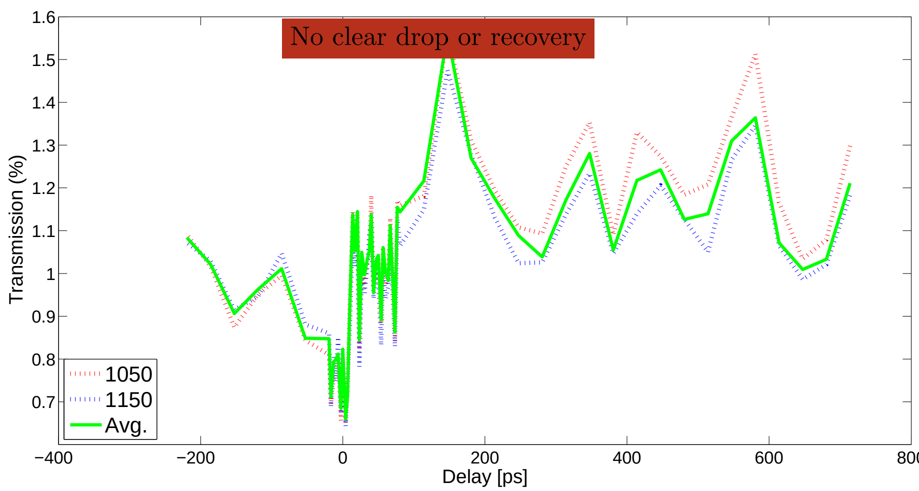

Figure 4.8: Experiment no. 1 of transmission spectroscopy of a 1100 nm pulse probe through a plasma in fused silica generated with a 800 nm pump pulse of 36.8 J/cm^2. Both probe and pump are 200 fs pulses focused with an NA of 0.25. Integration time of spectrometer is 50 ms.

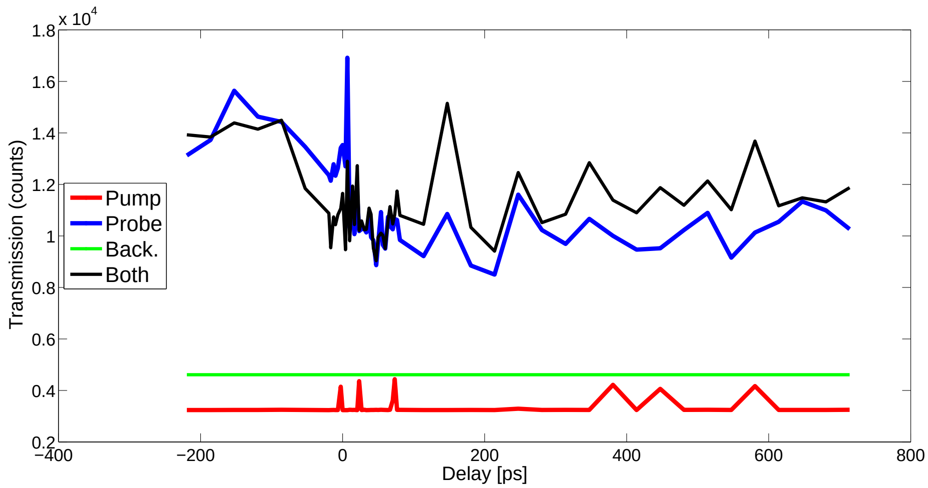

Figure 4.9: Raw data of experiment no. 1 (see figure 4.8). Shown are the transmission plots with just the background (green), only the pump (red), only the probe (blue) and with both the pump and probe (black).

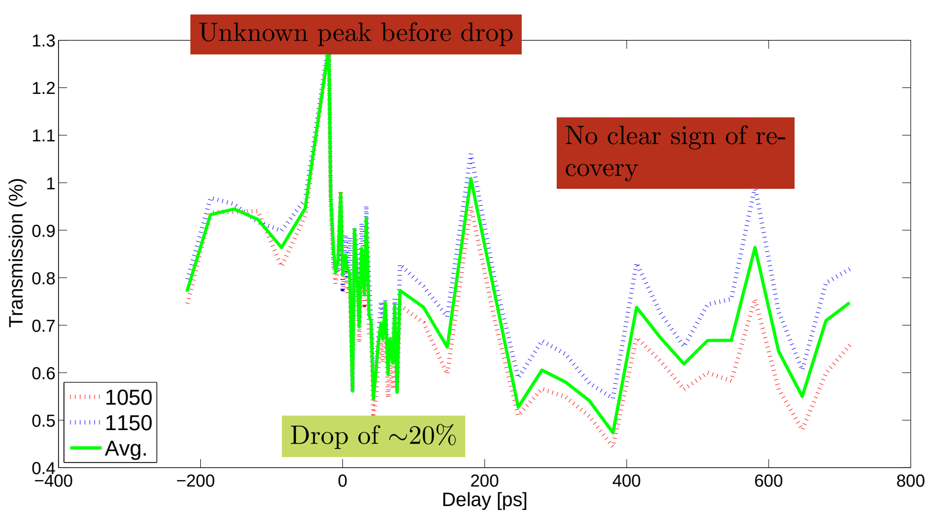

Figure 4.10: Experiment no. 2 of transmission spectroscopy of a 1100 nm pulse probe through a plasma in fused silica generated with a 800 nm pump pulse of 36.8 J/cm^2. Both probe and pump are 200 fs pulses focused with an NA of 0.25. Integration time of spectrometer is 50 ms.

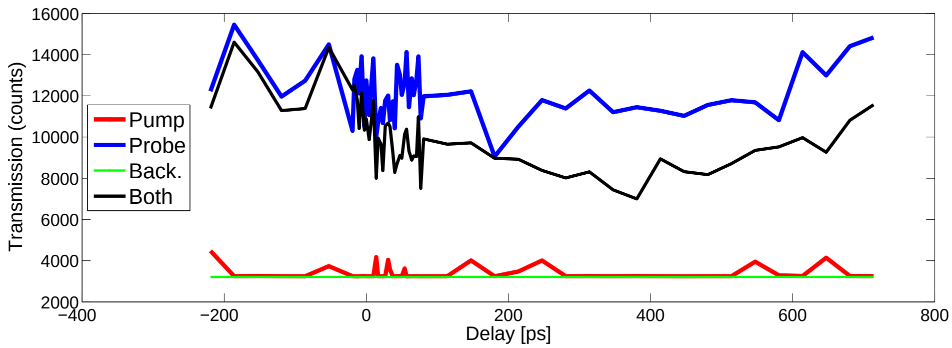

Figure 4.11: Raw data of experiment no. 2 (see figure 4.10). Shown are the transmission plots with just the background (green), only the pump (red), only the probe (blue) and with both the pump and probe (black).

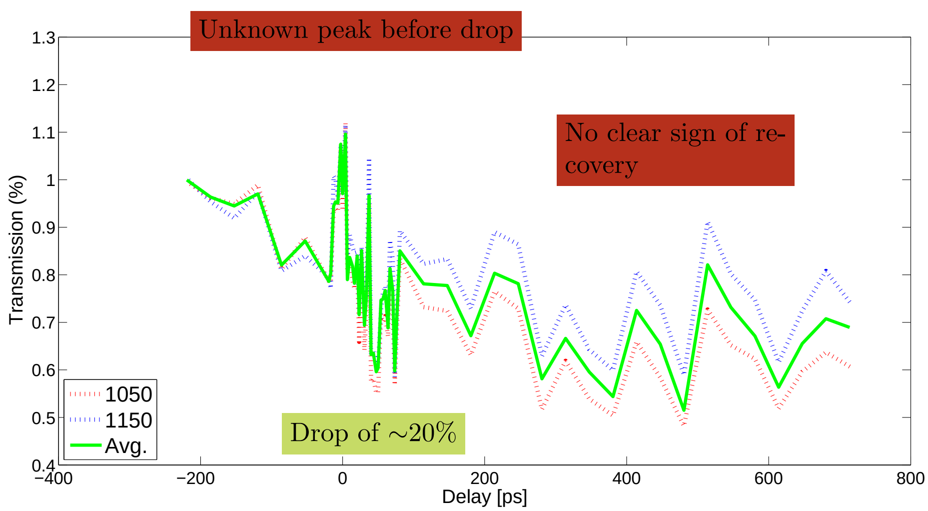

Figure 4.12: Experiment no. 3 of transmission spectroscopy of a 1100 nm pulse probe through a plasma in fused silica generated with a 800 nm pump pulse of 36.8 J/cm^2. Both probe and pump are 200 fs pulses focused with an NA of 0.25. Integration time of spectrometer is 50 ms.

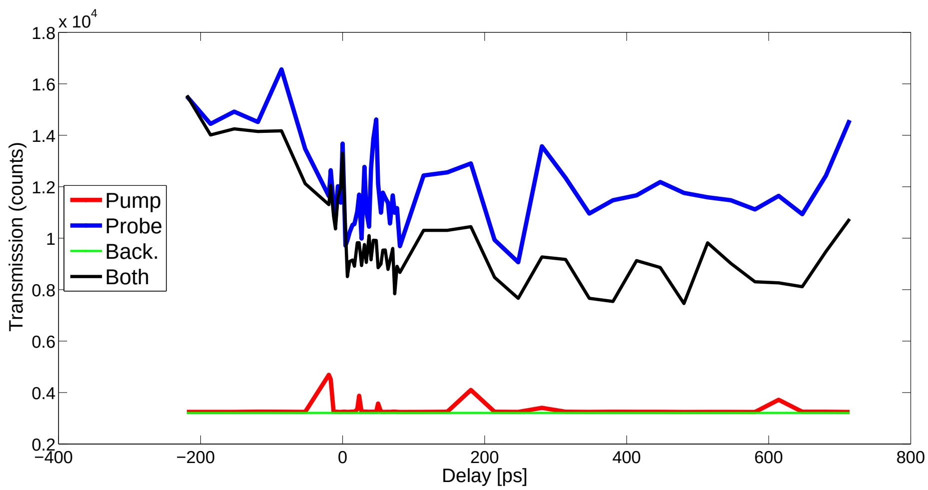

Figure 4.13: Raw data of experiment no. 3 (see figure 4.12). Shown are the transmission plots with just the background (green), only the pump (red), only the probe (blue) and with both the pump and probe (black).

Pump fluence: 37.6 J/cm^2

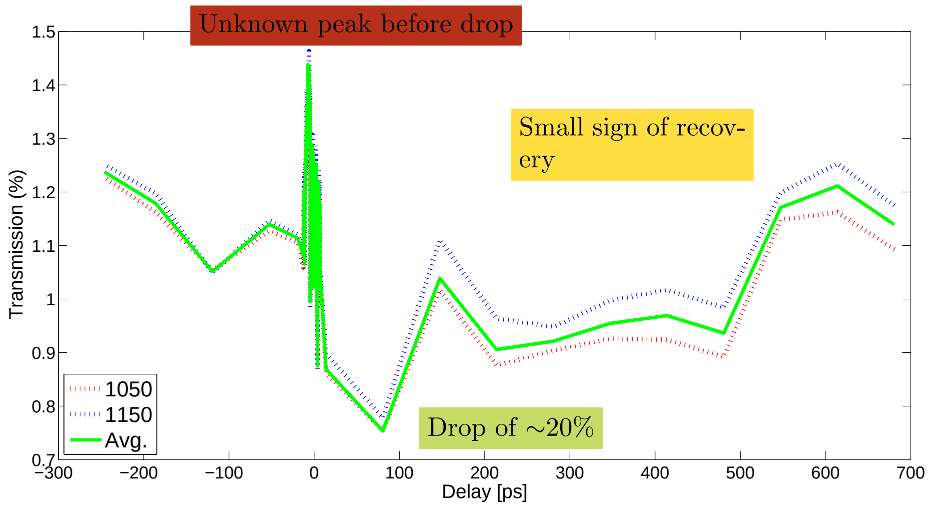

Figure 4.14: Experiment no. 1 of transmission spectroscopy of a 1100 nm pulse probe through a plasma in fused silica generated with a 800 nm pump pulse of 37.6 J/cm^2. Both probe and pump are 200 fs pulses focused with an NA of 0.25. Integration time of spectrometer is 150 ms.

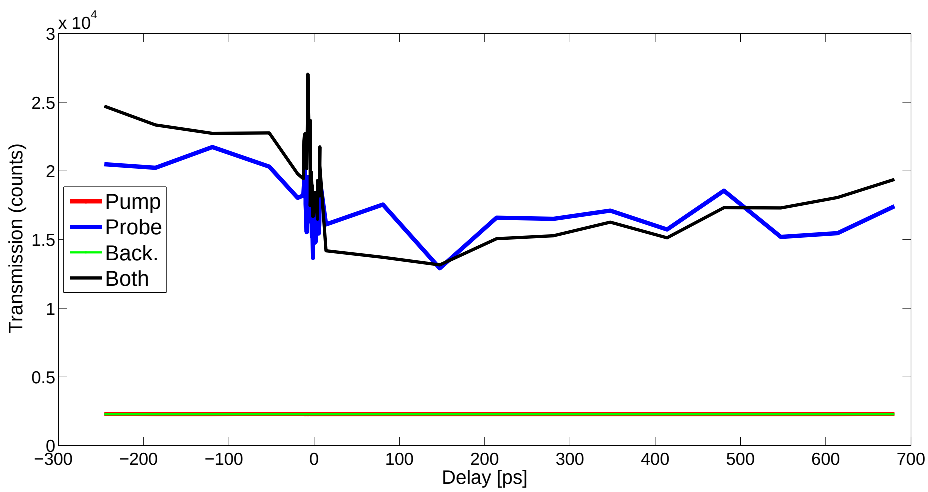

Figure 4.15: Raw data of experiment no. 1 (see figure 4.14). Shown are the transmission plots with just the background (green), only the pump (red), only the probe (blue) and with both the pump and probe (black).

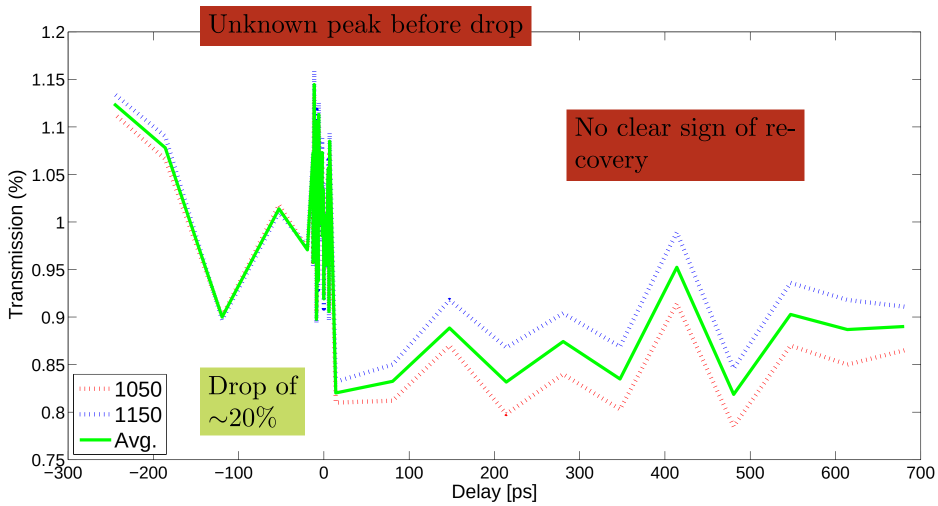

Figure 4.16: Experiment no. 2 of transmission spectroscopy of a 1100 nm pulse probe through a plasma in fused silica generated with a 800 nm pump pulse of 37.6 J/cm^2. Both probe and pump are 200 fs pulses focused with an NA of 0.25. Integration time of spectrometer is 150 ms.

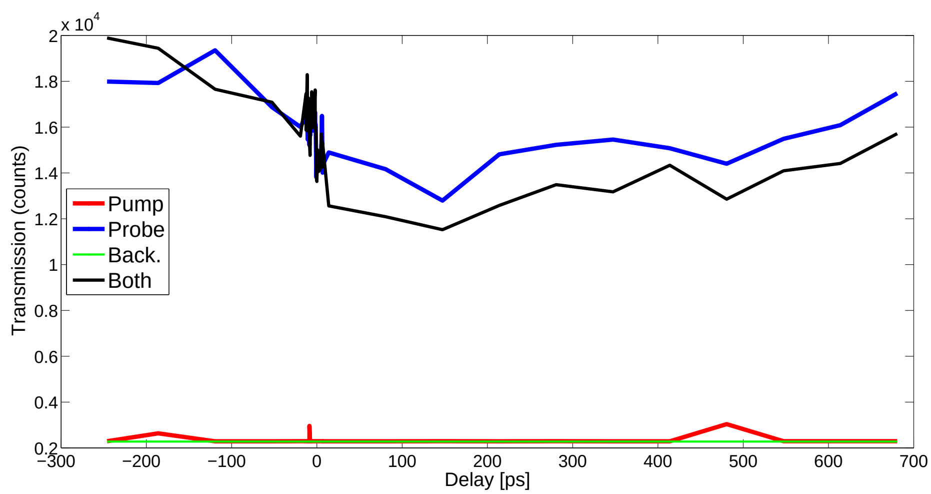

Figure 4.17: Raw data of experiment no. 2 (see figure 4.16). Shown are the transmission plots with just the background (green), only the pump (red), only the probe (blue) and with both the pump and probe (black).

4.3 Arrival of difference wavelengths

Though the achromatic doublet collects the generated supercontinuum light and makes it collinear, there will always be a difference in arrival time for the different wavelengths. This is due to a different optical path length caused by two reasons.

Firstly, the refractive index of a lens is dependent of the wavelength of the incident light which means that every wavelength is refracted differently. Secondly, the doublet collects light within a certain 'cone' from the sapphire plate and thus rays of the light hit the lens at different heights under different angles.

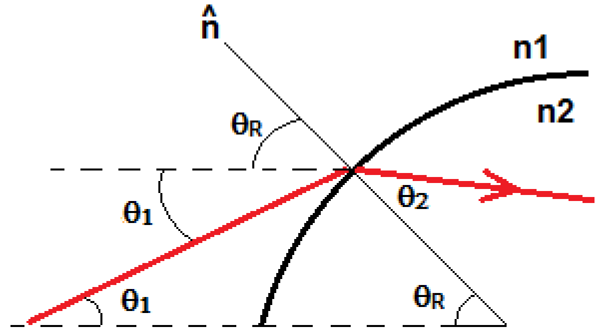

Figure 4.18: Ray based model for calcuating the trajectory time for different wavelengths

If one assumes that the light can be considered as different rays going through the lens, the trajectory of a ray and the time it will take to get to the focus point can be calculated. In this model a lens is considered as a medium with two interfaces (front and back) at which the light is refracted. Within the medium the light continues its trajectory (see figure 4.18). The achromatic doublet is modeled as a lens with two different materials in between three interfaces. The radii of curvature, the distances between interfaces and the types of materials of the doublet are known (see table 4.1). In the model the sample is included as well as the focusing lens which is considered to be perfect (see figure 4.19). The formula to calculate the refracted angle is at each interface:

$$\theta_2 = \arcsin\left(\sin(\theta_1 + \theta_R)\frac{n1(\lambda)}{n2(\lambda)}\right) - \theta_R \tag{4.4}$$

Here, θ refers to the angles, which are relative to the horizontal axis and n refers to the refractive indices of the media. For the λ-dependence of n the database of SCHOTT is used [(3)]. In equation 4.4 Snell's law $\frac{\sin\theta_1}{\sin\theta_2} = \frac{n_2}{n_1}$ is used with the addition that the horizontal axis is rotated with respect to θ_R which is the angle

Achr. doublet medium 1	N-SF6HT
Achr. doublet medium 2	N-BAF10
Sample	Fused silica
Radius (interface 1)	79.1 mm
Radius (interface 2)	16.2 mm
Radius (interface 3)	-21.1 mm
Distance 1-2	1.5 mm
Distance 2-3	12.0 mm

Table 4.1: Lens specifications for the model

between the normal axis of the curve and the horizontal axis.

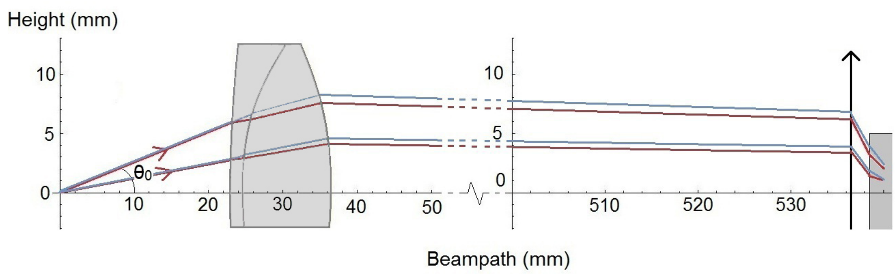

Figure 4.19: Setup used in the calculations of the model

In the model the experiment is simulated resulting in a setup with an achromatic doublet, containing three interfaces, a microscopic objective and finally the sample (see figure 4.19). Unfortunately, the microscopic could not precisely be simulated as the necessary specifications were not known. The lens is therefore treated as a perfect thin lens.

From the results of the model (see figure 4.20 and 4.21) one can see that the difference in arrival time is in the order of a few picoseconds, dependent of the wavelength and starting angle difference. To check this with experimental results the time step of the translation stage is set to less than a picosecond and focused on the range around the transmission drop (i.e. where the pump and probe arrive at the same time).

From the experiment we find an arrival difference of roughly 4 ps between 500 and 700 nm which is in agreement with our model. However, it has to be noted that the model is simplified and assumes light is behaving as rays which is not necessarily the case.

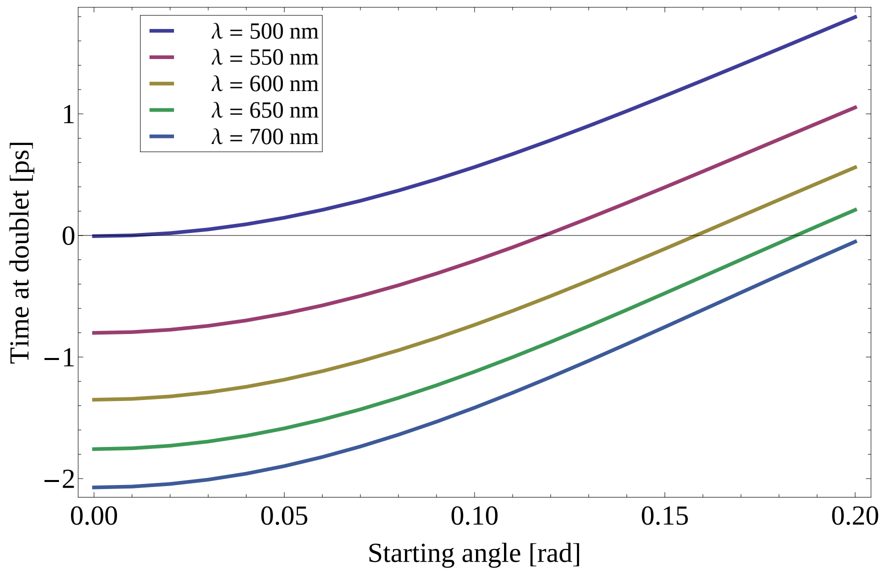

Figure 4.20: Calculated time of arrival dependent on the starting angle for different wavelength. The time is with respect to a ray of 500 nm with a zero rad starting angle.

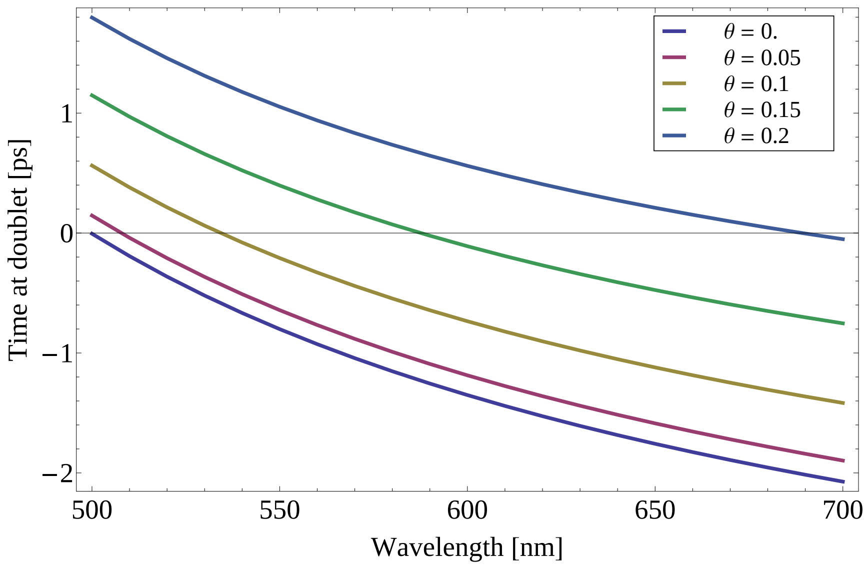

Figure 4.21: Calculated time of arrival dependent on the wavelength with different starting angles. The time is with respect to a ray of 500 nm with a zero rad starting angle.

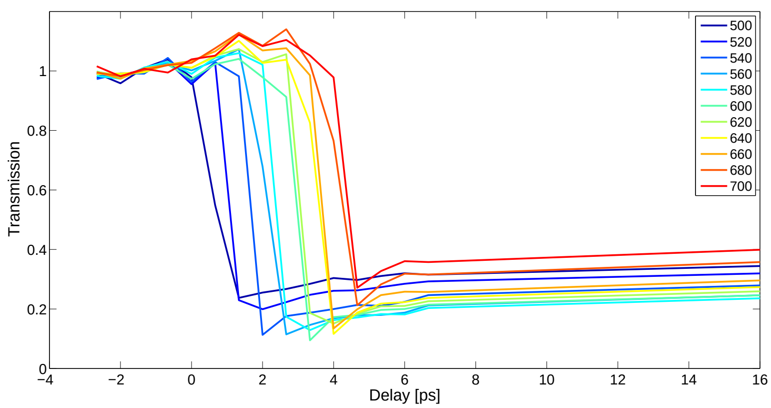

Figure 4.22: Measured time of arrival dependent on the wavelength with respect to the arrival time of light of 500 nm. Measured during an experiment of transmission spectroscopy of a 660 nm pulse probe through a plasma in fused silica generated with a 800 nm pump pulse of 8.3 J/cm^2. Both probe and pump are 200 fs pulses focused with an NA of 0.25. Integration time of spectrometer is 250 ms.

5 Conclusion

The goal of this paper was to study the dynamics of fs-laser modification of fused silica in a collinear setup.

Firstly, a proper drop in transmission was seen with probe light of 800 nm confirming that the excited electrons absorb and reflect a significant portion of the incident light. It could also be confirmed that the recovery occurred within a nanosecond. However, the manner of the recovery was somewhat different. Where in Witcher's experiment the recovery was smooth and gradually reaching it's former transmission, the results in this experiment show some abrupt slope changes.

Secondly, Witcher [(1)] suggested that wavelengths shorter than 750 nm are unaffected by the generated plasma. However, a clear drop in transmission was seen for 660 nm light which contradicts Witcher's statement. Unfortunately, the dynamics of the plasma recovery show an unexplainable behavior which questions the reliability of the results. Moreover, no proper transmission drop was seen for the infrared light due to the instability of the supercontinuum light.

Furthermore, when comparing the transmission profiles for different pump powers for both the 800 nm as the 660 nm probe, no clear relation between pump power and the drop in transmission could be seen. This has mainly to do with the sensible alignment of the experiment and its large influence on the drop.

Finally, the effect of the difference in arrival of various wavelength was measured and confirmed this with a ray-tracking model.

References

[1] J. Witcher, *Dynamics of Femtosecond Laser Modification Inside Fused Silica.* PhD thesis, University of California, Davis, 2011.

[2] J. B. Ashcom, R. R. Gattass, C. B. Schaffer, and E. Mazur, "Numerical aperture dependence of damage and supercontinuum generation from femtosecond laser pulses in bulk fused silica," *Journal of Optical Society of America*, vol. 23, no. 11, pp. 2317–2322, 2006.

[3] SCHOTT, "http://refractiveindex.info/."