Laser-plasma interaction in droplet-targets

Dissertation

zur Erlangung des akademischen Grades
doctor rerum naturalium (Dr. rer. nat.)

vorgelegt dem Rat der Physikalisch-Astronomischen Fakultät
der Friedrich-Schiller-Universität Jena

von Stefan Düsterer,
Master of Arts (University of Texas, Austin)
geboren am 9.1.1974 in Dinkelsbühl
Gutachter:

1. **Prof. Dr. R. Sauerbrey**  
   Institut für Optik und Quantenelektronik  
   Friedrich-Schiller-Universität Jena

2. **Prof. Dr. O. Willi**  
   Institut für Laser- und Plasmaphysik  
   Heinrich-Heine-Universität Düsseldorf

3. **PD Dr. P. Gibbon**  
   Central Institute for Applied Mathematics  
   Forschungszentrum Jülich


Tag der öffentlichen Verteidigung: 3. Dezember 2002
## Contents

1 Introduction 1

2 Experimental setup 6
   2.1 The laser system ........................................ 6
   2.2 The setup for the extreme ultraviolet (EUV) measurements ........... 6
      2.2.1 The vacuum chamber .................................... 7
      2.2.2 Characterization of the water droplets .................... 8
   2.3 EUV radiation detection ................................... 12
      2.3.1 The EUV spectrometer .................................. 13
      2.3.2 The pinhole spectrometer .............................. 18
      2.3.3 The EUV monochromator ............................... 18

3 Theoretical background 20
   3.1 Basic plasma physics ..................................... 20
   3.2 Laser-plasma interaction ................................ 22
      3.2.1 Plasma expansion ..................................... 23
      3.2.2 Collisional absorption mechanisms ...................... 24
      3.2.3 Collisionless absorption mechanisms .................... 29
      3.2.4 Laser-plasma interactions at ultra high intensities ........ 31
      3.2.5 MEDUSA - simulation of laser-plasma dynamics .......... 32
   3.3 Atomic physics in plasmas ............................... 33
      3.3.1 Thermodynamic equilibria ............................ 33
      3.3.2 Rate equations ...................................... 36
      3.3.3 The collisional radiative steady state code .......... 37
      3.3.4 Time-dependent rate equation model ................... 38
      3.3.5 Transition probabilities of $O^{5+}$ lines ................ 40
      3.3.6 Radiation transport .................................. 42
      3.3.7 Line broadening .................................... 43
4 Results of the EUV experiments
4.1 Acquiring EUV spectra ................................
4.2 Experimental test of the CRSS code .........................
4.3 The mass-limited droplet target ............................
  4.3.1 Dependence of the conversion efficiency (CE) on the laser pulse energy ..
  4.3.2 CE dependence on the laser pulse duration .................
  4.3.3 Droplet expansion and laser transmission ..................
  4.3.4 Prepulse dependence of the EUV CE ....................
  4.3.5 Dependence of the EUV CE on the prepulse energy ..........
  4.3.6 Measurement of x-rays ................................
4.4 Solid extended targets ................................
  4.4.1 The ice target ................................
  4.4.2 EUV CE of an extended quartz target ....................
  4.4.3 Preplasma dynamics ................................
4.5 Ion velocities ........................................
4.6 Characterization of the EUV source .........................
  4.6.1 Pulse duration of the EUV emission .....................
  4.6.2 Determination of the EUV source size ...................
  4.6.3 Angular distribution of the EUV emission ...............
5 Simulation of the EUV measurements .....................
  5.1 Simulation of the electron density and the ion temperature ............
  5.2 Laser energy deposition and EUV CE ........................
  5.3 Simulated EUV pulse duration ............................
  5.4 Comparison of the simulated and the measured EUV CE ..........
6 Ultra-high intensity measurements ....................
  6.1 Laser-triggered fusion ................................
    6.1.1 Introduction ................................
    6.1.2 Energy distribution of fusion neutrons ................
  6.2 Laser-triggered fission ................................
7 Conclusion .........................................
A Appendix: Description of the laser system

A.1 The oscillator - the front end

A.2 Laser pulse expansion and compression

A.3 Amplification stages

A.4 The prepulse unit

A.5 The characterization of laser pulses ranging from fs to ns

A.5.1 80 fs to 10 ps pulses

A.5.2 120 ps pulses

A.5.3 6 ns pulses

A.6 Focusing the laser beam

A.6.1 The focus

A.6.2 Determination of the intensity
1 Introduction

Talking of ‘light-matter interaction’, most people, even physicists, think of sunburn and solar cells. However, an intense laser pulse can heat up matter not just to some hundred degrees Kelvin, but rather up to millions of Kelvin, producing soft x-ray emitting plasma. By focusing the laser beam even tighter, temperatures of thousands of Kelvin can be achieved, triggering nuclear reactions, like fusion or photo-fission of nuclei. The work presented here will deal with the description and characterization of such a laser, with the efficient production of (very) soft x-rays, namely extreme ultraviolet (EUV) radiation, and laser-induced nuclear reactions.

The Jena TeraWatt (TW) laser system [Ziener, 2001] provides a peak power of 12 TW ($12 \times 10^{12}$ W) exceeding the power provided by a conventional power plant ($\sim 10^9$ W) by orders of magnitude. However, the light power of 12 TW lasts only for a tiny fraction of a second, in contrast to the power plant which operates continuously. A modest amount of laser energy, in the range of 1 J, is compressed into a very short period of time of only 80 femtoseconds ($80 \times 10^{-15}$ s). Electrons can move at most a few $\mu$m within 80 fs, while ions are essentially immobile during such a short time. This extremely powerful laser is one of only a few lasers reaching such powers world-wide. As one can imagine, important questions in basic physics, concerning for example relativistic plasma physics [Düsterer et al., 2001b, Düsterer et al., 2001c, Schwoerer et al., 2001] or melting dynamics of matter [Feurer et al., 2001], can be answered by conducting experiments with such a laser. However, the dominant part of this thesis is of great interest for industrial applications. Why is basic physics research, performed with a TW laser of commercial interest?

The reason is simple. The smallest structures on a computer chip have to become smaller and smaller in order to keep up with the persistent demand for faster computers. The only cost-effective way to produce millions of transistors on one single computer chip is lithography. However, the structure sizes are ultimately limited to about half the wavelength of the light used for the lithographic production steps. Thus, the currently used excimer laser technology, with wavelengths of 248 nm and 193 nm, can only be scaled down to structure sizes, or more precisely structure periodicities, of 90 nm. However, structures of 65 nm periodicities are demanded by the semiconductor roadmap, the guideline for the progress of the computer industry within the next 15 years for the year 2007 [ITRS, 2001]$.^1$ Only lithography using a shorter - much shorter - wavelength can assure the progress of the computer industry. Today, it is not possible to produce efficient lasers having such short wavelengths. As stated above, intense laser pulses are capable of heating matter to enormously high temperatures. Matter, or rather plasma, at such temperatures emits a significant amount of radiation at wavelengths considerably shorter than 100 nm. This fact finally answers the

$^1$A structure periodicity of 65 nm might be produced with not jet industrially applied F$_2$-excimer lasers working at 157 nm or some new technologies. However, it will not be possible to produce periodicities of only 45 nm, demanded for 2009, with excimer laser technology.
question why laser-plasma interaction is an interesting topic for the industry. By optimizing the laser parameters, the emission of short wavelength radiation can be significantly enhanced and in turn, a much smaller and more cost effective laser tailored for the specific application can yield the same amount of short wavelength radiation as a large, expensive and un-optimized laser system [Düsterer et al., 2001a]. Thus, understanding the relevant laser-plasma interaction processes can be important for future lithography technology.

From the point of lithography, basically every wavelength that is smaller than 40 nm could have been used [Bjorkholm, 1998]. The wavelength region of interest between 5 nm and 40 nm is the range of extreme ultraviolet radiation (EUV). However, imaging optics has to be constructed. Lenses were quickly ruled out since EUV radiation is absorbed in all known materials within micrometers. On the other hand, efficient mirrors with a reflectivity better than 60% can only be produced for 11 nm and 13 nm [Attwood, 1999]. For several reasons, the choice was made for 13 nm - the future lithography wavelength. To date, almost every detail of a future lithography tool has been designed and optimized, leaving only small room for additional improvement [Attwood, 1999, Lerner, 2000, Stulen & Sweeney, 1999]. Only the heart of such an tool, the 13 nm source is still not determined. The current approaches are far away from the demanded output of several hundred (!) watts of 13 nm radiation on average [Stamm et al., 2002]. Besides the laser-plasmas acting as potential 13 nm source, gas discharge devices are also possible candidates for EUV lithography sources [Klosner et al., 1997]. However, their scalability to high average powers is questionable. On the other hand, a laser-based 13 nm source is almost arbitrarily scalable. If one laser fails to produce the demanded EUV power, one can in principle use two or more lasers shooting onto the same target. This, however, is the scientific approach and not very economic. Thus, the main goal is to optimize the laser-plasma interaction in such a way as to produce as much 13 nm radiation with as low laser energy as possible. The major part of the work presented here addresses the question how the conversion efficiency of laser energy into 13 nm radiation can be improved. The optimization of the production of 13 nm radiation is the challenge for the fabrication of the next-generation of computer chips.

Studies of the EUV emission from laser-produced plasmas have been performed with solid extended metal targets since the early eighties [Carol et al., 1980, Spitzer et al., 1996]. It was soon found that such targets eject a significant amount of macroscopic target fragments, accelerated by thermal stress between the million degree hot center of the plasma and the cold, surrounding material [Trucano et al., 1995]. These fragments referred to as debris destroy nearby sensitive optics [Richardson et al., 1993]. A different approach utilizes targets just the same size as the focal region of the laser. Generally, liquid jets or small droplets with diameters of several tens of µm are used as target. These so-called mass-limited targets provide the chance of almost debris-free operation [Berglund et al., 1998, Richardson, 1998, Rymell & Hertz, 1993]. The question remains what target material is best. A number of materials suitable for mass-limited targets emit radiation around 13 nm, but their particular efficiencies are quite different [Schriever et al., 1998a]. Basically, two
candidates for the target material are currently under investigation: oxygen and xenon. Liquid Xe is a promising material [Kubiak et al., 1998], however, it is difficult to get a stable liquid Xe jet or even small droplets and moreover Xe is extremely expensive. Therefore, the work presented here was performed on oxygen as EUV emitting element. Fivefold ionized oxygen $\text{O}^{5+}$ emits very efficiently at 13.0 nm. There are many materials containing oxygen, like quartz, but the challenge is to produce a mass-limited target. The easiest way to get a small volume of oxygen ions into the laser focus is the use of water droplets as target. Water is cheap, non-toxic and relatively easy to handle and is thus ideal for basic research as well as for industrial applications.

At the start of this work, no systematic investigation of the conversion efficiency on the various laser parameters or target sizes was known for any target applicable in EUV lithography. Focusing on mass-limited targets relying on oxygen as target element, many differing numbers for the conversion efficiency were found in the literature. In the experiments by the Hertz group [Rynell & Hertz, 1993], water droplets with a diameter of $\sim 10 \mu m$ were illuminated with laser pulses from a Q-switched Nd:YAG laser (700 mJ, 8 ns) [Malmqvist et al., 1996] resulting in a conversion efficiency of only 0.01% in a solid angle of 4$\pi$. However, the conversion efficiency obtained by Constantinescu et al. [Constantinescu et al., 2000] with a similar laser (frequency-doubled Nd:YAG, 400 mJ / 10 ns) and $\sim 20 \mu m$ droplets reached 0.1% in 4$\pi$. Richardson et al. reported even conversion efficiencies of up to 0.6% in 4$\pi$ from larger water droplets (60 $\mu m$), again using a Q-switched Nd:YAG laser [Schriever et al., 2000]. The measured conversion efficiencies vary over more than one order of magnitude without obvious explanation.

The current work describes the first systematic investigation of the dependence of the EUV emission on the laser parameters like pulse duration, pulse energy and pulse shape. These experiments shall finally result in the understanding of the dependencies of the EUV emission on the laser parameters and the consequential introduction of scaling laws for these parameters. A major part of the work was the investigation of the influence of the laser pulse duration on the conversion efficiency. Much effort was put into the modification of the laser system so that it was flexible enough to provide pulse durations spanning over five orders of magnitude. This is the first measurement investigating the 13 nm emission of an industrially relevant target with pulse durations ranging from the femtosecond to the nanosecond regime [Düsterer et al., 2001d]. The only work known covering a similar range of pulse durations was performed by the Milchberg group [Parra et al., 2000]. They used a krypton and argon rare gas clusters for their experiments. However, these elements do not show significant emission at 13 nm and they are of no interest for lithography applications. Despite the different target material and size, these measurements can also be explained by the models developed in this thesis.

Not just one single laser pulses but rather a sequence of two laser pulses enlarges the parameter field for the optimization of the EUV emission tremendously. A weak laser pulse, the prepulse, precedes the main laser pulse and creates an expanding plasma sheath at the target surface. The
main pulse now interacts not with the dense target surface but with the thin preformed plasma, altering the interaction completely. It is a well known fact that a prepulse can alter, and in particular enhance, the emission of x-ray radiation from laser produced plasma [Kühlke et al., 1987]. By optimizing the preplasma conditions, several groups [Andreev et al., 2002, Dunne et al., 2000, Nakano et al., 1996] could increase the laser-plasma emittance in the soft x-ray region, focusing on extended solid targets. Even feedback-controlled optimization of the prepulse parameters was successfully performed for solid targets [Feurer, 1999]. But only in one experiment was the soft x-ray emission (at ~3 nm) resulting from a mass-limited target enhanced by introducing an ultraviolet prepulse [Berglund et al., 1996]. In this work, not only will the influence of the prepulse energy and delay between pre- and main pulse be explored, but by changing the prepulse and main pulse duration, it will be shown how tailored prepulses can optimize the conversion efficiency further [Düsterer et al., 2002].

The underlying physics of the laser-plasma interaction, spanning from hydrodynamics at nanosecond long laser pulses to nonlinear interactions when femtosecond pulses are applied to the target, can be described to some extent by analytic equations. Here however, the plasma dynamics were simulated, allowing to visualize the plasma evolution temporally and spatially resolved. To capture the plasma temperature and density at any time after the interaction helps to determine the fundamental processes and variations for the differing laser pulse durations. Furthermore, numerical modeling of the atomic physics in hot plasma allows the determination of the optimum plasma conditions for efficient 13 nm production.

To explore the EUV wavelength range, the Jena TW laser was not operated at full power. Turning up the laser system, it is possible to reach intensities of $5 \times 10^{19}$ Wcm$^{-2}$. These enormous intensities, exceeding the threshold for plasma creation over ten million times, can accelerate a fraction of the electrons and ions within the interaction region up to several MeV or hundreds of keV, respectively. It is easy to see how nuclear reactions can occur in such an environment.

Focusing the TW laser pulse onto the water droplets as used for the EUV experiments, but exchanging the ‘regular’ water by heavy water (D$_2$O), fusion of deuterium nuclei can be observed. In such a hot plasma, two deuterium nuclei fuse together resulting in a $^3$He nucleus and a neutron. The energy distribution of the fusion neutrons can be used as tracer for the ion temperature within the billion degree hot plasma [Ditmire, 2002, Hilscher et al., 2001, Zweiback et al., 2000]. Thus, the neutron spectrometry can be used as noninvasive thermometer for extremely hot plasma. It is known from several experiments [Mackinnon et al., 2001, Snavely et al., 2000] that ions cannot only be accelerated within the laser plasma, but under certain conditions ions can be ripped off the backside of a thin target and accelerated outside the actual laser interaction region. It will be shown that the droplet target, in contrast to the bulk target, can be employed to provide an insight to the various acceleration mechanisms [Karsch et al., 2002].

---

2The radiation in the region between 0.3 nm and 5 nm is called soft x-rays [Attwood, 1999].
The harsh environment of the hot plasma cannot only lead to fusion reactions, but nuclei can also be split into fragments. Electrons accelerated by the intense laser pulse to energies of several MeV give rise to MeV Bremsstrahlung photons when colliding with an ion. These high energetic photons in turn can split nuclei. By measuring the number of fission products, this photo-fission can be utilized as detection scheme for highly energetic photons in the MeV range [Cowan et al., 2000, Düsterer et al., 2001b, Düsterer et al., 2001c, Ledingham et al., 2000, Schwoerer et al., 2001] and [Schwoerer et al., 2002].
2 Experimental setup

2.1 The laser system

The heart of all experiments described in this work is the Jena TeraWatt (TW) laser system (see Fig. 2.1). The laser produces pulses with durations varying from conventional 6 ns pulses down to ultra-short 80 fs. The energy contained in a single laser pulse can be changed from several mJ up to 1 J. Together with a focal spot diameter ranging from 3 µm to 20 µm FWHM\(^1\), interactions with intensities starting from a moderate \(10^{12}\) Wcm\(^{-2}\) up to an ultra-high \(5 \times 10^{19}\) Wcm\(^{-2}\), causing the electrons to quiver relativistically in the laser field, can be explored. To investigate the laser-plasma interaction with matter for such a wide range of parameters, the laser system has to be rather flexible. The Jena TW laser system is based on Ti:sapphire as amplification material. The chirped pulse amplification (CPA) technique is used to increase the initial pulse energy several million times. The basic idea of this amplification scheme is that a short and weak femtosecond pulse is reversibly stretched in time by a factor of 1,000 to 10,000 of its initial pulse duration. The stretched pulse is then amplified and afterwards compressed to almost its initial duration [Strickland & Mourou, 1985]. Without stretching the laser pulse, the amplification would lead to intensities sufficient to undergo undesired nonlinear interactions (e.g. self-phase modulation, self-diffraction and self-focusing) or even cause damage in the amplification material. Using the CPA technique, the energy of the initial pulse can be amplified 100 million fold within only three amplification stages.

The modular conception of the laser system makes it possible to achieve the broad range of laser parameters described above, by changing or bypassing some modules. All the modules as well as the measurement of the pulse duration and the laser intensity will be discussed in detail in Appendix A.

2.2 The setup for the extreme ultraviolet (EUV) measurements

EUV radiation is strongly absorbed in all materials due to a vast number of atomic resonances and strong scattering. For example, 100 µm of air at atmospheric pressure has a transmission of only 50% for radiation at a wavelength of 13 nm [Gullikson]. Therefore, to get an absorption of less than 1% for the 13 nm radiation while propagating from the laser-produced plasma to the EUV detector (∼80 cm), a pressure of less than \(10^{-3}\) mbar was needed. Thus, all experiments had to take place in vacuum. In order to perform the experiments on a long-term basis and under controlled

\(^1\)Full width at half maximum
2.2. The setup for the extreme ultraviolet (EUV) measurements

conditions with a complicated setup, a new interaction vacuum chamber that was exclusively used for the experiments described here had to be designed and built.

2.2.1 The vacuum chamber

Fig. 2.2 shows the 50 cm diameter (60 cm height) vacuum vessel built for the EUV experiments described in Chap. 4. The main chamber is evacuated by a 45 m$^3$/h pre-pump and a 450 l/s turbo pump reaching an end pressure of $< 10^{-5}$ mbar without operation of water droplet target and $5 \times 10^{-4}$ mbar when the droplet source was turned on. The laser beam with a diameter of 43 mm was focused by an off-axis parabolic mirror with a focal length of 119 mm to a focal spot diameter of 20 $\mu$m (for details see appendix A). Located in the middle of the chamber, one can see the most important, fragile and difficult part of the experiment - the droplet source. Usually, water is one of the most unwanted materials in vacuum physics when ultrahigh vacuum conditions have to be reached. Once water contaminates the chamber, it is hard to remove. But to investigate the EUV radiation resulting from water droplets, a way to control water in vacuum had to be found. A commercial 10 $\mu$m diameter capillary from Siemens Elema, initially produced for large scale plotters, acted as nozzle. A description of the droplet formation follows below. In order to produce a stable chain of droplets, a backing pressure of 50 bar was required for the water. A large volume of gas (4 l of nitrogen gas at $\sim 50$ bar) pressurized a small volume (0.2 l) of water within a pressure reservoir (see Fig. 2.7) giving a reasonably constant pressure over several
2.2. The setup for the extreme ultraviolet (EUV) measurements

Figure 2.2: A picture of the interior of the vacuum vessel. A more schematic view on the same setup is shown in Fig. 2.7.

hours. The small diameter of the nozzle allows a flux of only 0.28 ml/min, thus, the sparse amount of water is enough to operate the droplet source for about 12 hours. Since the vapor pressure of water ($\sim 22$ mbar for 20° degree centigrade [Weast, 1980]) is considerably higher than the demanded pressure of $< 10^{-3}$ mbar, the water had to be removed from the interaction chamber by a differential pumping scheme. About 10 mm below the nozzle, the droplets enter a 0.7 mm diameter aperture (conical device in the lower part of Fig. 2.3) connected to a vacuum hose that is fed through the wall of the vacuum vessel. This hose is connected to an additional 8 m$^3$/h pre-pump bringing the water vapor out of the vacuum. Most of the water is extracted by this mechanism. However, water accidentally hitting the edge of the 0.7 mm hole evaporates quickly. The remaining part of the water freezes, and something looking like a tree of ice starts to grow. Unfortunately, this tree grows directly in the direction of the nozzle, and once the ice reaches the nozzle, it will be damaged. To avoid freezing, the area around the 0.7 mm hole is heated to 100° centigrade. The extraction of the droplets out of the interaction chamber has not been described before and was therefore patented [Ziegler et al., 2001].

2.2.2 Characterization of the water droplets

As early as in the 17th century, it was realized that a water jet is not a stable configuration and eventually breaks up into droplets. But the main mechanisms were not known until mid of the 19th century when Lord Rayleigh found that the droplets are formed by the surface tension, acting against the inertia of the fluid. Since that time, modeling of water flows is an active field of research [Eggers, 1997].
2.2. The setup for the extreme ultraviolet (EUV) measurements

The most important requirement for a stable water flow is a low Reynolds number (below ~1000) indicating a laminar flow [Berglund et al., 1998]. The Reynolds number, which is basically a measure of the kinetic energy in relation to the friction, is defined as

$$Re = \frac{\rho v d}{\eta} \quad (2.1)$$

for a liquid jet with a density \(\rho\), a velocity \(v\) and a diameter \(d\). \(\eta\) denotes the viscosity of the fluid. The Reynolds number is calculated to be 600 for the parameters used in the experiment (\(v=60\) m/s, \(\rho = 1\) g/cm\(^3\) and \(\eta = 10^{-3}\) Pa s for water [Gouge & Fisher, 1997]). And indeed, a laminar flow can be observed at the nozzle (see lowest picture in Fig. 2.4). As stated above, a jet is not stable and breaks up spontaneously into irregularly sized droplets (see left part of Fig. 2.5). This breakup occurs at a distance \(L\) below the nozzle, when the surface tension (\(\sigma = 0.073\) N/m for water [Gouge & Fisher, 1997]) has had enough time to amplify small random perturbations. \(L\) can be given as [Berglund et al., 1998]:

$$L = 12v\left(\sqrt{\frac{\rho d^3}{\sigma}} + \frac{3\eta d}{\sigma}\right) \quad (2.2)$$

Consistent with the experimental findings, \(L = 3.0\) mm for the parameters used in the experiment.

To create a stable chain of equally spaced and equally sized droplets, the water jet has to be modulated. A piezo crystal located above the nozzle induced pressure modulations in the jet, which in turn imprint slight variations to the jet diameter. The modulation causes the jet to break up into a reproducible chain of regularly spaced droplets much faster than the random fluctuations occur, as can be seen in Fig. 2.4.

However, Fig. 2.5 shows that the stable droplet formation can only be achieved within a small range of modulation frequencies between 0.8 MHz and 1.8 MHz. The main parameter \(x\) controlling the droplet formation is given by the ratio of the jet diameter \(d\) and the wavelength \(\lambda\) of the modulation:

$$x = \frac{\pi d}{\lambda} = \frac{\pi df}{v} = 0.52 \ f [MHz] \quad (2.3)$$
2.2. The setup for the extreme ultraviolet (EUV) measurements

Figure 2.4: The breakup of the water jet at the nozzle exit (left side) is shown for different modulation frequencies. The images were taken in air (long image) and in vacuum (small image). No modulation frequency was applied in the lower picture, leaving the jet undisturbed (for the first 3 mm behind the nozzle). The images were taken with a microscope using a single fs-laser pulse as flash light to illuminate the droplets. The picture was turned for better presentation. The droplets are moving from top to bottom in the experiment.

The wavelength can also be expressed in terms of fluid velocity $v$ divided by the modulation frequency $f$. The fluid velocity was calculated by knowing the distance between the droplets and the modulation frequency to be 60 m/s for a backing pressure of 50 bar. Since $v$ is constant, $x$ is directly proportional to the modulation frequency. For values of $x < 1$, the breakup of the jet is dominated by the modulation frequency, whereas for $x > 1$, irregular breakup is theoretically predicted [Eggers, 1997] - droplets start to merge into bigger ones. According to equation (2.3), stable droplet formation can be expected for driving frequencies $< 1.9$ MHz. For modulation frequencies below 0.8 MHz ($x < 0.35$), the second harmonic of the modulation frequency starts to dominate [Eggers, 1997]. The break-up at twice the driving frequency is expected. Indeed, the onset of the second harmonic can be seen in Fig. 2.4 for a driving frequency of $f = 0.8$ MHz, but however, the two ‘second harmonic’, droplets tend to merge into one droplet. This is not the case for $f = 0.7$ MHz anymore (see Fig. 2.5). The upper and lower limit predicted by the calculation is in good agreement with the measurement as shown in Fig. 2.5.

The last question concerning the droplet production is whether there are significant differences
in the droplet formation in vacuum compared to air. Fig. 2.6 shows the droplet chain imaged in vacuum (upper row) and in air (lower row). The speed of the droplets, which is proportional to the droplet spacing for constant modulation frequency, is about 5% higher in vacuum since the friction of the ambient air is not present. The droplet size remains unaffected, indicating that the evaporation of the water is rather small compared to the droplet radius. This leads to the conclusion that the droplets are still water droplets several mm below the nozzle and not ice balls, since a considerable fraction (~20%) of the droplet has to evaporate to remove enough heat for the remaining droplet to freeze. Furthermore, the droplets move with a velocity of 60 m/s and thus remain only for about 170 μs within the vacuum chamber before entering the differential pumping aperture - not long enough to evaporate.

In conclusion, a stable chain of droplets with 20 μm diameter and a volume of $V_0 = 4.2 \times 10^{-9}$ cm$^3 \simeq 1.4 \times 10^{14}$ molecules can be produced with a modulation frequency of 1.0 MHz. By tuning the driving frequency within the range of stable droplet formation, it is possible to vary the droplet diameter between 15 μm (i.e. $V = 0.42 V_0$) and 25 μm (i.e. $V = 2.0 V_0$).

---

**Figure 2.5:** The images of the droplets, falling from top to bottom, were acquired for various modulation frequencies between 0.7 and 2.1 MHz. The pictures were taken from the region 4 mm below the nozzle in vacuum. There is only a small frequency region (0.8 to 1.8 MHz) in which the droplet formation is stable and reproducible.

**Figure 2.6:** The images of the droplets (flying from left to right in the picture) recorded for 1.0 MHz modulation. Droplets in vacuum (upper droplets) exhibit a slightly higher speed but the same size compared to droplets in air.
Synchronization between the droplets and the laser pulses

In contrast to commonly used Q-switched lasers which can be triggered externally, the front end of the Jena TW laser is a mode-locked Ti:Sapphire oscillator that cannot be triggered. In the experiments described here, the laser provided the master trigger, and the droplets were triggered by the laser. The pulse repetition frequency of the laser oscillator is 82 MHz (see Sect. A.1). This frequency was subdivided to ∼1 MHz by a delay generator (DG535 Stanford research systems). There is a whole range of frequencies around 1 MHz which can be produced by the subdivision of the 82 MHz, but only very few of them are also phase-locked to the 10 Hz repetition rate of the amplifiers. It is possible to produce a 1 MHz signal which is stable locked to the 10 Hz repetition rate of the laser pulses by fine tuning of the subdivision within the GHz range. To influence the relative phase of the droplet modulation frequency in respect to the 10 Hz of the laser pulse, an electronic phase shifter was built, changing the droplet position with respect to the laser focus. Using this device, the vertical position of the droplet was adjusted such that the laser pulse hit a droplet centrally. In air, this synchronization worked for hours without any adjustment, whereas as soon as the experiments were performed in vacuum, the vertical position of the droplets drifted in respect to the laser focus. After about one minute, the droplets were only partially hit. This could be counterbalanced by a manual fine tuning of the synchronization with the phase shifter. The mechanism responsible for the drift of the droplets was identified by Hemberg et al. [Hemberg et al., 2000] to be the absence of thermal equilibrium right at the tip of the nozzle. The nozzle is continuously cooled by the evaporating water, changing the nozzle radius slightly which influences the droplet velocity. This drift lasts for the first few hours of operation in vacuum. Thus, many experiments were performed without synchronizing the droplets to the laser.

2.3 EUV radiation detection

The exploration of the EUV region within the electro-magnetic spectrum is a relatively recent research topic. One of the most characteristic features of the EUV radiation is the very short absorption length for all materials (typically < 1 µm) due to myriads of atomic resonances [Attwood, 1999]. This strong absorption turned out to be the bottleneck for measuring EUV radiation. Special care has to be taken in order not to absorb the radiation before it reaches the active part of the detector. Moreover, only specially designed detectors are capable of measuring the EUV radiation at all. Two different approaches were chosen in the present work in order to measure the EUV radiation resulting from the laser-plasma interaction. First, an imaging transmission grating spectrometer equipped with a backside illuminated CCD was used to record the spectra of the line emission of several O$^{5+}$ transitions in the wavelength region from 5 to 20 nm. The spectrometer can be coupled to a streak camera to provide time-resolved EUV spectra. This EUV spectrometer was absolutely calibrated in the spectral window between 10 nm and 15 nm at a synchrotron source.
Second, a much simpler device, an *EUV monochromator*, was constructed in order to measure only the radiation at $13.0 \pm 0.2$ nm.

**Figure 2.7:** The picture shows a three-dimensional impression of the experimental setup used for the EUV measurements.

### 2.3.1 The EUV spectrometer

**Design**

The basic setup of the spectrometer, constructed by Jasny et al. [Jasny et al., 1994], is shown in Fig. 2.8 (see also Fig. 2.7). The EUV radiation emitted by the plasma is collected by the imaging mirror, subsequently dispersed by the transmission grating and finally the spectrum is recorded with a CCD camera. The main advantage of the EUV spectrometer is the imaging mirror. In contrast to a standard pinhole spectrometer (Sect. 2.3.2) which collects typically not more than a tiny solid angle of $10^{-8}$ sr of the emitted EUV radiation with a spectral resolution of $\lambda/\Delta \lambda \geq 500$,
2.3. EUV radiation detection

the toroidal imaging mirror of the described spectrometer is capable of collecting \(5.9 \times 10^{-4}\) sr. This rather large solid angle enables the acquisition of EUV spectra from single laser shots. The sensitivity is enhanced at the expense of the much lower spectral resolution of only about 20.

A toroidal mirror images the EUV radiation from the laser-plasma source 1:1 onto the CCD, thus the plasma acts as ‘slit’, comparing the setup to a ‘regular’ spectrometer. However, for particular laser parameters (high laser pulse energy and long pulse duration), the source size which is imaged onto the CCD can exceed 100 \(\mu\text{m}\) in diameter, thus degrading the spectral resolution. In addition, the rectangular shape of the imaging mirror causes aberrations, blurring the image even more (see Fig. 2.9).

Figure 2.9: The picture shows a CCD image cutout of the minus first order of the spectrometer in which the four lines, discussed in the next chapters, can be identified. The enlarged region shows that the resolution of the 13 nm peak is limited by the source size.

The rectangular mirror (20 \(\times\) 100 mm) is located at a distance of 360 mm from the laser-plasma. The toroidal radii are 5375 mm in the plane of the drawing (Fig. 2.8) and 24 mm in perpendicular direction. The mirror is designed in a way that the EUV radiation impinges on the toroidal surface at incidence angles greater than 86°. Since the mirror is coated with nickel, having a refractive index smaller 0.9977 for the measured wavelength region \([\text{Gullikson}]\), all EUV radiation is reflected under total external reflection. A gold transmission grating with 2000 lines/mm was inserted in-between the mirror and the detector CCD. The free-standing 250 nm thick gold bars have a period of 500 nm. Since the slits between the bars have the same size as the bars themselves, the modulation of the grating diffraction with the single-slit diffraction cancels out the 2\(^{\text{th}}\) order of diffraction. The bars are intersected by a supporting grid with periods of several \(\mu\text{m}\), not affecting the EUV diffraction. A micro-channel plate (MCP) was used as EUV detector in the original setup \([\text{Jasny et al., 1994}]\). However, since the entire spectrometer setup had to be absolutely calibrated by synchrotron radiation, a backside illuminated thinned CCD was chosen. In contrast to the MCP, showing saturation effects \([\text{Giudicotti et al., 1994}]\), the response of a CCD is linear for pulsed (laser-plasma) as well as for continuous (synchrotron) radiation. A regular, front side illuminated CCD is very insensitive for EUV radiation due to a \(\sim 100\) nm thick SiO\(_2\) oxide layer on top of the chip which is essentially opaque for EUV radiation \([\text{Li et al., 1995}]\). To circumvent this problem, a backside illuminated thinned CCD chip was chosen. The substrate of the chip is removed, and the chip is illuminated from the backside. This results in an oxide layer \(<10\) nm.
which reduces greatly the EUV absorption of the CCD chip [Li et al., 1995]. The CCD chip has 1024×256 pixels, each pixel is 24×24µm in size with a dynamic range of 16 bit (DO420-BN from Andor). During the measurement the CCD chip is cooled down to -55° centigrade to keep the dark current below 5 counts per pixel and 5 seconds (which is the typical integration time). This thermal noise is negligible since the typically acquired spectra show several thousand counts per pixel (e.g. see Fig. 2.11). In order to prevent contamination of the fragile CCD chip with water from the target chamber, the vacuum at the camera was separated from the vacuum within the interaction chamber by a 200 nm thick polyimide foil. This foil transmits only 30% of the 13 nm radiation, but the lifetime of the CCD chip was greatly enhanced due to the water-free vacuum.

**Calibration of the spectrometer at a synchrotron (BESSY II)**

One of the major goals of the project was to measure *absolute numbers* for the EUV photon yield produced by a certain laser pulse energy. With this information, the conversion efficiency (CE) of the EUV yield can be given. It is possible to calculate the transmission or reflection for each component of the spectrometer, but the uncertainty for this kind of calculation is rather large. Therefore, the entire spectrometer, as it was used in the experiments, was calibrated with a source of known photon flux. The only reliable EUV photon source for which the number of photons, the angular radiation distribution and the spectrum can be precisely calculated is a synchrotron. Electrons moving with relativistic energies emit radiation in a small forward pointing cone when their direction is changed, i.e. when they are accelerated. Using just one bending magnet to change their direction, the resulting spectral distribution of the radiation is a wide continuum. However, by undulating the electrons in a magnetic field, periodically changing its sign, the resulting spectrum becomes narrower. The line width $\lambda/\Delta\lambda$ is equal to the number of periods of the magnetic field $N$ which is typically on the order of 100 [Attwood, 1999]. However, the electrons undulate not perfectly harmonically, thus radiation from higher harmonics is always present in the spectrum, as can be seen in the calculated undulator spectrum in Fig. 2.10 as well as in the measured spectrum in Fig. 2.11. The calibration of the spectrometer was performed in close cooperation with the Physikalisch-Technische Bundesanstalt (PTB) at the new Berlin electron storage ring for synchrotron radiation (BESSY II).

Since the EUV beam produced by the synchrotron has a diameter of several mm, a pinhole (0.5×0.5 mm²) had to be placed in the object plane. This pinhole was imaged 1:1 onto the CCD as shown in Fig. 2.11. The calibration measurements were performed with an electron current of 60 nA circulating in the synchrotron, resulting in $4.6 \times 10^7$ photons per second in the 13 nm peak (see Fig. 2.10). Typical exposure times ranged between 5 and 60 seconds, testing the linearity of the CCD response.

By comparing the area under the peaks at 13 nm for the calculated (Fig. 2.10) and the measured
2.3. EUV radiation detection

(Fig. 2.10) spectra, the response (CCD counts per photon) of the spectrometer was calculated to be 0.010 counts per photon at 13 nm. This procedure was repeated for 10 nm to 15 nm in 1 nm steps. Thus, the spectrometer was absolutely calibrated in the spectral range of 10 nm to 15 nm.

Calibration of the Zirconium filters

In recording the emission spectra of the laser-plasma interaction, the strayed laser light would overexpose the CCD camera completely. Therefore, a filter has to be used which blocks the entire visible light, but transmits as much as possible of the EUV radiation. As already pointed out, the EUV radiation is strongly absorbed in all materials, however, there are several elements transmitting a fair amount of 13 nm radiation when used as thin foils. The transmission for the most promising materials was scanned by using an internet database for wavelength-dependent transmission data [Gullikson]. The highest transmission is achieved by thin beryllium foils (a 200 nm thin foil transmits \( \sim 75\% \) 13.0 nm radiation), which unfortunately become highly toxic when destroyed. The second best choice is zirconium (Zr). According to the database, a 200 nm thick Zr foil still transmits 52% of the incident 13.0 nm radiation, and the transmission is almost constant over the range of interest between 10 nm and 15 nm. Since the Zr foil was not mounted in the spectrometer (as there is no visible light emitted by the synchrotron) while calibrating the spectrometer at BESSY II, the transmission of the Zr filter was not included in the calibration factor. In order to check the calculated transmission values of the database, the wavelength-dependent transmission of a thin Zr foil \( T_{Zr}(\omega) \) was measured. The spectrally resolved signal from the spectrometer is the product of the EUV radiation emitted by the plasma times the spectrometer response \( S(\omega) \) and
the Zr transmission $T_{200\text{nm Zr}}$. The transmission for a given filter thickness $d$ is given by $T_{Zr} = e^{-d/d_0}$, $d_0(\omega)$ is the material and wavelength-dependent 1/e-thickness. It is possible to determine $T_{Zr}(\omega)$ by performing the same experiment twice with identical laser parameters, providing the same $S(\omega)$ for the two experiments. Thus, recording the two spectra with a Zr filter thickness of 200 nm for the first experiment and 400 nm Zr for the second run, the spectrometer response $S(\omega)$ cancels out as soon as the fraction of both measured spectra is calculated:

$$\frac{S(\omega) \times e^{-400nm/d_0(\omega)}}{S(\omega) \times e^{-200nm/d_0(\omega)}} = e^{-200nm/d_0(\omega)} = T_{200\text{nm Zr}}(\omega)$$

(2.4)

Thus, the Zr transmission of a 200 nm thick foil $T_{200\text{nm Zr}}(\omega)$ was experimentally obtained as function of the wavelength. The measured and the calculated transmission agree well, as shown in Fig. 2.12.

**Acquiring time-resolved spectra**

The time-dependence of the EUV spectrum can be recorded by using of a streak camera coupled to the transmission grating spectrometer. The streak camera (from Kentech instruments Ltd.) was mounted between the actual spectrometer and the CCD camera. Incoming EUV photons produce photo-electrons at the photo-cathode of the streak camera. These electrons are accelerated onto a phosphorescent screen by a static electric field. The image on the phosphorescent screen is exactly the image (the spectrum) initially imprinted on the cathode by the EUV photons. This screen is finally imaged to the CCD camera yielding just the EUV spectrum. Not much is gained by transferring the photon spectrum into an electron current and back to photons again, but by applying a very fast sweeping voltage ramp perpendicular to the spectral dimension, the electrons are deflected to different angles according to their time of arrival. The outcome of the device is a two-dimensional image with one direction providing the spectral information of the radiation and the perpendicular axes shows the temporal evolution for each spectral component (see e.g. [Feurer & Sauerbrey, 1996]).
2.3. EUV radiation detection

2.3.2 The pinhole spectrometer

In addition to the imaging transmission grating spectrometer that collects a rather large solid angle of radiation but with a poor spectral resolution, a high resolution pinhole spectrometer\(^2\) was used for some experiments. The spectrometer consists of a 50µm × 500µm rectangular pinhole located 68 cm away from the plasma, imaging the plasma to a backside illuminated CCD that is located 40 cm behind the pinhole. A 10,000 lines/mm transmission grating right behind the pinhole was used as dispersive element. The spectrometer provided a high resolution\(^3\) of \(\lambda/\Delta\lambda=500\), at the expense of the collected solid angle of only 4.3 ×10\(^{-9}\) sr.

2.3.3 The EUV monochromator

The transmission grating spectrometer provides the full set of information over a large spectral range. These data are valuable for determining various plasma parameters, but it is not necessary in all cases to acquire the full information. Therefore, a small device, the EUV-monochromator tool specially tuned for 13.0 nm radiation was constructed and built.

As shown in Fig. 2.13, the monochromator consists of a 800 nm Zr foil to block the visible light and a molybdenum-silicon multilayer mirror optimized for a maximum reflectivity at 13.0 nm for an angle of incidence of 7.5°. The Mo-Si mirror consists of 40 alternating Mo and Si layers, each with a thickness of 6.4 nm (~20 to 30 atomic layers). Fig. 2.14 shows the reflection curve for such a mirror, calculated by an online database [Gullikson]. The reflection bandwidth of 0.4 nm is considerably larger than the investigated 13 nm radiation line width, thus the mirror reflects the entire 13 nm radiation without spectral clipping.

![Figure 2.13: Scheme of the EUV monochromator tool. The visible (laser) light is blocked by the Zr-filter. Out of the entire range of EUV wavelength entering the device, only the 13 nm radiation is reflected by the multilayer mirror and detected by the photodiode.](image)

The 13 nm photons reflected from the multilayer mirror were detected by a GaAsP Schottky diode (G1127-02 from Hamamatsu) without a glass protection window. The signal from the diode was tenfold amplified by a preamplifier located within the interaction chamber, close to the diode. In the experiment, the monochromator tool was positioned about 10 cm away from the laser-plasma to get a sufficiently large signal. The amplified voltage peak from the diode was measured

\(^2\)The spectrometer was provided by the Fraunhofer Institut für Lasertechnik (ILT) in Aachen

\(^3\)According to ILT (unpublished).
2.3. EUV radiation detection

with an oscilloscope. It was carefully checked that the diode signal was proportional to the EUV emission of the target by calibrating the tool against the absolutely calibrated transmission grating spectrometer. The calibration result can be seen in Fig. 2.15.

**Figure 2.14:** The figure shows the reflection curve of the Mo-Si multilayer mirror mounted in the EUV-monochromator. The reflection for longer and shorter wavelengths outside the shown region is well below 2%. The reflection curve was calculated by an online database [Gullikson] for unpolarized EUV radiation at an angle of incidence of 7.5°.

**Figure 2.15:** Calibration of the EUV monochromator against the transmission grating spectrometer. The EUV signal from a laser shot is simultaneously recorded by both devices. One dot represents one laser shot. The response of the monochromator is indeed linear on the EUV photon yield.
3 Theoretical background

The experiments described in this thesis deal with a rather complex state of matter far away from our 40 eV world. Temperatures of several million degrees are achieved during the interaction of matter with laser light. Electrons are torn away from the atom, and plasma evolves. Besides laser-produced plasma, such hot and dense plasmas can only be found in the interior of stars. Here on earth, the enormous energy density present in the laser-produced plasma is far away from a thermal equilibrium with the surrounding world. As a consequence, the plasma is inherently short-lived. The physics described here takes place on time-scales of picoseconds ($10^{-12}$ s) and nanoseconds ($10^{-9}$ s). To get a qualitative and most of all, a quantitative picture of the various processes taking place in such an exotic state of matter, a theoretical description of the plasma and atomic physics under extreme conditions is essential.

3.1 Basic plasma physics

Plasma is basically a many-body system composed of electrons and ions that are coupled to one another by their electric and magnetic fields. Because of the vast number of electrons and ions taking part in the interaction ($\sim 10^{15}$ in case of the water droplets) an adequate theoretical description has to average over all particles. Macroscopic parameters ($T_e, T_i, n_e, n_i$ and $Z$) are introduced describing most phenomena quite well. The parameters $n_e$ and $n_i$ denote the electron and ion density in units of number of particles per cm$^3$. Created out of neutral matter, plasmas are generally charge-neutral. Summing up all the charges of the electrons and ions no net charge is left. In other words, the product of the number of ions and the average charge state of the ions $Z$ equals the number of free electrons:

$$n_e = Z n_i$$  \hspace{1cm} (3.1)

$T_e$ and $T_i$ denote the electron and the ion temperature, respectively. The temperature can be defined for a thermal equilibrium via the Maxwellian energy distribution function $f(E_{kin})$ [Salzmann, 1998]:

$$f(E_{kin}) \, dE_{kin} = \frac{2}{\sqrt{\pi} \, (k_B T)^{3/2}} \, \sqrt{E_{kin}} \exp \left( - \frac{E_{kin}}{k_B T} \right) \, dE_{kin}$$  \hspace{1cm} (3.2)

Concentrating on the electrons, the mean thermal velocity $v_e$ for electrons moving in one direction is linked to the electron temperature by [Attwood, 1999]:

$$E_{kin} = \frac{1}{2} m_e v_e^2 = \frac{1}{2} k_B T_e$$  \hspace{1cm} thus  \hspace{1cm} $v_e = \sqrt{\frac{k_B T_e}{m_e}}$$  \hspace{1cm} (3.3)

with the electron mass $m_e$. $k_B$ is the Boltzmann constant, conveniently expressed in units of eV/K ($k_B = 8.6 \times 10^{-5}$ eV/K) [Attwood, 1999]. For practical reasons, the temperature will often be given in energy units of eV, according to the relation 1 eV $\equiv 1.2 \times 10^4$ K.
Electrons and ions are influenced by the electric field $E$ produced by the nearby charges. Due to the much larger mass of the ions, usually only the motion of the electrons is considered. Following Kruer [Kruer, 1988], the electric field can be decomposed into two separate fields $E_1$ and $E_2$ with different spatial scales. $E_1$ has fluctuations on a scale below the so-called Debye length $\lambda_D$ (see below), which is the length over which the field contribution of a single charge is shielded by the surrounding electrons. This rapidly fluctuating microfield is due to random collisions between individual charges. Therefore, *collisional* processes (see Sect. 3.2.2) are linked to $E_1$. $E_2$ on the other hand is related to deviations from charge neutrality on a scale that is large compared to $\lambda_D$, averaging out small scale fluctuations. Thus, $E_2$ gives rise to *collective* or *collisionless* electron motion, like plasma waves (see Sect. 3.2.3). This classification provides a natural separation into collisional and collective behavior.

The Debye length can be given in terms of $n_e$ and $T_e$ as [Attwood, 1999]:

$$\lambda_D = \left( \frac{\varepsilon_0 k_B T_e}{e^2 n_e} \right)$$  \hspace{1cm} (3.4)

with the electron charge $e$ and the permittivity of free space $\varepsilon_0$. $\lambda_D$ is on the order of several nm for the plasmas relevant to the experiments ($n_e \sim 10^{21} \text{cm}^{-3}$ and $T_e \sim 100 \text{ eV}$). A closely related parameter is $N_D$, the number of particles within the so-called Debye sphere that denotes a sphere with the radius $\lambda_D$ [Kruer, 1988]

$$N_D = \frac{4\pi}{3} \lambda_D^3 n_e = 1.7 \times 10^9 \sqrt{\frac{T_e^3 \text{[eV]}}{n_e \text{[cm}^{-3}]}}.$$  \hspace{1cm} (3.5)

If there are many particles in the Debye sphere, the microscopic fluctuations related to $E_1$ are effectively screened and the macroscopic description of the plasma is more appropriate. Thus the more particles in the Debye sphere, the smaller the contributions of the collisions and thus, collective behavior becomes dominant. $N_D$ is about 50 for the parameters given above. The relatively small number gives already a hint that collisional as well as collective processes will have to be considered.

Another important parameter, related to the electron density, is the *plasma frequency* $\omega_p$. Subsequent to a distortion of the charge neutrality, e.g. by a short laser pulse or some random fluctuation, the electrons within the plasma tend to oscillate with the plasma frequency [Attwood, 1999]:

$$\omega_p = \sqrt{\frac{e^2 n_e}{\varepsilon_0 m_e}}$$  \hspace{1cm} (3.6)

This formula was used first by Lord Rayleigh\(^1\) in 1906 that treated the atom as a simple pumpkin model [Williams, 2001]. The plasma frequency $\omega_p$ marks a fundamental boundary between

---

\(^1\) He influenced a large part of physics. Note that he is also quoted for contributions on fluid dynamics (p. 8) and laser beam propagation (p. viii).
conducting and dielectric behavior in the interaction of electromagnetic waves with plasmas. For light with frequencies large compared to the plasma frequency, the electrons inertia retards their response and the *underdense* plasma behaves like a dielectric. As a result, the underdense plasma is rather transparent to the radiation. When the incident laser frequency is less than $\omega_p$, the electrons can respond and exclude the incident field, resulting in a reflection of wave energy from the so-called *overdense* plasma [Williams, 2001]. The electron density at which the plasma frequency equals the laser frequency $\omega_L$ is called the *critical electron density* $n_{\text{crit}}$, denoting the boundary between underdense (or *undercritical*) plasma and overdense (or *overcritical*) plasma. According to Eq. (3.6) the critical density is given by:

$$n_{\text{crit}} = \frac{\varepsilon_0 m_e \omega^2_L}{e^2} = \frac{1.11 \times 10^{21}}{\lambda_L^2 \text{[\mu m]}} \text{ cm}^{-3}$$  \hspace{1cm} (3.7)

with the laser wavelength $\lambda_L$. The critical density for the Ti:sapphire laser system $\lambda_L= 0.8 \mu m$ is, therefore, $n_{\text{crit}} = 1.7 \times 10^{21} \text{ cm}^{-3}$.

In the underdense plasma, electrons cannot react ‘fast enough’ to exclude the electromagnetic radiation, but the propagation of a one-dimensional plane wave of the form

$$E(r, t) = E_0 e^{-i(\omega_L t - kx)}$$  \hspace{1cm} (3.8)

with the wave vector $k = 2\pi/\lambda_L$ is influenced. The dispersion relation of a collisionless plasma is [Attwood, 1999]:

$$\omega^2_L = \omega^2_p + k^2 c^2$$  \hspace{1cm} (3.9)

The group velocity $v_g = \partial \omega_L / \partial k$ of the laser radiation as function of the electron density can be derived from Eq. (3.9) to be:

$$v_g = c \frac{n}{n_{\text{crit}}} = c \sqrt{1 - \frac{\omega^2_p}{\omega^2_L}} = c \sqrt{1 - \frac{n_e}{n_{\text{crit}}}}$$  \hspace{1cm} (3.10)

where $n$ is the refractive index of the plasma. It is to note that this dependence of the group velocity on the refractive index is not generally applicable. It is only valid for the dispersion relation given in Eq. (3.9). $n$ becomes imaginary and the electromagnetic wave decays evanescently for electron densities $n_e > n_{\text{crit}}$, i.e. for overcritical plasma.

### 3.2 Laser-plasma interaction

Several properties of a plasma have been reviewed above. The next concern is the production of plasma and the subsequent influence of the laser electromagnetic field on the plasma evolution. Focusing intense laser pulses with durations between several ns down to few fs onto any material, plasma will be created whenever the laser intensity exceeds a certain threshold. In case of water,
3.2. Laser-plasma interaction

the plasma formation threshold is about $3 \times 10^{10}$ Wcm$^{-2}$ for ns pulses and $3 \times 10^{12}$ Wcm$^{-2}$ for fs-pulses [Hammer et al., 1997]. Multi-photon absorption of the laser radiation, happening within the front part of the laser pulse, provides some free electrons. These will be accelerated by the electric field of the laser beam and subsequently transfer their energy to the surrounding cold material, eventually creating a hot plasma. However, there is no single model which would describe all phenomena [Gibbon & Förster, 1996]. To explain the dependence of the EUV conversion efficiency on the laser parameters, the knowledge of the interplay of the different processes is necessary. The most important processes will be discussed below.

3.2.1 Plasma expansion

After a laser pulse heated the plasma, a large fraction of the energy absorbed by the plasma will be converted into kinetic energy of the plasma expansion. According to the hydro-dynamics simulation, 30-80% (see Sect.3.2.5) of the absorbed energy drives the expansion depending on the plasma parameters.

By analogy with the ideal gas law, the thermal pressure is given by $n_e k_B T_e$, where the electrons are responsible for the pressure due to their much higher mobility, whereas the ions with mass $m_i$ counteract the expansion with their inertia $n_i m_i = n_e / Z m_i$ according to Eq. (3.1). Furthermore, using continuity and momentum equations and assuming adiabatic expansion, it is possible to connect the expansion velocity far away (several cm) from the initial laser-plasma interaction region with the initial plasma or electron temperature $T$ or $T_e$, respectively, and the initial charge state $Z$ short-time after the laser pulse. The equation for the ion acoustic speed $c_s$, which is identical to the expansion velocity, is given for a plain geometry by [Attwood, 1999, Huba, 1994]:

$$v_{exp} = c_s = \sqrt{Z \frac{\gamma k_B T_e}{m_i}}$$

(3.11)

$\gamma$ is the ratio of the specific heats equals $\gamma = \frac{5}{3}$, for three translational degrees of freedom. Eq. (3.11) assumes that the ion temperature is much smaller than the electron temperature, which is certainly true for short pulses (some ps) since it takes about 30 ps till the hot electrons thermalize with the ions, as will be calculated in Sect. 3.2.2. Thus, using pulse durations $> 30$ ps, the assumption of $T_e = T_i = T$ is fulfilled and one has [Tsui et al., 1993]:

$$v_{exp} = \sqrt{\frac{Z + 1)(\gamma + 1) k_B T}{m_i}} \quad \text{or in terms of energy : } \quad k_B T = \frac{2 E_{kin}}{(Z + 1)(\gamma + 1)}$$

(3.12)

where $E_{kin} = m_i / 2 v_{exp}^2$. This equation, however, neglects the fact that the expansion can be influenced by the recombination of ions. Energy released by the recombination of the ions can be transferred into kinetic energy. Assuming that the entire recombination energy is returned to the
plasma, the appropriate equation is [Tsui et al., 1993]:

\[ k_B T = \frac{2(\gamma - 1)E_{\text{kin}}}{(Z + 1)(\gamma + 1)\gamma} \]  

(3.13)

Treating the laser-plasma interaction as stationary problem which can be justified for nanosecond laser pulses, the kinetic energy of the ions can be linked to the laser intensity \( I_L \). According to Ref. [Puell, 1970] the relation is:

\[ E_{\text{kin}} \propto I_L^{4/9} \quad \text{thus} \quad v_{\text{exp}} \propto I_L^{2/9} \]  

(3.14)

Using the same model, the electron temperature of the laser heated plasma can be expressed by:

\[ T_e \propto I_L^{4/9} \]  

(3.15)

The velocity distribution of the expanding plasma is Maxwellian since the expansion takes place on a time-scale large compared to the equilibration time (see Eq. (3.28)). For a plasma which is quickly heated by a laser pulse that is short in comparison to the plasma expansion time, the electrons and ions start all simultaneously to expand. After some time \( t \), the spatial density gradient is a ‘projection’ of the velocity distribution. A few fast ions are far away - many slow ones are closer to the target surface. Mapping the velocity distribution into a spatial distribution results in [Attwood, 1999]:

\[ n_i(x) = n_i(0) e^{-x/v_{\text{exp}} t} \]  

(3.16)

with the initial solid state electron density of about \( 10^{23} \) cm\(^{-3} \). In general, the gradient of the electron density is expressed in terms of the scale-length [Gibbon & Förster, 1996, Attwood, 1999]:

\[ L = \left| \frac{n_e}{\nabla n_e} \right| \]  

(3.17)

The scale-length mentioned in the following sections always denotes the scale-length at the critical electron density \( n_e = n_{\text{crit}} \).

### 3.2.2 Collisional absorption mechanisms

**Basic processes**

So far, collisions have not been considered. However, up to laser intensities of \( 10^{15} \) Wcm\(^{-2} \), collisional absorption is the dominant process of the laser-plasma interaction at a laser wavelength of 800 nm (e.g. [Attwood, 1999, Gibbon & Förster, 1996, Giulietti & Gizzi, 1998]). Electrons quivering in the laser field eventually lose part of their kinetic energy by collisions with the surrounding ions, equilibrating the energy gained by the electromagnetic field to all particles within the neighborhood.
3.2. Laser-plasma interaction

To include the electron-ion collisions mathematically, the dispersion relation for collisionless plasma, Eq. (3.9), has to be modified by introducing the electron-ion collision frequency $v_{ei}$ in the following way [Attwood, 1999]

$$\omega_L^2 = \omega_p^2 \left(1 - \frac{i_v_{ei}}{\omega_p}\right) + k^2 c^2$$ (3.18)

resulting in a complex wave vector $k = k_{\text{real}} + ik_{\text{im}}$. Inserting the complex $k$ into Eq. (3.8) leads to an absorption term proportional to $e^{-k_{\text{im}}x}$. The wave is exponentially damped while propagating through the plasma. For the plasmas of interest in the experiments of the thesis $\omega_L \gg v_{ei}$.

Rearranging Eq. (3.18) under this constraint results in the collisional absorption coefficient:

$$k_{\text{im}} = \frac{v_{ei} \omega_p^2}{2 \omega_L} = \frac{v_{ei} n_e}{2 c \ n_{\text{crit}} \sqrt{1 - \frac{n_e}{n_{\text{crit}}}}}$$ (3.19)

The electron-ion collision frequency $v_{ei}$ for a Maxwellian electron velocity distribution (Eq. (3.2)) as function of the electron density $n_e$, the electron temperature $T_e$ and the average charge state $Z$ can be expressed by [Attwood, 1999, Giulietti & Gizzi, 1998]:

$$v_{ei} = \frac{e^4 Z n_e \ln \Lambda}{3 (2\pi)^{3/2} \epsilon_0^2 m_e^{1/2} (k_B T_e)^{3/2}} = 3 \times 10^{-6} \frac{Z n_e [\text{cm}^{-3}] \ln \Lambda}{(T_e [\text{eV}])^{3/2}} \text{s}^{-1}$$ (3.20)

The so-called Coulomb logarithm $\ln \Lambda$ is defined as logarithm of the ratio of the maximum $b_{\text{max}}$ and minimum $b_{\text{min}}$ impact parameter of the electron-ion scattering. $b_{\text{max}}$ corresponds to the Debye length $\lambda_D$ (Eq. (3.4)) beyond which the ion charge is effectively screened. $b_{\text{min}}$ is related to the thermal energy of the electrons. Equating the Coulomb energy at closest approach $e^2 Z/(4\pi \epsilon_0 b_{\text{min}})$ to the thermal energy $\frac{3}{2} k_B T_e$ leads to $b_{\text{min}} = Z/(6\pi \lambda_D^2 n_e)$, thus using Eq. (3.5) results in [Attwood, 1999]:

$$\ln \Lambda = \ln \left(\frac{b_{\text{max}}}{b_{\text{min}}}\right) = \ln \left(\frac{9 N_D}{2 Z}\right) = 22.8 + 0.5 \ln \left(\frac{T_e^3 [\text{eV}]}{Z^2 n_e [\text{cm}^{-3}]^3}\right)$$ (3.21)

However, due to the logarithmic dependence the Coulomb logarithm varies only slightly with the parameters. Rearranging Eq. (3.19) and omitting numerical constants the collisional absorption scales like:

$$k_{\text{im}} \propto \frac{n_e^2 Z}{T_e^{3/2} \sqrt{1 - n_e/n_{\text{crit}}}}$$ (3.22)

The absorption increases essentially quadratically with the electron density until the critical density $n_{\text{crit}}$ is reached, where the group velocity (Eq. (3.10)) becomes zero and the light is reflected. This leads to the important conclusion that the collisional absorption basically takes place in the vicinity of the critical density. This result explains also that short wavelength laser pulses are better absorbed than long wavelength pulses [Garban-Labaune, 1982]. $n_{\text{crit}}$ increases for shorter wavelengths (Eq. (3.7)), therefore, UV lasers can penetrate into denser plasma where the absorption is higher. This scaling will be important to explain the dependence of the EUV yield on the laser pulse duration and to understand the influence of prepulses.
The absorption coefficient \( k_{int} \) describes the absorption locally. The next step is to calculate the complete absorbed laser energy for a given model. There are basically two limits to be considered. First, a perfectly steep density gradient is assumed which is a good approximation for short pulses. Secondly, the absorption for a long scale-length plasma will be calculated which is appropriate for long laser pulses or for the interaction with a preformed plasma created by a prepulse.

The absorption of a plasma with a step-like density profile can be calculated for arbitrary angle of incidence \( \theta \) and an arbitrary laser polarization [Gibbon, 2002, Gibbon & Förster, 1996]. Having a smooth surface, such that scattering of the laser light can be neglected, the absorption \( A(\theta) \) can simply be calculated by \( 1 - R(\theta) \); where \( R(\theta) \) is the intensity reflection coefficient. The reflection is calculated by standard electro-dynamics, using the Fresnel equations [Gauthier, 2001]:

\[
A_{\text{step},s} = 1 - R_s(\theta) = \left| \frac{\cos(\theta) - \sqrt{\varepsilon - \sin^2(\theta)}}{\cos(\theta) + \sqrt{\varepsilon - \sin^2(\theta)}} \right|^2
\] (3.23)

where \( \theta \) is the angle between the target normal and the laser beam. This equation holds if the electric field vector undulates parallel to the target surface - so-called \( s \)-polarized light. For the polarization perpendicular to \( s \)-polarized light, which is \( p \)-polarized light, there is always a component of the electric field normal to the target surface. The reflection coefficient in this case is given by:

\[
A_{\text{step},p} = 1 - R_p(\theta) = \left| \frac{\varepsilon \cos(\theta) - \sqrt{\varepsilon - \sin^2(\theta)}}{\varepsilon \cos(\theta) + \sqrt{\varepsilon - \sin^2(\theta)}} \right|^2
\] (3.24)

\( \varepsilon \) is the dielectric constant of the plasma. Referring to the Drude model (cp. Eq. (3.18)), \( \varepsilon \) can be given as [Gauthier, 2001]:

\[
\varepsilon = 1 - \frac{n_e}{n_{crit}} \frac{1}{1 + i v_{ei}/\omega_L}
\] (3.25)

However, in laser-produced plasmas one will often encounter density gradients and no step-like distributions as pointed out in Sect. 3.2.1. Kruer [Kruer, 1988] solved Maxwell’s equations for an exponential density gradient, as in Eq. (3.16), with a scale-length \( L \gg \lambda_L \). The absorption of \( s \)-polarized light is given as:

\[
A_{L,s} = 1 - \exp \left( -\frac{8v_{ei}^* L}{3c} \cos^3(\theta) \right)
\] (3.26)

with the electron-ion collision frequency at the critical density \( v_{ei}^* \). The analysis for \( p \)-polarized light is somewhat more involved [Gibbon, 2002], but having the droplet target in mind, the most light impinging is \( s \)-polarized.

Both models show the highest absorption at \( \theta = 0 \) for \( s \)-polarized light which is slowly decreasing to 0 at grazing incidence. \( p \)-polarized light, on the other hand has an optimum absorption at an angle \( \theta > 0 \). This optimum angle (for \( L \gg \lambda_L \)) is the same as given in Eq. (3.32) for the collisionless absorption.
3.2. Laser-plasma interaction

**Equipartition times**

The laser pulse first of all accelerates electrons, since they are much lighter than the ions. After many collisions, electrons and ions finally will be in equilibrium and have the same temperature. Here, the timescales at which the particles get into equilibrium, i.e. having a Maxwell distribution, are calculated. Non-equilibrium electron populations, such as those created from resonance absorption (see Sect. 3.2.3), thermalize within the time-scale of $t_{ee}$, which is the Spitzer electron self-collision time [Salzmann, 1998]:

$$
t_{ee} = 0.29 \sqrt{\frac{m_e c^2 T_e^{3/2}}{n_e e^4 \ln \Lambda}} = 0.33 \left( \frac{T_e [eV]}{100} \right)^{3/2} \frac{10^{21}}{\ln \Lambda n_e [cm^{-3}]} ps \quad (3.27)
$$

It takes only about 60 fs until the electrons are in equilibrium for typical laser-plasma parameters, namely $T_e \sim 200$ eV and the critical density. As expected, the ions will take up more time. Many collisions between electrons and ions will be necessary to achieve equilibrium. The equipartition time between electrons and ions is given by [Salzmann, 1998]:

$$
t_{ei} \approx 1000 \times \frac{A}{Z^2} \times t_{ee} \quad (3.28)
$$

After about 40 ps, electrons and fivefold ionized oxygen will have about the same temperature. A similar time-scale was found in the plasma dynamic simulations (Sect. 3.2.5). Thus, electrons equilibrate fast enough and $t_{ee}$ can be neglected, while $t_{ei}$ has to be considered for laser pulses with durations of several ps or even less.

**Dependence of the collisional absorption on the laser pulse length**

Another important issue is the scaling of the absorption with the laser pulse duration. The simplest picture implies that the longer the driving laser pulse, the more collisions take place and the plasma is heated efficiently by the absorbed laser energy. In the interaction of ‘long’ laser pulses in the nanosecond regime with solid extended targets, almost 100% of the incident laser radiation is absorbed [Garban-Labaune, 1982] whereas the fraction of absorbed energy decreases for shorter pulses [Broughton & Fedosjevs, 1993, Eidmann & Schwanda, 1991, Fedosejevs et al., 1990] and [Garban-Labaune, 1982, Price et al., 1995].

The absorption of a laser pulse is best at densities close to the critical density as stated above. Therefore, the larger the volume with almost critical density or in other words the larger the scale-length $L$, the better the absorption [Andreev et al., 2002, Krueer, 1988]. Long laser pulses with a pulse duration of $\tau_L$ leave the plasma enough time to evolve a large scale-length $L$ according to $L \propto v_{exp} \tau_L$ [Kodama et al., 1987, Nakano et al., 1996]. Thus long pulses are better absorbed than short ones. In addition, the absorption decreases with the laser intensity, for reasons stated in the next section. Leaving the laser pulse energy constant, short pulses have a higher intensity than
longer pulses, decreasing the absorption for short ones even more. Further on, the electronic heat conductivity, which is proportional to the electron density, decreases for a larger plasma scale-length. The plasma with large \( L \) is widespread and not in direct contact with the solid density plasma at the target surface. Short scale-length plasmas are always close to the high density region at the surface. Thus the heat conductivity is lower for large \( L \) and the plasma cools sufficiently slower and therefore radiates for a longer time, as stated by Ref. [Nakano et al., 1996].

The influences of all these different effects on the EUV conversion efficiency are hard to model. Therefore, only empirical values for the scaling of the x-ray conversion efficiency (CE) have been determined so far. According to Refs. [Altenbernd et al., 1997, Broughton & Fedosjevs, 1993, Eidmann & Schwanda, 1991, Kodama et al., 1987], the CE increases with increasing pulse length \( \tau_L \). The CE (for 3 nm radiation) was found by Ref. [Altenbernd et al., 1997] to be proportional to \( \tau_L^{0.3\pm0.1} \) within the laser pulse duration range of 100 fs to 2 ps, as long as the laser pulse energy was kept constant. A similar scaling for even longer pulses was observed by Ref. [Kodama et al., 1987]. Furthermore, the increase of the laser absorption with the scale-length is important for the use of prepulses. A weak pulse preceding the main pulse enhances the scale-length seen by the main pulse and enhances the absorption as well [Andreev et al., 2002].

### Intensity dependence of the collisional absorption

Turning to the intensity dependence of the absorption coefficient, a mathematical description can be developed. A higher laser intensity \( I_L \), achieved by either enhancing the laser pulse energy or reducing the pulse duration, increases the collective quiver motion \( v_{\text{quiver}} = eE_L/m_e\omega_L \propto \sqrt{I_L\lambda_L^2} \) of the electrons in the electric field of the laser \( E_L \). The effective mean electron velocity \( v_{\text{eff}} = \sqrt{v_e^2 + v_{\text{quiver}}^2} \) therefore increases. Since the cross-section \( \sigma_{ei} \) for electron-ion collisions decreases with \( v_{\text{eff}}^{-4} \) [Giulietti & Gizzi, 1998]

\[
\sigma_{ei} = \frac{4\pi Z^4}{m_e^2 v_{\text{eff}}^4} \tag{3.29}
\]

the electron-ion collision frequency \( v_{ei} \) (Eq. (3.20)) and in turn the absorption coefficient (Eq. (3.19)) decrease for higher laser intensities. The appropriate correction for the absorption coefficient (Eq. (3.19)), resulting in an effective, intensity-dependent absorption coefficient, is given by [Attwood, 1999, Giulietti & Gizzi, 1998]:

\[
k_{\text{eff}}(I_L) = k_{\text{im}} \left[ 1 + \frac{3}{2} \left( \frac{v_{\text{quiver}}}{v_e} \right)^2 \right]^{-1} = k_{\text{im}} \left[ 1 + I_L \left( \frac{3}{2 e n_{\text{crit}} k_B T_e} \right) \right]^{-1} \tag{3.30}
\]

As the intensity gets higher, the electron-ion collisions become less frequent, decreasing the absorption, but an additional effect reduces the absorption as well. When the rate at which the electrons gain energy from the laser (\( \propto v_{ei} v_{\text{quiver}}^2 \)) becomes larger than the rate at which the electrons can thermalize with each other (\( \propto v_{ee} v_e^2 = Z v_{ei} v_e^2 \)), there will be a deficit of slow electrons
The collisional absorption decreases, since mainly slow electrons account for it. The transition from collisional to collisionless effects is, therefore, given by [Attwood, 1999, Giulietti & Gizzi, 1998]:

\[
\frac{v_{\text{e}}v_{\text{e}}}{v_{\text{quiver}}} > Z \frac{v_{\text{e}}v_{\text{e}}}{v_{\text{e}}} \Rightarrow Z^{-1} \left( \frac{v_{\text{quiver}}}{v_{\text{e}}} \right)^2 = \frac{I_L}{Z c n_{\text{crit}} k_B T_e} = 2.1 \times 10^{-13} \frac{I_L [\text{Wcm}^{-2}] \lambda^2 [\text{µm}]}{Z T_e [\text{eV}]} > 1 \tag{3.31}
\]

For parameters appropriate for the EUV measurements \((T_e \sim 150 \text{ eV}, Z \sim 5 \text{ and } \lambda = 0.8 \text{ µm})\) the collisional absorption is no longer the dominant process for intensities \(> 5 \times 10^{15} \text{ Wcm}^{-2}\). The temperature in the plasma rises so quickly during the first part of the laser pulse that collisions become ineffective for the longest time of the interaction. Instead, collisionless absorption mechanisms become more important [Gibbon & Förster, 1996], which will be (briefly) considered next.

### 3.2.3 Collisionless absorption mechanisms

There are a number of collisionless processes providing efficient absorption at intensities exceeding \(10^{15} \text{ Wcm}^{-2}\). The Brunel absorption [Brunel, 1987] and the anomalous skin effect are two mechanisms predominant in very steep density gradient plasma \((L \ll \lambda_L\), see Eq. (3.17)) which was not the case in the reported experiments. A mechanism demanding a certain plasma scale-length \(L\) is the resonance absorption. This is the most prominent collisionless mechanism, and it is also the dominant collisionless process in the laser-plasma interactions to be considered for the EUV experiments. In the standard picture, an obliquely incident p-polarized laser field excites a plasma wave at the critical density. This wave, which grows over a number of laser periods, will eventually be damped either by collisions and collisionless Landau damping at lower intensities or by wave breaking at higher intensities [Gibbon & Förster, 1996, Kruer, 1988]. The main difference to the collisional absorption is the fact that the laser couples its energy to the collective motion of the plasma wave, not to single electrons independently, even if both processes finally heat the surrounding material by collisions.

A laser beam impinging onto the target under an angle of \(\theta\) will not reach the critical density since the refractive index of plasma is below 1, and the laser beam is refracted from higher to lower electron densities. Thus, the highest density seen by the laser is \(n_e(\theta) = n_{\text{crit}} \cos^2(\theta)\) [Giulietti & Gizzi, 1998]. Therefore, the electric field of the laser has to tunnel to the critical density to excite the plasma wave. On the other hand, the electric field component of the laser which drives the plasma waves decreases for steeper angles of incidence (smaller \(\theta\)). Therefore, very large and very small \(\theta\) lead to a low coupling between the laser and the plasma wave. This leads to an optimal angle of incidence \(\theta_{\text{max}}\), for which the absorption becomes maximum depending on
3.2. Laser-plasma interaction

the scale-length [Fedosejevs et al., 1990, Giulietti & Gizzi, 1998]:

$$\sin(\theta_{max}) \approx 0.8\sqrt[3]{\frac{\lambda L}{2\pi L}}$$  \hfill (3.32)

This equation states that the optimum angle of incidence approaches normal incidence for large scale-length. It can also be shown that the absolute value of the absorption coefficient increases with increasing scale-length [Gauthier, 2001]. Therefore, by applying a prepulse and thereby enlarging $L$, the resonance absorption for the droplet targets increases, since the laser beam is predominantly impinging under normal or close to normal incidence onto a droplet. The theoretically predicted maximum absorption of laser radiation obtained by resonance absorption is however limited to $\sim 50\%$ [Forslund et al., 1975]. Just like in the case of collisional absorption, things become more complicated when collisionless absorption of realistic plasma profiles has to be calculated. Such problems can only be solved by computer simulations [Gibbon & Bell, 1992], yielding absorptions up to 80% in agreement with experiments [Bastiani et al., 1997, Fedosejevs et al., 1990, Teubner et al., 1996].

These numerical simulations show that resonance absorption predominantly heats fast electrons while the energy gain of the slow ones is low. This asymmetry yields in a non-thermal hot electron population, so-called supra-thermal electrons [Giulietti & Gizzi, 1998]. The fraction of absorbed laser energy converted to supra-thermal electrons depends on various parameters and can only be determined by experiments [Bastiani et al., 1997, Giulietti & Gizzi, 1998, Yu et al., 1999] and numerical simulations [Gibbon, 1994]. Typical values range between 1% and 20%. The temperature of the hot electrons $T_{hot}$ can be calculated by the following relation

$$T_{hot} = 3.2 \times 10^{-5} \sqrt[3]{I_L [\text{Wcm}^{-2}] \frac{\lambda}{L} [\mu\text{m}^2]} \text{keV}$$  \hfill (3.33)

which was obtained by using particle-in-cell simulations [Gibbon, 2002, Gibbon & Bell, 1992].

Also, laser light travelling within an extended region ($L \gg \lambda_L$) of underdense plasma produces electron and ion plasma waves by parametric instabilities. The damping of these waves result in supra-thermal electrons as well [Giulietti & Gizzi, 1998]. In particular, Raman instabilities and two-plasmon decay prevail for the laser parameters of interest. The presence of these waves was experimentally observed for 1 ps pulses and prepulse delays $> 100$ ps ($L > 1$). The quantized plasma oscillations, plasmons, typically carry half the laser photon energy $\frac{1}{2}\hbar\omega_L$. Thus, during the parametric interaction of the laser light with the plasmons, new photons with an energy of $\frac{3}{2}\hbar\omega_L$ are created [Giulietti & Gizzi, 1998, Veisz et al., 2002]. For a Ti:sapphire laser wavelength of 800 nm the 3/2 harmonic is expected at 533 nm, which was indeed observed as intense green emission from the laser-plasma interaction region.
3.2. Laser-plasma interaction

3.2.4 Laser-plasma interactions at ultra high intensities

It was already mentioned in the last section that at laser intensities of several times $10^{15}$ W cm$^{-2}$ a certain fraction of electrons is accelerated by the laser field to energies beyond the temperature of the mass of electrons. However, only a few percent of the electrons are suprathermal. This changes drastically as the intensity is increased towards $10^{19}$ W cm$^{-2}$. The higher the intensity of the laser pulse, the higher is the fraction of laser energy which is converted into hot electrons. Up to 20-30% [Malka & Miquel, 1996, Wharton et al., 1998, Wilks et al., 2001] for intensities around $10^{19}$ W cm$^{-2}$ or even 40% [Snavely et al., 2000] for $3 \times 10^{20}$ W cm$^{-2}$ of the laser energy is transferred to the suprathermal electron population. The temperature of these hot electrons increases up to several MeV, according to the ponderomotive scaling [Schwoerer et al., 2001, Wilks et al., 1992]:

$$T_{\text{hot}} \simeq 0.511 \left( \sqrt{1 + \frac{I_L [\text{W cm}^{-2}] \lambda_L^2 [\mu m^2]}{1.37 \times 10^{18}}} - 1 \right) \text{MeV} \quad (3.34)$$

The idea behind this scaling is that the electrons are accelerated by the magnetic field of the laser pulse, that is generally neglected in standard electro-dynamics. However, for the enormous intensities and thus large field strength in a ultrahigh intensity laser pulse, electrons can be pushed to energies of several MeV by the magnetic field of the laser light. The rather simple equation is, thus, in good agreement with several experiments for intensities $> 10^{17}$ W cm$^{-2}$ [Gibbon, 2002]. Note, that for intensities exceeding $\sim 2 \times 10^{18}$ W cm$^{-2}$, at $\lambda_L = 0.8 \mu m$, the hot electron temperature is higher than the energy related to the rest mass of the electron and the plasma physics becomes relativistic.

These hot electrons collide eventually with the ions and the surrounding matter. The electron trajectory is bent during such a collision, giving rise to bremsstrahlung emission. The spectral intensity $W_{\text{brems}}$ of the bremsstrahlung for a Maxwellian non-relativistic electron distribution with a temperature of $T_e$ is given by [Giulietti & Gizzi, 1998]:

$$W_{\text{brems}}(\hbar \omega) = \frac{32 \pi}{3} \sqrt{\frac{2 \pi}{3k_B T_e m_e}} Z e^6 n_e^2 \exp \left( -\frac{\hbar \omega}{k_B T_e} \right) \quad (3.35)$$

Having electrons with temperatures of several hundred keV bremsstrahlung photons with energies far above 1 MeV can be produced. Thus, focussing an intense laser pulse onto a target, visible light ($\lambda_L = 800$ nm) can be converted into MeV radiation far beyond the energy of atomic transitions. This equation is a good approximation for the experiments described in Sect. 6.2. Calculations for the relativistic regime demanded for higher laser intensities can be found in Ref. [Ewald et al., 2002].

Unfortunately, the spectrally integrated radiation emission is not proportional to $T^4$ as for Planck radiation of a black body, but only increasing with $\sqrt{T_e}$, however, it is the only way to produce such high energetic photons within a laser-plasma.
3.2. Laser-plasma interaction

3.2.5 MEDUSA - simulation of laser-plasma dynamics

It is almost impossible to treat the laser-plasma interaction, continuously varying in time and space, by the analytical formulas discussed above. The help of a simulation program was needed in order to obtain the time- and space-dependent electron and ion density \( n_e(x,t) \) and \( n_i(x,t) \) as well as the electron and ion temperature \( T_e(x,t) \) and \( T_i(x,t) \) respectively, and the fluid velocity \( v(x,t) \). Part of the output of such a simulation can be seen in Fig. 3.1. The simulation of the laser-plasma interaction was performed with the computer code MEDUSA [Christiansen et al., 1974, Giulietti & Gizzi, 1998], which is a one-dimensional Lagrangian code. The program simulates the laser absorption and the subsequent plasma dynamics. It solves the hydrodynamic equations for the continuity of mass and charge and the equations of motion, dominated by the thermal pressure. Also, thermodynamic equations are included to simulate the heating of the plasma. The ions are always treated as ideal gas whereas for the electrons, a Thomas-Fermi equation of state [Salzmann, 1998] was used to include quantum mechanical effects. The ionization dynamics was computed by the Saha equation. It is also possible to switch to the more accurate average atom model (e.g. [Salzmann, 1998]) which however requires about 100 times more CPU time. Fortunately, the results of both models do not differ substantially for the simulated plasma parameters. The heat conduction is calculated using the Spitzer-Härm conductivity in which the heat flux is taken to be proportional to the temperature gradient. For high temperature gradients, which typically occur for short pulses, a flux limiter can be set. Relying to experimental results (e.g. [Hauer et al., 1984]) the flux limit was set to 0.1.

![Figure 3.1](image)

**Figure 3.1:** The ion temperature (left) and the electron density (right) are plotted as function of the distance from the target for various times after the laser pulse \( (E_L = 20 \text{ mJ}, \tau_L = 100 \text{ ps resulting in } 6 \times 10^{13} \text{ Wcm}^{-2}) \). The collisional laser absorption is calculated according to Eq. (3.19). The collisionless absorption however is not directly calculated by the program, but has to be specified. One can force MEDUSA to deposit a certain fraction of the laser energy (typically 20%) at the critical density. MEDUSA does not simply deposit 20% of the incident laser energy, but rather calculated the ab-
sorption within the undercritical plasma. Thus the fraction refers only to the amount of laser energy reaching the critical density. Since this ‘collisionless’ absorbed energy produces supra-thermal electrons, it is possible to define a fraction of the collisionless absorbed energy which is converted into supra-thermal electrons. This fraction is typically set to about 10-20% for ps and fs laser pulse durations. Thus, in agreement with experiments [Bastiani et al., 1997, Eidmann et al., 1983, Giulietti & Gizzi, 1998], 2-4% of the incident laser energy is converted to supra-thermal electrons at intermediate laser intensities. The influence of these two parameters is discussed in some detail in Chap. 5. The heating induced by shock waves [Salzmann et al., 1983] which are launched by the ablating surface material, is taken into account as well.

Further input parameters are the pulse duration and intensity for up to four independent Gaussian laser pulses as well as the delays between the pulses. Furthermore, the atomic number and atomic mass of the target material have to be given. The preferred geometry has to be chosen. Besides the planar geometry, which was used in most calculations, in principal, the code can calculate a spherically symmetric plasma expansion. This would be adequate to model the laser irradiated water droplet, but the intensities demanded for the simulation of the experiments were too high for the spherical calculation mode.

The simulation results obtained with MEDUSA are presented in Chap. 5.

3.3 Atomic physics in plasmas

Hot plasmas are a rather unfriendly environment. Enormous electric fields, frequent collisions and highly energetic photons alter the atomic physics of ions within a plasma compared to the physics of isolated ions. Energy levels, line width, population densities of levels and many more parameters rely on the plasma conditions. The dependencies that are important for the understanding of the EUV production are reviewed within this section.

3.3.1 Thermodynamic equilibria

In complete thermodynamic equilibrium (TE), all particles, ions and electrons, and the radiation are all in mutual equilibrium, all having equal temperatures. The Planck equation (Eq. (3.36)) describes the radiation, the Boltzmann equation (Eq. (3.39)) the ratio of excited levels and the Maxwell distribution (Eq. (3.2)) holds for the velocity of the particles. Most models are somehow based on the thermodynamic equilibrium, however the latter is rarely found in laboratory plasmas. Nevertheless, the TE is quite useful to get rough estimates on plasma temperatures and radiation yields to be expected by the advanced theory and the experiment. The Planck equation for the
energy density of the radiation is given by [Thorne et al., 1999]:

\[ \rho(T, \omega) d\omega = \frac{2\hbar \omega^3}{\pi c^3} e^{\hbar \omega/k_B T} - 1 d\omega \] (3.36)

This distribution has a maximum at a particular photon energy \( \hbar \omega_{\text{max}} \) for a given temperature \( k_B T \).

The relation between both quantities is given by Wien’s law [Attwood, 1999]:

\[ \hbar \omega_{\text{max}} = 2.82 \times k_B T [\text{eV}] \] (3.37)

This equation states that a plasma with a temperature of 34 eV radiates in a way that the maximum of the emitted radiation is at 95 eV equivalent to 13 nm.

To get a rough estimate of the conversion efficiency of laser light into a narrow region around 13 nm, one can integrate \( \rho(T, \omega) \) over this region and normalize the integral with the total emitted radiation. This wavelength region of interest is the reflection bandwidth of the Mo-Si mirrors which about 0.4 nm (see Sect. 2.3.3). It turns out that the plasma temperature at which the 13 nm radiation, normalized by the total emitted radiation energy, reaches a maximum is not found at 34 eV but rather at 24 eV. This can be explained by the fact that the distribution for 34 eV is broader compared to the emission characteristic for 24 eV. Thus the total energy is distributed over a larger wavelength region, leaving less energy for a specific wavelength. At an ion temperature of 24 eV 1.8% of the totally emitted radiation is radiated into the small region around 13 nm. Relying on laser-plasma simulations, about 40% of the laser radiation is absorbed by the plasma and furthermore, only 50% of the absorbed energy contributes to the thermal energy (whereas the remaining 50% are consumed for the plasma expansion) one ends up with about 0.4% conversion efficiency. This value is indeed in the experimentally obtained range of conversion efficiencies. The prediction that the best conversion can be expected at a plasma temperature of 24 eV is also quite close to the results of advanced calculations (Sect. 3.3.3) and the experiment (Sect.4.2).

In fact, the condition that the radiation is in equilibrium with the particles, in other words that the plasma emits a Planckian continuum, is not fulfilled since the radiation of the plasma is emitted as lines and not as continuum (see e.g. Fig. 4.1). However, the Planckian radiation distribution sets an upper limit to the radiation emitted by line emission. It envelopes indeed any radiation distribution as long at least the Boltzmann distribution is satisfied [Thorne et al., 1999]. Thus, the CE obtained for the Planckian distribution can be taken as upper limit to the experimentally determined CE.

By making assumptions that are much closer to the experimental conditions, it is nevertheless possible to describe the ratio of excited levels of an ion in an analytical form. There are basically two equilibria to be considered: The local thermodynamic equilibrium (LTE) and the corona equilibrium.
The local thermodynamic equilibrium (LTE)

The local thermodynamic equilibrium is an approximation to the physical environment present in high density plasmas where the radiation is not in equilibrium with the plasma. Comparing the mean free path of photons and electrons, it turns out that the photons reach 100 to 10^4 times further than the electrons with the same energy [Salzmann, 1998]. This is true for low Z-plasmas and temperatures up to 1 keV - the region of interest in this work. Therefore, the µm-sized laser-produced plasma is expected to be almost transparent in a certain range of frequencies. Radiation escapes from the plasma and is, therefore, not in equilibrium with the particles (electrons and ions). In general, upper levels can be populated by absorption of ambient radiation or by energy transfer due to electron collisions. In order that the shortfall of the radiation excitation does not matter, the collisional excitation due to electron impact has to be dominant. Collisions dominate for electron densities of [Giulietti & Gizzi, 1998, Thorne et al., 1999]:

\[ n_e [\text{cm}^{-3}] \gg 1.7 \times 10^{14} \sqrt{T_e [\text{eV}] \Delta E^3 [\text{eV}]} \]  

(3.38)

To fulfill this inequality, \( n_e \) has to be greater than \( \sim 10^{21} \text{ cm}^{-3} \) for a temperature of 24 eV and the 13 nm transition (\( \Delta E \sim 100 \text{ eV} \)). As it will turn out later, the dominant part of the 13 nm radiation is indeed emitted at this lower boundary of the electron density.

Neglecting the contribution of the radiation, the population of excited levels can be calculated, within the same ionization state, can be calculated without knowing detailed information about cross-sections and transition probabilities. This is a very useful feature of the LTE. As pointed out, the levels are populated by electron-ion collisions. By considering a two-level atom (\( l \) denoting the lower level and \( u \) the upper one), the collisional transition rate coefficients are expressed as \( C_{l \rightarrow u} \) and \( C_{u \rightarrow l} \) for the collisional excitation and de-excitation, respectively. The knowledge of the transition rates is not essential, since taking the fraction of the populations of the upper \( n_u \) and the lower \( n_l \) level, all details cancel out and the Boltzmann distribution is left over: [Thorne et al., 1999]

\[ \frac{n_u}{n_l} = \frac{C_{u \rightarrow l}}{C_{l \rightarrow u}} = \frac{g_u}{g_l} e^{-\Delta E/k_B T} \]  

(3.39)

\( g_{l,u} \) are the statistical weights of the particular level. Taking the numbers for the upper level of the 13 nm transition (4d) and (2s) ground-state (see Fig. 3.5 and Tab. 3.2) and a temperature of 24 eV, about 6% of the ions are in the upper excited level. Since only about \( \frac{1}{4} \) of the ions are in the ground state of the considered O^{5+} ionization stage, calculated by the Saha equation [Salzmann, 1998], the simple form of a two-level LTE calculation predicts that about 2% of the oxygen ions are in the upper 13 nm level. Certainly, this is an upper limit since in real life, many more levels are involved, and detailed rate coefficient models are necessary (see next section).
3.3. Atomic physics in plasmas

The corona equilibrium

The second equilibrium to be considered is the coronal equilibrium, which is applicable to plasmas with a lower electron density than the LTE\(^2\). The assumptions are that the dominating part of the excitation is still due to electron collisions but the density is low enough to ensure that the de-excitation is dominated by spontaneous radiative decay and not by collisional de-excitation. Now the detailed information about the spontaneous decay (Einstein coefficient) \(A_{u\rightarrow l}\) and \(C_{l\rightarrow u}\) are demanded since they do not cancel out and the two-level corona equilibrium results in [Salzmann, 1998, Thorne et al., 1999]:

\[
\frac{n_u}{n_l} \propto n_e \frac{A_{u\rightarrow l}}{C_{l\rightarrow u}}
\] (3.40)

However, \(A_{u\rightarrow l}\) and \(C_{l\rightarrow u}\) are constant for a given transition and plasma temperature\(^3\). Therefore, the population of the upper level increases with the electron density, implying more collisions, until LTE limit is reached. The LTE limit is given by the Boltzmann distribution (Eq. (3.39)) which gives the highest possible density of excited states in an equilibrium.

Summarizing the predictions of the TE, LTE and the corona equilibrium, the population of the upper 13 nm level will be highest at temperatures of about 24 eV. Furthermore, the relative population density increases steadily with the electron density until it saturates at the LTE limit of \(\sim 2\%\) at an electron density of about \(n_e \sim 10^{21} \text{ cm}^{-3}\). These results, achieved by using very simple models, agree surprisingly well with the much more advanced calculations presented in the next section (see Fig. 3.2).

3.3.2 Rate equations

To get a more accurate dependence of the population density of a specific level on the ion temperature and electron density, a detailed model of the atomic physics has to be applied to the problem. It is necessary to sum over all involved levels, to include populating and depopulating ionization and recombination processes by photons and electron collisions inducing transitions to the next ionization stages as well as excitation and de-excitation within the same ionization level. The basic scheme of such a rate equation model is as follows [Kawachi & Fujimoto, 1995, Salzmann, 1979]:

\[
\begin{align*}
\frac{d n_i}{dt} &= \sum_{j<i} n_e n_j C_{j\rightarrow i} - \left\{ \sum_{j<i} F_{i\rightarrow j} + \sum_{j>i} C_{i\rightarrow j} \right\} n_e + \sum_{j<i} A_{i\rightarrow j} n_i + \\
&\sum_{j>i} \left\{ [F_{j\rightarrow i} n_e + A_{j\rightarrow i}] n_j - S_{i} n_e n_i + [\alpha_i n_e + \beta_i + \gamma_i] n_e n_{O \text{ VII}} \right\}
\end{align*}
\] (3.41)

\(^2\)For a detailed discussion of the parameter ranges in which the coronal equilibrium is valid and for the introduction of various correction terms to the simple formula (3.40) see Ref. [Salzmann, 1998]

\(^3\)The actual values of A and C will be calculated in Sect. 3.3.5
The population density $n_i$ of the level $i$ is connected with the spontaneous transition probability $A_{i\rightarrow j}$ from $i$ to $j$. $C_{i\rightarrow j}$, $F_{j\rightarrow i}$ and $S_i$ are the rate coefficients for electron impact excitation, de-excitation\(^4\) and ionization, respectively. So far only transitions within the lithium-like ionization level were considered. To include the recombination transitions from helium-like ions to lithium-like ions $\alpha_i$, $\beta_i$ and $\gamma_i$ denoting the rate coefficients for three-body recombination, radiative and dielectronic recombination are included. $n_{\text{O VII}}$ and $n_{\text{O VI}}$\(^5\) (in Eq. (3.42)) represent the ground-state of helium-like and lithium-like ion density, respectively. All these coefficients have to be either calculated by approximation formulas or taken from a database. A more detailed description is beyond the scope of this work and can be found e.g. in [Salzmann, 1998].

The set of equations can either be solved time-dependent, demanding the time-dependence of the ion temperature or the electron density (see Sect. 3.3.4), or a steady state situation can be assumed for the so-called collisional radiative steady state (CRSS) model (see Sect. 3.3.3). In the second case, the population density of the particular levels can be calculated as function of the ion temperature and the electron density. Ionization and recombination processes are balanced causing the time derivative of $n_i$ to vanish, and the coupled set of differential equations is reduced to a set of linear equations with the general form:

$$n_i = R(n_e, T)_i; n_e n_{\text{O VII}} + I(n_e, T)_i; n_e n_{\text{O VI}} \quad (3.42)$$

summarizing the various coefficients appearing in Eq. (3.41) in a recombination coefficient $R(n_e, T)_i$ and a ionization coefficient $I(n_e, T)_i$, both depending on the electron density and temperature.

### 3.3.3 The collisional radiative steady state code

A computer program, similar to the one described in [Salzmann & Szichman, 1987], was used to calculate the population densities $n_i$ of about 50 levels as function of the electron density and temperature for all ionization stages of oxygen according to Eq. (3.42). Among other levels, the relative population density of the O\(^{5+}\)(4d) level, the upper level of the 13 nm transition, was computed for a series of electron densities between $10^{18}$ and $7 \times 10^{21}$ electrons/cm\(^{-3}\), and for temperatures from 10 eV to 100 eV. Results of the code are shown in Fig. 3.2.

For every given electron density, there is a temperature at which the O\(^{5+}\)(4d) density has a maximum. These maxima are plotted in Fig. 3.2 as function of the electron density. The population density of the 4d level, relative to the total ion density, increases monotonically with the electron density from $\sim 10^{-4}$ at an electron density $n_e = 10^{18}$ cm\(^{-3}\) to $\sim 10^{-2}$ at $n_e = 10^{21}$ cm\(^{-3}\), as expected by the coronal equilibrium calculation. But at even higher densities, when the LTE conditions start to prevail in the plasma, the relative density decreases accordingly, because the

\(^4\)For reasons of clarity the excitation and de-excitation get different symbols.

\(^5\)The nomenclature was borrowed from astronomy.
collisions distribute the bound electron population among more states. The temperatures at which the maxima occur are around 20 eV at the low density limit and increase smoothly to $25 - 30$ eV when LTE conditions are reached. The overall maximum of the curve is slightly above 1% when the electron density is $2 \times 10^{21}$ cm$^{-3}$ and the temperature is $\sim 26$ eV. The maximum population density is only a factor of 2 lower than the simple two-level LTE estimation in Eq. (3.39) predicted. The population density has no sharp maximum as function of the temperature. It decreases to 80% of the maximum within $\pm 6$ eV.

Since the emitted 13 nm radiation is proportional to the population density of the O$^{5+}(4d)$ level, Fig. 3.2 states that the major part of the 13 nm radiation originates from a plasma with a density of $n_e = 10^{21}$ cm$^{-3}$ and a temperature of $T = 25 - 30$ eV.

### 3.3.4 Time-dependent rate equation model

It is also possible to solve the set of equations in Eq. (3.41) retaining the time-dependence. The simulation code used for this calculation is described in [Ralchenko & Maron, 2001]. The code can only calculate the temporal behavior of the various levels when the plasma evolution is predetermined. Therefore, the time-dependent ion temperature at a particular electron density was needed as input. These data were provided by the MEDUSA simulation results for 1 ps laser pulses with an energy of 20 mJ preceded by a 2 mJ prepulse (upper part of Fig. 3.3). The lower part of Fig. 3.3 shows the temporal evolution of the various ionization levels starting from neutral oxygen up to fully stripped ions.

The oxygen ions are quickly ionized up to fully ionized oxygen, subsequently recombining when the plasma cools down. O$^{6+}$ and O$^{5+}$ are dominating the ionization levels for almost 1 ns, while O$^{8+}$ and O$^{7+}$ last only for a short time due to the high ionization potential (see Tab. 3.1). Indeed, a strong emission at 2.2 nm was observed in the experiments, resulting from O$^{6+}$ and O$^{7+}$ to some extend [Kelly]. The population of the O$^{5+}$ ions peaks at an ion temperature of 32 eV, slightly
higher than expected from the CRSS code. The time-dependency indeed alters the population distribution in contrast to the steady state model, however, only slight changes have been observed. The subsequent recombination to neutral oxygen is caused by the fact that the input temperature data are limited to a particular value of the density, thus neglecting the plasma expansion. The expanding low density plasma is still highly ionized even for \( \mu s \) after the laser-plasma interaction, as measurements indicated (Sect. 4.5).

![Figure 3.3:](image)

In addition, the population density of the \( \text{O}^{5+} \) (4d) was calculated by the time-dependent model. As expected the 4d level is populated by recombining \( \text{O}^{6+} \) as can be seen in Fig. 3.4 (solid lines). The more interesting question is to look at the population density dynamics of the \( \text{O}^{5+} \) and \( \text{O}^{6+} \) ions at different electron densities. Fig. 3.4 shows the population density for an electron density of \( 10^{21} \text{ cm}^{-3} \) (solid lines), like in Fig. 3.3, and at a lower density of \( 5 \times 10^{19} \text{ cm}^{-3} \) (dashed lines). As expected from Fig. 3.2 the \( \text{O}^{5+} \) (4d) contributions for the lower density decreased by a factor of 6 compared to \( n_e = 10^{21} \text{ cm}^{-3} \). In addition, the recombination occurs at later times since collisions are less frequent in the lower density plasma. Later times in turn imply a lower ion temperature. The maximum of the \( \text{O}^{5+} \) (4d) population occurs at \( \sim 20 \text{ eV} \), again consistent with Fig. 3.2. The completely different behavior of the population density for various electron densities shows again the important impact of the electron density on the atomic physics. The optimization of only the ion temperature, like for a Planck emitter, is not sufficient to get efficient radiation from an ion in a hot plasma.
3.3. Atomic physics in plasmas

3.3.5 Transition probabilities of O\textsuperscript{5+} lines

To get information from the measured spectra about the temperature and the electron density of the region emitting 13 nm radiation, knowledge about the transition probabilities of the various lines and, in addition, information about absorption of the radiation on its way out of the plasma are essential.

The number of photons emitted per unit time and cubic centimeter in the transition from the upper $u$ state to the lower $l$ state is given by [Salzmann, 1998]:

$$P_{u\rightarrow l} = n_u A_{u\rightarrow l}$$

(3.43)

$P_{u\rightarrow l}$ is proportional to the density of excited upper states $n_u$ and to $A_{u\rightarrow l}$, the Einstein coefficient for the spontaneous radiative decay which is related to the oscillator strength $f_{u\rightarrow l}$ and the energy difference of the transition $\Delta E_{u\rightarrow l}$ in the following way [Huba, 1994, Salzmann, 1998]:

$$A_{u\rightarrow l} = \frac{2c}{(\hbar c)^2} \frac{e^2}{m_e c^2} (\Delta E_{u\rightarrow l})^2 f_{u\rightarrow l} = 4.3 \times 10^7 (\Delta E_{u\rightarrow l}[\text{eV}])^2 f_{u\rightarrow l} \text{ s}^{-1}$$

(3.44)

c is the speed of light and $m_e$, $e$ are the electron mass and charge, respectively. The oscillator strength\textsuperscript{6} is closely connected to the wave functions of the respective state and can be calculated by first principles, however, the database of the CRSS code already provides $f_{u\rightarrow l}$ for the lines of consideration. By inserting the numbers from table 3.2 for the 13 nm transition one gets $A_{u\rightarrow l} = 4.6 \times 10^{10} \text{ s}^{-1}$. The reciprocal value of the Einstein A coefficient is the radiative life-time of the upper level. Without collisions, an excited electron on the O\textsuperscript{5+}(4d) level would decay under emission of a 13 nm photon within 20 ps.

\textsuperscript{6}Sometimes, the equation for the Einstein $A$ coefficient can be found including the statistical weights $g_{u,l}$. In this case, $f$ has to be taken as the excitational oscillator strength: $f_{l\rightarrow u} = -g_u f_{u\rightarrow l}/g_l$ [Salzmann, 1998].
As already pointed out in Sect. 3.3.1, the influence of the electron collisions on the transitions cannot be neglected for high electron densities. The rate coefficient for the collisional excitation \( C_{l\rightarrow u} \) can be expressed in the simplest form by van Regemont’s formula: [Huba, 1994, Salzmann, 1998]

\[
C_{l\rightarrow u} = 1.6 \times 10^{-5} \frac{f_{u\rightarrow l} \langle g(u,l) \rangle}{\Delta E_{l\rightarrow u} \sqrt{T_e}} \exp \left( -\frac{\Delta E_{l\rightarrow u}}{T_e} \right) \text{cm}^3/\text{s}
\]  

(3.45)

where energy and temperature are given in eV. \( \langle g(u,l) \rangle \) is the Gaunt factor that accounts for quantum mechanical corrections. \( \langle g(u,l) \rangle \) is generally taken to be \( \sim 0.2 \) for ions [Huba, 1994]. The excitations per second are given by \( n_e C_{l\rightarrow u} \), while the reciprocal of this value represents the mean time between collisional excitations. Inserting the numbers for the 13 nm transition, the collisional excitations occur in average every 30 ps for \( T_e = 30 \) eV and \( n_e = 10^{21} \text{cm}^{-3} \). The collisional de-excitation coefficient \( C_{u\rightarrow l} \) is calculated taking the Boltzmann equilibrium [Huba, 1994]:

\[
C_{u\rightarrow l} = g_l/g_u \exp(+\Delta E_{u\rightarrow l}/T_e) C_{l\rightarrow u}.
\]

Thus, de-excitations happen on average about 10 ps after the excitation. As already inferred by Eq. (3.38), the collisional de-excitation becomes dominant at \( n_e \sim 10^{21} \text{cm}^{-3} \). Indeed, the collisional de-excitation is twice as frequent as the spontaneous radiative decay at this electron density, thus justifying the LTE assumptions. The effective life-time of the upper state is, therefore, influenced by radiative and collisional de-excitation and can be given by

\[
\tau_{\text{eff}} = (A_{u\rightarrow l} + n_e C_{u\rightarrow l})^{-1}.
\]

Concentrating on the ratio of two radiative transitions, the influence of the electron collision and the numerical constants cancel out and

\[
\frac{P_{u\rightarrow l}}{P'_{d'\rightarrow l'}} = \frac{n_u(\Delta E_{u\rightarrow l})^2 f_{u\rightarrow l}}{n'_u(\Delta E'_{d'\rightarrow l'})^2 f_{d'\rightarrow l'}}
\]  

(3.46)

is left over. \( \Delta E_{u\rightarrow l} \) and \( f_{u\rightarrow l} \) are known (see table 3.2), and \( n_u \) can either be taken from the CRSS code as function of the electron density and the plasma temperature, or the ratio of \( n_u \) to \( n'_u \) can
be approximated by the Boltzmann distribution (Eq. 3.39)). Therefore, knowing the theoretical line ratio, one could in principle determine \( T_i \) and \( n_e \) by comparing the theoretical CRSS line ratios with the experiment. Unfortunately, the line ratios are not strongly density-dependent in the region of interest (see Fig. 4.2).

Looking at the measured line ratio (see Sect. 4.2), the 11.6 nm and the 15.0 nm lines are very weak compared to the 13 nm line compared to the ratios obtained by Eq. 3.46. The reason is found in the differing absorption of the lines. The propagation of the radiation within plasma is, therefore, reviewed in the next section.

3.3.6 Radiation transport

The radiation is attenuated by basically two different mechanisms while propagating in the plasma. First, the radiation is absorbed by bound-free (photoelectric) and free-free (inverse bremsstrahlung) transitions and attenuated by scattering. These are non-resonant mechanisms attenuating all lines of consideration by an approximately equal amount. Second, the radiation is reabsorbed by the reverse emission process - the resonance absorption. This absorption mechanism (bound-bound) is different for the lines, since their lower levels are not the same (see Fig. 3.5). The absorption coefficient \( \kappa_{l \rightarrow u} \), which is defined per unit length, depends (like the emission) on the oscillator strength \( f_{u \rightarrow l} \) [Salzmann, 1998].

\[
\kappa_{l \rightarrow u}(\omega) = \frac{2\pi^2 e^2 \hbar c}{m_e c^2} \frac{g_l}{g_u} n_i n_l f_{u \rightarrow l} L(\omega)
\]  

(3.47)

\( g_{u,l} \) are the statistical weights of the levels, and \( L(\omega) \) is the line shape that can be approximated by \( L \approx 2/\pi \Gamma \) for a Lorentzian line profile with \( \Gamma \) as the FWHM of the line [Salzmann, 1998]. The measured line shape, as will be shown in Sect. 4.1, is indeed a Lorentz-like line with a line width of 0.8 eV for the 13 nm line. The same line width was also used for the calculation of the absorption of the other lines. The ion density \( n_i \) and the population density of the lower level \( n_l \) again are taken from the CRSS output for the optimum EUV conditions, namely a temperature of 25 eV and an electron density of \( \sim 10^{21} \text{ cm}^{-3} \). \( \kappa_{l \rightarrow u} \) was thus calculated by inserting the values in Eq. (3.47) as shown in Tab. 3.2.

The 13 nm radiation can indeed escape from the plasma more easily than the 11.6 nm and the 15.0 nm radiation. To obtain the total attenuation of the radiation penetrating through the surrounding low density plasma, all contributions of the various densities have to be summed up. The electron density, and thus ion density, changes with the distance from the target (see e.g. Fig. 3.1). The further away from the initial target surface, the lower the ion density is, resulting in a smaller \( \kappa(x) \). A measure for the absorption by the plasma is the optical thickness \( \tau \) [Giulietti & Gizzi, 1998]:

\[
I_{\text{detector}} = I_0 e^{-\tau} \quad \text{with} \quad \tau = \int_0^{\text{detector}} dx \kappa(x)
\]

(3.48)
3.3 Atomic physics in plasmas

<table>
<thead>
<tr>
<th>$\lambda$ / nm</th>
<th>$\hbar\omega$ / eV</th>
<th>Lower level</th>
<th>Upper level</th>
<th>$g_l$</th>
<th>$g_u$</th>
<th>$f_{u\rightarrow l}$</th>
<th>$\kappa$/mm$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>11.6</td>
<td>106.8</td>
<td>$(1s^2)\ 2s$</td>
<td>$(1s^2)\ 4p$</td>
<td>2</td>
<td>6</td>
<td>0.11</td>
<td>14</td>
</tr>
<tr>
<td>13.0</td>
<td>95.3</td>
<td>$(1s^2)\ 2p$</td>
<td>$(1s^2)\ 4d$</td>
<td>6</td>
<td>10</td>
<td>0.12</td>
<td>0.35</td>
</tr>
<tr>
<td>15.0</td>
<td>82.6</td>
<td>$(1s^2)\ 2s$</td>
<td>$(1s^2)\ 3p$</td>
<td>2</td>
<td>6</td>
<td>0.45</td>
<td>56</td>
</tr>
<tr>
<td>17.3</td>
<td>71.6</td>
<td>$(1s^2)\ 2p$</td>
<td>$(1s^2)\ 3d$</td>
<td>6</td>
<td>10</td>
<td>0.67</td>
<td>2.0</td>
</tr>
</tbody>
</table>

Table 3.2: The table summarizes the important parameters for prominent EUV lines important for the analysis of the experiments. $\lambda$ is the wavelength and $\hbar\omega$ the photon energy of the respective transition. The nomenclature $(1s^2)$ denotes that the two inner shell electrons are in the s level during the transition. This is the case for all considered O$^{5+}$ transitions, and therefore, the term $(1s^2)$ will be skipped in the text. $g_{u,l}$ are the statistical weights of the upper and lower level. The oscillator strength $f_{u\rightarrow l}$ was taken from the CRSS code data base. Finally, the absorption coefficient $\kappa$ was calculated for $n_e = 1.4 \times 10^{21}$ cm$^{-3}$ and $T_i = 25$ eV (see text).

$\tau$ is the integration over all the absorption taking place from the origin of the radiation (position of about $n_{\text{crit}}$) to the detector. The radiation arriving at the detector was attenuated by a factor of $e^{-\tau}$, resulting from the absorption in the undercritical plasma. If $\tau \ll 1$, almost no absorption took place and the plasma is optically thin whereas $\tau \gg 1$ implies significant absorption, and the plasma is said to be optically thick.

The density is decreasing rapidly after a few tens of $\mu$m away from the target. Therefore, the optical thickness for the 13 nm line $\tau \ll 1$, and no opacity related corrections have to be taken into account. This is not the case for the 11.6 nm and the 15.0 nm lines. These are resonance lines since the lower level is the ground state of the ion thus, resulting in a higher absorption than the 13 nm line (see Tab. 3.2). The absorption coefficients determined by the experiments (see Sect. 4.2) are in good agreement with the calculated values.

3.3.7 Line broadening

It is interesting not just to look at line ratios but also to the line shape itself which is also influenced by the plasma parameters. In principle there are three different line broadening mechanisms to be considered.

The ultimate limit of each line width is given by the natural line width which is defined via the uncertainty principle [Huba, 1994]:

$$\Delta E \Delta t = \hbar \Delta \nu \Delta t \geq \hbar = 4.14 \times 10^{-15} \text{ eV s} \quad (3.49)$$

$\Delta t = A_{u\rightarrow l}^{-1}$ was already calculated with Eq. (3.44) to be about 20 ps. The resulting line width in terms of $(\lambda/\Delta\lambda)_{\text{nat}}$ is on the order of $2 \times 10^4$. Due to the thermal motion of the ions the line is also
broadened by the Doppler shift resulting in a Gaussian line shape. The Doppler width is given by [Huba, 1994]:

\[
\left( \frac{\lambda}{\Delta \lambda} \right)_{\text{Doppler}} = 1.3 \times 10^4 \sqrt{\frac{m_i}{T}}
\]

for \( m_i \) expressed in atomic mass units and \( T \) in eV. Considering oxygen ions \( (m_i = 16) \) and a plasma temperature of 30 eV, \( (\lambda/\Delta \lambda)_{\text{Doppler}} \) becomes 9500.

The third broadening effect which is dominating for high electron density plasmas (see Sect. 4.1) is the so-called Stark broadening. The electric field induced by electrons, zipping around the ion and colliding with it, changes the energy levels of the ion, in analogy to the Stark effect. The upper and lower level of the radiative transition change continuously their potential energy, thus the energy difference is not sharply defined during the emission and the line width increases. The effect is certainly related to the energy of the electrons and most of all on the electron density. The higher the density the more frequent are collisions.

Starting from Eq. (3.45), H. Griem developed a semiempirical formula to calculate the line broadening due to electron impact [Schriever et al., 1998a, Griem, 1968]:

\[
\left( \frac{\lambda}{\Delta \lambda} \right)_{\text{coll}} = 7.7 \times 10^{23} \frac{\sqrt{T_e [\text{eV}]} \times 1}{n_e [\text{cm}^{-3}]} \left\{ \frac{1}{l} |R^2|l + \frac{1}{u} |R^2|u \right\}
\]

where the electron temperature \( T_e \) and density have to be known as well as the matrix elements \( R^2 \) of the atomic transitions. The matrix elements can be given in a strict way only for hydrogen like ions:

\[
\langle l, u | R^2 | l, u \rangle = \frac{n^2}{2Z^2} \left\{ 5n^2 + 1 - 3\ell(\ell + 1) \right\}
\]

with the quantum numbers \( n, \ell \) for the upper and lower state. However, the equation can be used as good estimate for the present purpose by substituting the charge state \( Z \) by an effective charge state \( Z_{\text{eff}} \). Arguing for the 13 nm line, an electron in a 4d orbital, which is quite far away from the nucleus, in a lithium-like ion moves in an electric potential generated by the nucleus minus the two innermost electrons. Altogether this potential is similar to that generated by a hydrogen-like ion with a nuclear charge \( Z = 8 - 2 = 6 \). A more advanced method, relying on empirical values and allowing that the screening of the inner electrons might not be unity, is the introduction of a screening factor \( \sigma \) defined by: \( Z_{\text{eff}} = 8 - \sigma \) [Salzmann, 1998, Thorne et al., 1999]. Using the empirical energy eigenvalues \( E_n \) for the fivefold ionized oxygen, \( Z_{\text{eff}} \) can be calculated according to:

\[
E_n = \frac{Z_{\text{eff}}^2}{n^2} \times 13.6 \text{ eV} \quad \Rightarrow \quad Z_{\text{eff}} = n \sqrt{\frac{E_n [\text{eV}]}{13.6}}
\]

\( E_n \) is given by the ionization energy minus the excitation energy of the respective level (see Fig.3.5). \( Z_{\text{eff}} \) turns out to be rather close to 6 (\( \sigma_{4d} = 1.96 \) and \( \sigma_{2p} = 1.91 \)), as expected. This consistency with the hydrogen model justifies its use. Finally, the values of \( Z_{\text{eff}} \) for the upper and the lower level are used in Eq. (3.52) rather than just \( Z \).
4 Results of the EUV experiments

The aim of this chapter is to present the measurements which were performed in order to investigate the influences of various parameters on the EUV production, to find trends and to develop scaling laws. The understanding of the various contributing processes shall finally result in the precise optimization of the EUV yield. The EUV emission of fivefold ionized oxygen $\text{O}^{5+}$ was measured for different laser pulse durations ranging from fs to ns regime and for various laser pulse energies. Prepulses were also introduced, to change the preplasma density distribution interacting with the main laser pulse. Diverse types of targets were investigated, changing the droplet size, the target geometry and the target material. In order to compare this wide range of different experiments, the conversion efficiency (CE) is introduced. The CE is defined as the fraction of energy emitted in the 13 nm line compared to the incident laser pulse energy. Using this definition it is possible to compare the EUV emission produced by completely different laser parameters. It is also the number of interest for the industry dealing with the next-generation EUV-lithography. The high EUV average power demanded by future lithographic applications can only be economically successful with an optimized CE.

Five-fold ionized oxygen has an isolated line at 13 nm, which emits in a spectrally narrow window compared to the reflection curve of the multilayer mirrors used to image the EUV radiation in lithographic applications. To compare the CE of this single line to sources, emitting a broad continuum, like xenon, the CE is related to a certain wavelength region. The generally accepted range is 2.5% of the bandwidth (BW) of 13 nm, meaning the region of $13.00 \pm 0.16$ nm, comparable to the multilayer reflection bandwidth.

4.1 Acquiring EUV spectra

The EUV spectrometer described in Sect. 2.3.1 was used to acquire the spectra of the emission of fivefold ionized oxygen ions ($\text{O}^{5+}$) in the EUV spectral region. The droplets, as described in Sect. 2.2.2, acted as oxygen containing target. The EUV radiation from a single small droplet hit by an intense laser pulse was recorded. The raw data of a recorded spectrum range over a broad wavelength region from 5 nm to 18 nm, as can be seen in e.g. Fig. 4.12 or 4.13. Zooming to the region of interest around 13 nm, four emission lines can be identified. All four lines result from fivefold ionized oxygen (see Tab. 3.2). To correct for the wavelength-dependent response of the spectrometer and in order to determine the number of detected EUV photons, the spectrometer raw data were multiplied with the calibration curve obtained at the BESSY II synchrotron (see Sect. 2.3.1). A typical spectrum presenting the number of photons collected by the spectrometer is shown in Fig. 4.1. The lines show a FWHM of 0.8 nm, which is about ten times the line width determined with a high resolution spectrometer. As already discussed in Sect. 2.3.1, the
4.2. Experimental test of the CRSS code

The spectrum recorded by the transmission grating spectrometer is broadened by the finite source size and additional aberrations of the imaging mirror. These spectra are certainly sufficient to get information about the 13 nm photon yield and the line ratios for various laser parameters. However, to look at the line width in a narrow range around 13.0 nm, high resolution spectra were recorded by a pinhole spectrometer (see Fig. 4.3).

So far only the number of photons in the very small collection solid angle of the spectrometer ($5.9 \times 10^{-4}$ sr) has been determined. To calculate the conversion efficiency for the total emission in the solid angle of $4\pi$, it is essential to know the angular emission characteristic of the EUV radiation, which indeed was found to be isotropic (see Sect. 4.6.3). Therefore, the measured number of photons within the small solid angle of the spectrometer can be scaled to $4\pi$, just by using the geometric ratio. Thus, typical numbers of photons, emitted from the laser-produced plasma hit by just one laser pulse, range between $10^{11}$ to $10^{14}$ photons within 2.5% bandwidth around 13.0 nm and $4\pi$. As will be shown, the number of EUV photons emitted by the plasma strongly depends on the laser parameters.

![Figure 4.1: A typical EUV spectrum obtained for a water droplet target, recorded with the spectrometer described in Sect. 2.3.1. The width of the lines (FWHM ~0.8 nm) is not dominated by the emitted line width, but is rather due to the the finite source size and aberrations of the spectrometer. Some properties of the for prominent lines can be found in Tab. 3.2.](image)

**4.2 Experimental test of the CRSS code**

First of all, it is important to experimentally verify the predictions of the CRSS code (see Sect. 3.3.3), since all theoretical models and the interpretation of the results of the plasma dynamics simulation are based on the CRSS calculation results. This model states that the major part of the 13 nm radiation, produced by fivefold ionized oxygen, results from plasma with a temperature of 25-30 eV and an electron density of about $2 \times 10^{21}$ cm$^{-3}$. As discussed in the theoretical part of the work (see Sect. 3.3.5 to 3.3.7), it is possible to obtain the temperature and the electron density of the region of emission, by comparing the line ratios of different transitions and the
4.2. Experimental test of the CRSS code

measured line width of the 13 nm emission respectively, to theoretical models. First, the temperature is determined by the line ratios. This experimentally obtained temperature and the line width were combined to evaluate the electron density. The experiments described in this section are all performed with the 20 µm water droplet target.

The ratio between the 13 nm line and its neighboring lines can be used to determine the ion temperature. Looking at Tab. 3.2, the first attempt is to compare the optically thin 17.3 nm line (O⁵⁺(3d)→O⁵⁺(2p)) to the likewise optically thin 13.0 nm line (O⁵⁺(4d)→O⁵⁺(2p)). The line ratio calculated by Eq. (3.46) is 0.53, which is in good agreement with the measured line ratio which was determined to lie in the narrow region between 0.45 and 0.60 for the whole range of applied laser parameters. Although the upper levels differ in the principal quantum number and thus, in the potential energy, the line ratio calculated based on the CRSS code results does not depend on the plasma temperature in the region between 20 eV and 40 eV. This surprising finding seems to be due to the complicated recombination dynamics. Both levels are populated by recombining O⁶⁺ ions and therefore, the population dynamics has to be considered for population density of the O⁵⁺(4d) and the O⁵⁺(3d) level. It turns out that the population of the energetic higher level (O⁵⁺(4d)) is higher than the population of the lower one, and thus using the Boltzmann distribution instead the CRSS output (see Sect. 3.3.5) completely fails to describe the observed line ratio at any temperature. Thus, the 17.3 nm/13.0 nm line ratio cannot be used to infer the temperature in the emission range.

Figure 4.2: Comparison of the calculated and measured line ratios. The symbols denote the ratios calculated with the CRSS code output for 3p→2s/4d→2p (stars) and 4p→2s/4d→2p (circles) for ne = 1.4 × 10²¹ cm⁻³ (blue) and ne = 1.4 × 10²⁰ cm⁻³ (black), respectively. The green line was calculated using just the Boltzmann distribution to obtain the ratio of the population density. The shaded areas represent the ratios observed in the measurement.

A somewhat more complicated approach is the comparison of the optically thin 13 nm line and the optically thick 11.6 nm (O⁵⁺(4p)→O⁵⁺(2s)) and 15.0 nm (O⁵⁺(3p)→O⁵⁺(2s)) lines. The 13 nm radiation can easily escape from the plasma, in contrast to the radiation of the other two lines (see Sect. 3.3.6). To correct for the absorption of the resonance lines, the ratio of the lines with the same principal quantum number (4p→2s/4d→2p), which is independent of the temperature, was scaled to the experimental data, as shown in Fig. 4.2. The scaling according to Eq. (3.48)
resulted in $\tau_{4p \rightarrow 2s} = 0.25$. This value corresponds well with the calculated absorption coefficient $\kappa$ (Tab. 3.2). Integrating the absorption of the radiation along their way out of the source region (relying on the simulated density profiles), the radiation has to propagate through the equivalent of a $\sim 20 \, \mu\text{m}$ plasma column with $n_e \sim 2 \times 10^{21} \text{cm}^{-3}$. Using these values, the calculated $\tau$ (Eq. (3.48)) is equal to the experimentally determined optical thickness. Thus, $\tau_{3p \rightarrow 2s}$ can be calculated using the absorption coefficients from Tab. 3.2.

Being able to correct for the absorption, the comparison of the experimental and the calculated ratio of $3p \rightarrow 2s/4d \rightarrow 2p$, tells the ion temperature. The temperature dependence of the line ratios is due to the change of the population density for the upper levels with temperature according to Eq. (3.46). The ratio of the population density can either be calculated by the CRSS code or the Boltzmann distribution. Fig. 4.2 shows the absorption corrected line ratio calculations for the CRSS model (symbols) and the much simpler two-level Boltzmann distribution (green line). Both results agree well with respect to the temperature dependence, but differ in magnitude. This is not surprising since only two levels are accounted for in the Boltzmann model. The CRSS code provides the population density not only as function of the temperature, but also as function of the electron density. The line ratio is plotted for two electron densities, however the ratios are not substantially different for higher or lower electron densities than the ones shown. Thus, no assumptions about the electron densities have to be made. The intersection with the experimental region lies between 28 eV and 32 eV. This result is in good agreement with the time-dependent rate equation model (Sect. 3.3.4) and only slightly higher than the optimum temperature range of 25-30 eV, predicted by the CRSS calculation. The experimental line ratios were obtained for laser pulse durations ranging from 200 fs up to 6 ns and laser pulse energies from 20 mJ to 200 mJ. Despite the broad range of laser parameters spanning orders of magnitude, the line ratios vary only within $\pm 15\%$. Taking a look at Fig. 4.13, which shows the spectrometer data from 5 nm to 18 nm, it is astonishing that the relative ratios of the four lines lie in a narrow range, while distinct differences at shorter wavelength can be seen. This result implies that the EUV radiation (11-17 nm) is not emitted from a plasma region which is directly heated by the laser pulse, but rather from an expanded region as the plasma cools down. Indeed, the simulation showed much higher plasma temperatures (200-300 eV) for the laser-plasma interaction region. Temperatures around 30 eV do not evolve until the plasma has already expanded substantially (see Sect. 5.1).

To get information about the electron density in the EUV emitting region, spectroscopic methods can help as well. As discussed in Sect. 3.3.7, the line width can give information about the electron density. The 13 nm line acquired with the high resolution pinhole spectrometer, described in Sect. 2.3.2, is shown in Fig. 4.3. This line was fitted with a Voigt profile [Thorne et al., 1999] which can determine the relative influence of the Lorentzian and the Gaussian contribution to the line profile. This fit states that the line is indeed entirely Lorentzian and the Gaussian contribution is well below 1%. Thus, Doppler broadening is not important. The line width of the $\text{O}^{5+}(4d) \rightarrow (2p)$ transition, measured for 1.8 ps laser pulses, was determined as 0.087 nm (FWHM) equivalent to
4.3 The mass-limited droplet target

The water droplets described in Sect. 2.2.2 acted as laser target for the experiments presented in this section. The dependence of the EUV conversion efficiency on various laser parameters and droplet sizes was measured. These results will be compared to the results for extended targets (Sect. 4.4) and to the simulations described in Chap. 5.

Large high average-power lasers, which will be used in future lithographic applications will not be able to generate focal spots smaller than 20 µm in diameter. Thus, to measure with industrially relevant parameters and to heat the droplets as homogeneously as possible, the size of the laser focal spot was adjusted to the droplet size. To do so, the off-axis paraboloid had to be adjusted well in order to avoid astigmatic line foci. However, the resulting focal spot is much smaller than the droplet diameter (see Sect. A.6.1). Hence, the droplets were moved 100 µm out of the actual focus to match the laser beam diameter to the droplet (see Fig. A.4). The laser transmission was used for the fine tuning of the droplet position. The transmission of the laser through the

\[
\left( \frac{\Delta \lambda}{\lambda} \right)_{\text{exp}} = 150.
\]

The width of the line is again almost independent of the laser parameters. This width is too large to be explained by the natural line width and therefore the Stark broadening is the dominant effect. The measured line width has to be corrected for the finite spectrometer resolution. The denoted value of \(\frac{\Delta \lambda}{\lambda} = 500\) (see Sect. 2.3.2) was taken as upper limit and \(\frac{\Delta \lambda}{\lambda} = 320\), measured for the nearby O\(^{6+}\) 3d→2p line, was used as lower limit for the spectrometer resolution. Inserting the measured parameters in Eq. (3.51) results in an electron density of \(n_e = (1.4 \pm 0.2) \times 10^{21} \text{ cm}^{-3}\) for the EUV emitting plasma.

The CRSS results represent the temperature and the electron density of the EUV emitting region well, thus further calculations and simulations can be based on the CRSS output.

4.3 The mass-limited droplet target

The high resolution spectrum of the 13 nm region was acquired with the pinhole spectrometer (see Sect. 2.3.2). The O\(^{5+}\) (4d)→(2p) transition with a FWHM of 0.087 nm and the close-by O\(^{6+}\) (3d)→(2p) line with a FWHM of 0.041 nm are shown. The Si L-absorption edge (at 12.4 nm) of the SiO\(_2\) oxide layer on top of the CCD chip can clearly be seen.

Figure 4.3: The high resolution spectrum of the 13 nm region was acquired with the pinhole spectrometer (see Sect. 2.3.2). The O\(^{5+}\) (4d)→(2p) transition with a FWHM of 0.087 nm and the close-by O\(^{6+}\) (3d)→(2p) line with a FWHM of 0.041 nm are shown. The Si L-absorption edge (at 12.4 nm) of the SiO\(_2\) oxide layer on top of the CCD chip can clearly be seen.

\[
\left( \frac{\Delta \lambda}{\lambda} \right)_{\text{exp}} = 150.
\]

The width of the line is again almost independent of the laser parameters. This width is too large to be explained by the natural line width and therefore the Stark broadening is the dominant effect. The measured line width has to be corrected for the finite spectrometer resolution. The denoted value of \(\frac{\Delta \lambda}{\lambda} = 500\) (see Sect. 2.3.2) was taken as upper limit and \(\frac{\Delta \lambda}{\lambda} = 320\), measured for the nearby O\(^{6+}\) 3d→2p line, was used as lower limit for the spectrometer resolution. Inserting the measured parameters in Eq. (3.51) results in an electron density of \(n_e = (1.4 \pm 0.2) \times 10^{21} \text{ cm}^{-3}\) for the EUV emitting plasma.

The CRSS results represent the temperature and the electron density of the EUV emitting region well, thus further calculations and simulations can be based on the CRSS output.

4.3 The mass-limited droplet target

The water droplets described in Sect. 2.2.2 acted as laser target for the experiments presented in this section. The dependence of the EUV conversion efficiency on various laser parameters and droplet sizes was measured. These results will be compared to the results for extended targets (Sect. 4.4) and to the simulations described in Chap. 5.

Large high average-power lasers, which will be used in future lithographic applications will not be able to generate focal spots smaller than 20 µm in diameter. Thus, to measure with industrially relevant parameters and to heat the droplets as homogeneously as possible, the size of the laser focal spot was adjusted to the droplet size. To do so, the off-axis paraboloid had to be adjusted well in order to avoid astigmatic line foci. However, the resulting focal spot is much smaller than the droplet diameter (see Sect. A.6.1). Hence, the droplets were moved 100 µm out of the actual focus to match the laser beam diameter to the droplet (see Fig. A.4). The laser transmission was used for the fine tuning of the droplet position. The transmission of the laser through the
4.3. The mass-limited droplet target

droplet was continuously observed during the experiments (see Fig. 2.7). When the droplets were positioned too far away from the focus, the transmission rose and the position was readjusted.

All the EUV yields shown result from single shot measurements. The EUV radiation was detected either by the transmission grating spectrometer or the EUV-monochromator. Between 20 and 1000 EUV yields from single laser shots were averaged in order to get a reliable value for the CE. This method enables the characterization of each single laser-plasma interaction, therefore minimizing systematic errors possibly occurring in measurements averaging over thousands of shots.

The error bars of the CE (see e.g. Fig. 4.5), not included in every plot, result from the shot to shot fluctuations of the EUV radiation, due to fluctuations in the laser energy as well as in the plasma interaction, since the focus varies about ± 5 µm from shot to shot in respect to the droplets. The fluctuations can be traced back to vibrations induced by the vacuum pumps and to the vibrations of the entire house, in which the laboratory is located in the third floor.

4.3.1 Dependence of the conversion efficiency (CE) on the laser pulse energy

First of all, the influence of the laser pulse energy on the CE was investigated. The pulse energy was varied in the range of 20 mJ, determined by the detection limit of the EUV detectors, up to 200 mJ. The conversion efficiency as a function of the laser pulse energy was measured for various pulse durations, starting with 200 fs, 1.2 ps, 1.8 ps, 10 ps up to 120 ps and 6 ns. The behavior of the CE on the laser pulse duration was found to be similar for all measured pulse durations, even though they differed by almost five orders of magnitude. Fig. 4.4 shows the energy dependence for various pulse durations. All sub-nanosecond pulses show a more or less distinct peak at 50 mJ. This seems to be a universal feature, independent of the laser pulse duration. The peak is most dominant and yields the highest CE for a laser pulse duration of 120 ps (Fig. 4.5). As will turn out in the following discussion, the peak is due to the limited number of particles within the target. A completely different behavior was found for large extended targets with an almost unlimited number of particles which can interact with the laser pulse. The results for the extended targets will be discussed in Sect. 4.4.

The reason for the energy scaling of the CE is simple: The number of atoms in the interaction region of the laser is fixed for a droplet target. By assuming an equilibrium situation as the simplest model, the laser energy necessary for a given droplet size to provide optimum conditions for each atom to efficiently emit 13 nm radiation can be calculated. For a given water droplet diameter of 20 µm, the number of molecules contained by the droplet is ∼ 1.4 × 10^{14}. An energy of 0.35 keV is needed for each oxygen atom to be ionized to the desired O^{5+}(4d) ionic state (cp. Tab. 3.1). Furthermore, the plasma created has to be heated to at least 25 eV. Each water molecule contributes with 7 free electrons and three ions (O^{5+} and two protons) to the plasma. Multiplication with the
4.3. The mass-limited droplet target

The mass-limited droplet target

number of degrees of freedom for these 10 particles results in 0.4 keV per molecule for the plasma heating. Therefore, ionization and heating of the entire droplet to 25 eV requires $\sim 15$ mJ.

This is a lower limit for the laser energy per pulse, since not the entire laser energy impinging on the droplet is absorbed by the plasma. According to experimental results the fraction of absorbed laser energy is $\sim 40\%$ for 120 ps pulses (e.g. [Garban-Labaune, 1982, Wood et al., 1988]) and decreases with the pulse duration to about 20% for the 1 ps pulses [Price et al., 1995], in good agreement with the values calculated by the MEDUSA simulation. It should also be noted that not the entire absorbed energy contributes to the heating. About 50% is consumed by the plasma expansion. Summing up all these numbers, a laser pulse energy of 50 mJ to 100 mJ for 120 ps to 200 fs pulses, respectively, should yield optimum plasma conditions and thus the highest conversion efficiency. Less energy than 50-100 mJ does not heat the plasma enough and the fraction of O$^5+$ and O$^6+$ is reduced. Energy is consumed to get O$^4+$ but the ion will not contribute to the 13 nm radiation and the conversion efficiency decreases. On the other hand, applying more energy per laser pulse the plasma is heated to higher temperatures giving rise to helium- and hydrogen-like oxygen, which radiate soft x-rays. The conversion efficiency decreases again since energy is spent on heating that does not contribute to the 13 nm radiation.

The differing behavior of the CE for 6 ns pulses can be attributed to the expansion of the droplet during the laser-plasma interaction (see Sect. 4.3.3). Since the laser focal spot of 20 $\mu$m diameter is smaller than the expanded droplet, the laser pulse does not heat the entire droplet but rather only part of it. The rest is blown off by the plasma expansion. The laser interacts with an effectively smaller droplet and the maximum in the CE should be found at a pulse energy below 20 mJ. Ref. [Vogt et al., 2001] indeed reports a maximum CE at 4 mJ for 3 ns pulse duration focused onto a 10 $\mu$m diameter H$_2$O jet.
4.3. The mass-limited droplet target

The model states that the laser pulse energy needed for an optimum CE is proportional to the number of ions in the interaction volume (respectively, the droplet volume). Despite its simplicity the model can explain the general behavior of the measured CE quite well (see Fig. 4.4). As already pointed out in the introduction, various differing numbers for the CE can be found in the literature. All these published conversion efficiencies indicate that the CE rises with the droplet size, a feature which can also be explained by the model discussed.

Using different droplet sizes is an alternative approach for testing this simple model, which states that the CE depends on the droplet volume. As pointed out in Sect. 2.2.2, the droplet source is capable of providing droplets with volumes ranging from half to double the volume compared to the 20 µm droplets. For 120 ps pulses, for which the dependence of the CE on the laser pulse energy is particularly distinctive, it is to be expected that the best conversion is found at laser pulse energies below 50 mJ for droplet diameters below 20 µm, whereas higher energies should result in a higher CE for bigger droplets. Indeed, Fig. 4.6 shows exactly this behavior. The variation of the CE with the droplet size was also measured for the other pulse durations, however, the difference was not as obvious as for the 120 ps pulses. The changes were only close to the size of the error bars.

![Figure 4.6: The dependence of 13 nm CE on the laser pulse energy was measured for 15 µm diameter droplets (0.42 times the volume of 20 µm diameter droplets) and 25 µm diameter droplets (2.0 times the volume of 20 µm diameter droplets). The lines indicate the position of the maximum CE. The laser pulse duration was 120 ps.](image)

4.3.2 CE dependence on the laser pulse duration

The EUV CE was measured for laser pulse durations varying over almost five orders of magnitude. By looking at the measured results, as shown in Fig. 4.7, it becomes clear that the CE is a rather complicated function of the laser pulse duration. Again, as for the dependence of the CE on the laser pulse energy, a maximum in the CE was found. The position of the maximum is in turn independent of the variation of the laser pulse energy, even though the intensity of the laser interaction varies by one order of magnitude. Thus the position of the peak does indeed depend on the pulse duration, rather than on the intensity. The outstanding role of the 120 ps laser pulses
can also be seen in spectra taken for the different pulse durations (see Fig. 4.13). 120 ps pulses can obviously heat the droplet plasma to higher temperatures than shorter and longer pulses do. It is quite surprising that not the ns pulses, but rather 120 ps laser pulses, yield the highest CE. According to the theoretical discussion, the laser absorption and thus the heating of the plasma should increase steadily with the pulse duration (see Sect. 3.2.2). However, all these arguments are based on (infinitely) extended targets, and indeed for such type of target the CE increases continuously from ultra-short to ns pulses (see Sect. 4.4). The deviation of the results for the droplets can again be traced to the mass-limitation of the target. The plasma tends to expand (see Sect. 4.3.3) as soon as the laser pulse impinges onto the target. The expansion is rather small for short pulses up to 100 ps, but cannot be neglected for ns pulse durations anymore. As will be shown in the next section, the droplet is diluted by the expansion within half a ns to a cloud of undercritical plasma. The scenario for ns laser pulses is the following: The first part of the laser pulse starts to heat the droplet and plasma streams away. After only a fraction of the laser pulse arrived at the center of the droplet, it has been diluted to undercritical plasma, providing a lower absorption coefficient (see Eq. (3.22)). The trailing edge of the laser pulse is less efficiently absorbed, resulting in a lower CE. It has to be emphasized that the absorption is not zero in the undercritical plasma, still it is far below the optimum. Summarizing the model, one can state: The longer the laser pulse, the higher the CE, until the plasma expansion starts to degrade the absorption and the CE falls. According to this argument, the optimum pulse duration is equivalent to the time $t_{\text{crit}}$, after which the plasma density falls below the critical density $n_{\text{crit}}$. $t_{\text{crit}}$ is proportional to the droplet diameter, as will turn out in the next section. This also leads to the scaling that the optimum pulse duration for achieving the highest EUV CE is proportional to the droplet diameter (see Sect. 4.3.3).

Summarizing the results from Figs. 4.4 and 4.7, in Fig. 4.8, it turns out that for mass limited
targets, an optimum CE is obtained if laser pulse energy and laser pulse duration are separately adapted to the target, contrary to bulk targets where the intensity seems to be the crucial parameter (Sect. 4.4). For the given 20 μm diameter droplet target there is exactly one optimum within

\[ \text{EUV CE in } \% \quad \frac{4}{G53} \quad 2.5 \% \quad \text{BW} \]

\[ 6 \text{ns} \quad 120 \text{ ps} \quad 2 \text{ ps} \quad 10 \text{ ps} \quad 200 \text{ fs} \]

Laser pulse duration

\[ \begin{array}{ccccccc}
0.03 & 0.04 & 0.06 & 0.08 & 0.09 & 0.11 & 0.13 \\
0.14 & 0.16 & 0.17 & 0.19 & 0.21 & 0.23 & \end{array} \]

Laser pulse energy in mJ

\[ \text{Figure 4.8: The plot shows in a compact form the dependence of the EUV CE on the laser pulse energy and duration (summary of Fig. 4.4 and Fig. 4.7). There is only one optimum laser pulse duration and laser pulse energy for a given droplet size.} \]

the laser parameter field of laser pulse duration and laser pulse energy resulting in a maximum EUV CE. The existence of other maxima outside the measured region has been ruled out by the discussion in the last sections. The CE can be substantially improved by choosing optimum laser parameters.

### 4.3.3 Droplet expansion and laser transmission

In order to confirm the model of the droplet expansion, the transmission of the laser pulse through the droplet and the ion expansion velocity were measured.

The transmission of the laser beam was monitored by a pyroelectric energy meter (Gentec ED 500), as shown in Fig. 2.7. When the droplet chain was not synchronized with the laser, some of the laser pulses hit a droplet completely, yielding a low transmission, and others only partially or not at all - equivalent to a transmission of 1. It is important to note that the laser energy after subtraction of the transmission is not equal to the absorbed energy since the major part of the laser energy is reflected. The EUV signal is plotted versus the laser transmission for nanosecond (6 ns) and picosecond laser pulses (see Fig. 4.9). Surprisingly, all transmission measurements for ps pulses (120 ps down to 0.2 ps) showed the same dependence. In case of short pulse durations, a high conversion efficiency can only be achieved for transmissions below 20%, one has to hit the droplet centrally. However, a transmission of 0% cannot be achieved, since the laser focus has a Gaussian distribution, thus the wings of the focal spot, about 10% of the energy, lay outside of the droplet radius. The hydrodynamic reaction of the plasma can be neglected during such short pulses, therefore the droplet diameter does not change while the laser interaction. The plasma gradient can be approximated by a step-like density profile (small scale-length). As discussed in Sect.
4.3. The mass-limited droplet target

3.2.2, the absorption of a step-like plasma gradient depends on the angle of incidence. Normal incidence provides the highest absorption, for s-polarized light, decreasing to zero absorption for grazing incidence - all the laser light is reflected. The case for p-polarized light is somewhat more involved. The absorption does not simply decrease with the angle of incidence. However, the geometry of the interaction is such that the horizontally polarized laser light impinges on the vertically (unsynchronized) moving droplets (cp. Fig. 2.7). If the laser impinges not centrally onto the droplet, it impinges above or below the center of the droplet, but not on the side of the droplet. Thus, the angle of incidence changes for the fraction of light impinging s-polarized onto the droplet whereas the p-polarized light impinges always under almost the same angles.

If the droplet is hit centrally the laser impinges under normal incidence for the dominant area of the droplet and the absorption is maximum. On the other hand, if the laser hits the droplet not concentrically, the absorption will decrease significantly. To calculate the dependence of the absorbed laser radiation as function of the transmitted laser energy, a 20 µm diameter plasma sphere with the angular dependence of the reflection and absorption characteristics given by Eq. (3.23) was assumed. The critical density $n_{\text{crit}}$ was taken as electron density and $v_{ei}$ was calculated for an electron temperature of $T_e = 300$ eV, according to the MEDUSA simulations. However, the angular dependence is not strongly dependent on these parameters. Knowing the dependence of the absorption on the angle of incidence, the laser energy which is absorbed when a Gaussian laser beam impinges on the droplet can be calculated. In the calculation, the center of the laser beam was moved away from the droplet center step by step, recording the transmitted and the absorbed energy. The line in Fig. 4.9 shows the absorbed laser energy, scaled to the measured data, as function of the transmission. The laser beam interacting with the droplet was approximated by a Gaussian energy distribution with a FWHM of 10 µm and a 40 µm diameter foot containing 10% of the laser energy (see e.g. Fig. A.4). The data measured for short pulses can indeed be explained by modeling the droplet as reflecting plasma sphere with a step-like plasma gradient.

The stars in Fig. 4.9 show the transmission data for the 6 ns laser pulses. The EUV signal has a significantly different behavior. The transmission does not show small values ($< 30\%$) at all and most of the measurements yielded transmissions below 50%. The absence of low transmission values can be explained by the plasma expansion. The droplet already expands significantly during the pulse, such that the trailing edge of the laser pulse sees only low density plasma and thus the absorption is low as discussed in the last section. As a consequence, the conversion efficiency for long pulses stays far below the value for short ones, and well adapted pulses. While in the short-pulse case one has to hit the droplet centrally to get a high EUV output, for the ns pulse the EUV yield falls linearly with the transmission. The long laser pulse creates a sufficiently long scale-length, such that the absorption is not strongly dependent on the angle of incidence. $L$ dominates the exponent in Eq. (3.26) and the absorption is close to 1 for most angles of incidence. Thus, most of the laser energy hitting the droplet is indeed absorbed. All the laser energy which is not detected by the energy meter is absorbed the target and, therefore, the CE is expected to decrease.
4.3. The mass-limited droplet target

linearly with increasing transmission, as observed in the measurement.

The transmission measurements presented only an indirect hint on the expansion. Therefore, the plasma expansion velocity was directly measured in order to check if the droplet expands fast enough at all to dilute substantially within nanoseconds. The time after which the droplet plasma becomes undercritical is denoted with \( t_{\text{crit}} \). Assuming within a very simple model that all particles are moving outwards of the interaction region with the expansion velocity \( v_{\text{exp}} \), the mean density becomes critical once the droplet expanded to a critical radius \( r_{\text{crit}} \). This can be expressed in

\[
 t_{\text{crit}} = r_{\text{crit}} \times v_{\text{exp}}.
\]

The expansion starts with an initial radius of \( r_0 = 10 \mu m \) and solid state density \( n_0 \approx 2 \times 10^{23} \text{cm}^{-3} \). Therefore, the critical radius

\[
 r_{\text{crit}} = \sqrt[3]{\frac{n_0}{n_{\text{crit}}}} \times r_0
\]

(4.1)
is calculated to be 50 \( \mu m \). The expansion velocity was measured, by using a Faraday cup (Sect. 4.5). For all pulse durations and laser pulse energies used in the EUV measurements, the peak of the expansion velocity always lay between 1.0 and \( 2.6 \times 10^7 \text{ cm/s} \). Thus, \( t_{\text{crit}} \) can be expected to be in the range of \( \sim 200 \text{ – 500 ps} \). Despite the simplicity of the model, the results are in good agreement with the measurements and the plasma-dynamic simulations. This is true for our droplet size where the expansion time is \( \sim 400 \text{ ps} \) and, looking at the literature [Parra et al., 2000, McNaught et al., 2001], \( t_{\text{exp}} \) was found to be \( \sim 200 \text{ ps} \) for 4 to 7 \( \mu m \) diameter droplets consisting of heavy Kr and Ar, also in remarkable agreement with this model.

Figure 4.9: The transmission of several laser pulses that were unsynchronized to the 20 \( \mu m \) droplets is shown for short (\( \leq 120 \text{ ps} / \text{circles} \)) and long (6 ns / stars) pulses. The line indicates the calculated absorption for a step-like plasma gradient at the droplet surface.

Another independent method to determine the size of the expanded droplet is provided by the 13 nm source size, as determined in Sect. 4.6.2. According to the theoretical discussion (see Fig.3.2), the 13 nm radiation is mainly emitted from plasma at the critical density. Thus, the radius of the 13 nm source size is thought to be equivalent to the critical radius in Eq. (4.1). Indeed, the 13 nm source size was measured to be between 35 \( \mu m \) and 90 \( \mu m \), depending on the laser parameters.
4.3.4 Prepulse dependence of the EUV CE

The laser-plasma interaction described so far was concerned with laser pulses impinging on a solid density target \((n_e \sim 10^{23} \text{cm}^{-3})\). If the density of the plasma with which the laser pulse interacts is decreased the interaction changes dramatically. Experimentally, this is performed by introducing a prepulse, preceding the main pulse (for the production of prepulses see Sect. A.4). This prepulse, typically containing 2% to 10% of the main pulse energy and the same pulse duration as the main pulse, creates a plasma at the surface of the target without evaporating the whole droplet. This plasma, the so-called preplasma, expands, leading to a plasma gradient that changes with time. The longer the delay after the prepulse hit the droplet, the more spread out and diluted the preplasma becomes. Thus, by changing the delay between the prepulse and the main pulse, the plasma environment interacting with the main pulse can be altered. The spatial extent of the plasma and the scale-length increases, whereas the electron density, the plasma temperature and the mean charge state decreases with longer delay times. In addition geometrical effects, like de-focusing of the laser beam, take place for large density gradients. The plasma gradient acts like a concave lens, since the refractive index in the undercritical plasma is below 1 (see Eq. (3.10)). This, in turn, reduces the laser intensity on the target.

First, the CE was measured as function of the prepulse delay for laser pulse durations of 1.8 ps and 120 ps with prepulse delays of up to 11 ns as shown in Fig. 4.10. The measurements were carried out for constant prepulse (2 mJ) and main pulse (25 mJ) energies to get a general overview of the influence of prepulses on the CE. A rapid increase of the CE as function of the prepulse delay is observed. The increase, present for both pulse durations, is much more distinct for the short pulses (1.8 ps) as compared to the 120 ps long pulses. The CE, measured for the 1.8 ps pulses increases by a factor of 15 until a maximum CE of almost 2% in \(4\pi\) is reached at a prepulse delay of 3–4 ns. The preplasma, streaming away from the droplet, dilutes for longer delay times and the influence on the laser-plasma interaction of the main pulse is less effective, thus the CE decreases for even longer delays. The CE produced by 120 ps pulses, on the other hand, increases

![Figure 4.10: The (2 mJ) prepulse dependence for 20 µm water droplets is shown for long delays. The measurement was performed for laser pulse durations of 1.8 ps and 120 ps with a main laser pulse energy of 25 mJ.](image-url)
only by a factor of 2.5 for prepulse delays of up to 2 ns while the CE stays constant for longer delays.

As next step, the CE was investigated in detail for short prepulse delays at which the increase is especially pronounced. The CE was measured for prepulse delays up to 700 ps (Fig. 4.11) in 100 ps steps for different pulse energies and a wider range of pulse durations. The energy of the prepulse was again 2 mJ for each measurement, whereas the energy of the delayed main pulse varied between 20 mJ and 100 mJ. The prepulse energy was kept constant to provide the same preplasma conditions for different main pulse energies. For each laser pulse energy and each delay, the EUV emission was measured for a laser pulse duration of 200 fs, 1.8 ps, 10 ps and 120 ps. No experiments were performed for a laser pulse duration of 6 ns, since no prepulse-dependent effect can be expected within the feasible delay times. The EUV CE as function of the prepulse delay, measured for 20 mJ laser pulses is shown in a compact form in Fig. 5.6 for all four measured laser pulse durations. This figure pinpoints the unique status of the 2 ps pulses regarding to high CE. The experiment shows, as the most important result, that the CE increases with increasing prepulse delay (up to 700 ps) for each measured laser pulse energy as well as for each pulse duration. However, there are substantial differences in the behavior of the CE as function of the various laser parameters. The optimum CE was found for a laser pulse energy of 20-50 mJ for 1.8 ps pulses and 20 mJ or slightly below for 120 ps. Higher pulse energies (100 mJ) result in a lower CE (for the same prepulse delay) and on the other hand for laser pulse energies considerably below several mJ, the laser-heating of the target is not sufficient to produce EUV radiation anymore resulting in a lower CE as well. Thus, the measurement indicates the existence of an optimum laser pulse energy of about 20 mJ as soon as a prepulse is introduced. The optimum pulse energy is lower in the experiments with prepulses compared to the measurements without prepulses (see Fig. 4.5). Especially interesting is the dependence of the CE on the laser pulse

![Figure 4.11: Experimental results on the dependence of the 13 nm CE on 2 mJ prepulses for different main pulse energies at a laser pulse duration of 1.8 ps (a) and 120 ps (b) in the region of 0 to 700 ps prepulse delay. The error bars, the same for all data points, result from shot to shot fluctuations.](image-url)
4.3. The mass-limited droplet target

duration. Without introducing a prepulse, the CE is lower for short (∼ 2 ps) laser pulses compared to 120 ps pulses (see Fig. 4.7). By contrast, using a prepulse, the highest CE for 1.8 ps pulses is significantly higher than the best CE for 120 ps pulses (for the same prepulse delay). With the introduction of a prepulse, the pulse length for the best CE is shifted to shorter pulses. Thus, the simple model for the scaling of the CE with the pulse duration as introduced in the Sect. 4.3.2 is not valid for complicated laser pulse forms. The expansion of the droplet does not seem to be the dominant factor influencing the EUV CE as soon as a prepulse is introduced.

Not only was the EUV CE measured for different prepulse parameters but also the spectra covering the entire EUV range. The spectra produced by the 120 ps pulses were found to be almost independent on the introduction of prepulses, whereas the spectra recorded for 1.8 ps pulses were strongly prepulse-dependent. The plasma became hotter as the prepulse delay was increased. The emission at short wavelengths rose in respect to the 13 nm emission. Comparing Fig. 4.12 with Fig. 4.13 a certain analogy between the spectra of the 120 ps pulses and the 1.8 ps pulses with prepulse can be seen, both show a strong emission at short wavelength. This equality is no coincidence, but will be explained in Sect. 5.1 by the simulation results.

**Figure 4.12:** The spectrum for a 2 ps laser pulse ($E_L=50$ mJ, 700 ps prepulse delay) preceded by a prepulse is compared to the spectrum without prepulse. Both spectra were normalized on the 13 nm peak, to simplify the comparison. The use of a prepulse enhances the plasma temperature.

**Figure 4.13:** Spectra were recorded for different laser pulse durations ($E_L=50$ mJ, without prepulse). The highest plasma temperatures were achieved with the 120 ps pulses.

Summarizing the experiments, using a prepulse enhances the CE distinctly, while the optimum laser parameters shift from 120 ps pulse duration and 50 mJ laser pulse energy to 2 ps and 20 mJ, respectively. The EUV CE measured for 1.8 ps pulses was increased by a factor of 15 up to 2% in $4\pi$ and 2.5% BW just by introducing a prepulse, while the CE of 120 ps laser pulses remained almost constant within a factor of 2.5. The optimum CE obtained with the use of a prepulse is 8 times higher than the best CE measured without subsidiary prepulse. Finally, comparing the
measurements with prepulses, the best CE obtained for 2 ps laser pulses is still 3 times higher than the best value recorded for 120 ps laser pulses. To explain these surprising results, simulations were performed, as described in the next chapter.

### 4.3.5 Dependence of the EUV CE on the prepulse energy

In the experiments discussed so far, the prepulse energy was always kept constant at 2 mJ to provide the same preplasma conditions for different main pulse energies. To optimize the CE, however, it is interesting to study the influence of the energy contained by the prepulse on the CE. Fig. 4.14 summarizes the experimental and simulated (see Chap. 5) results. With regard to the experiment in which one laser pulse is split into two replica to create pre- and main pulse, the sum of the prepulse energy and the main pulse energy was kept constant for the simulation. Thus, a prepulse fraction of 50% is equivalent to two equally strong double pulses.

![Figure 4.14: The dependence of the EUV CE on the prepulse energy for 2 ps laser pulses (20 mJ / 700 ps prepulse delay). The measured as well as the simulated (see Chap. 5) data are shown.](image)

It can be seen that the introduction of a weak prepulse enhances the CE rapidly. Increasing the energy of the prepulse further leads to a hotter preplasma which expands faster. This in turn leads to a longer plasma scale-length enhancing the laser absorption (see Sect. 3.2.2 and 3.2.3). However, for prepulses containing a large portion of the total energy, the lower energy in the main pulse starts to affect the interaction. The main pulse is not strong enough to heat the plasma sufficiently and the CE falls down to the initial value without prepulse when all the laser energy (100%) contained in the prepulse.

The fraction of the energy contained by the prepulse is limited to 10% for 20 mJ pulses in the laser system used. The low CE at 0% and 100% prepulse energy leads to the prediction of an optimum prepulse energy fraction. This optimum could not be achieved with the current laser setup (blue dots in Fig. 4.14), and therefore, the simulation of the EUV CE (see next chapter) was used to determine the optimum which was found at 20% prepulse energy (Fig. 4.14). These
results lead to the promising prediction that an even higher EUV CE than the one measured could be achieved by optimizing the prepulse energy.

### 4.3.6 Measurement of x-rays

The images which were recorded by the CCD camera of the transmission grating spectrometer (described in Sect. 2.3.1) can be used to obtain information about the emission of hard x-rays from the plasma. By analyzing the acquired images the spectral range up to photon energies of $\sim 30$ keV can be measured. Looking very closely at the CCD-image recorded by the spectrometer (see Fig. 2.9) one can find single pixels with high readings lying outside the actual spectrum. These single pixels are hit by one x-ray photon with an energy of several keV. These x-ray photons result from bremsstrahlung originating from hot supra-thermal electrons, produced by collective absorption mechanisms (see Sect. 3.2.3). The x-ray photon is absorbed within the pixel and produces electron-hole pairs, which are detected as counts. Every electron-hole pair needs 3.66 eV of energy to be created, therefore e.g. an x-ray photon with an energy of 3.7 keV produces about thousand pairs [Scholze et al., 2000]. In addition, the CCD was calibrated by irradiating it with monochromatic x-ray photons produced by an x-ray tube.

![Figure 4.15: The figure shows a histogram of x-ray photons detected on the CCD chip of the spectrometer. 200 fs laser pulses yield considerably more x-rays with a higher temperature compared to 1.8 ps pulses at a laser pulse energy of 20 mJ.](image)

By analyzing the CCD images and counting the number of pixels that recorded equivalent counts, an x-ray histogram can be extracted. Fig. 4.15 shows the result for 2 ps (blue) and for 200 fs laser pulses (red). Not surprisingly there are on average fewer x-ray photons counted at higher than at lower photon energies. The decrease is indeed exponential, as expected from a Maxwellian hot electron distribution, thus a hot electron temperature can be assigned to the spectrum (see Sect. 3.2.4). The fs-pulses show ten times more hard x-rays and a higher temperature ($T_{\text{hot}} \sim 4$ keV) as compared to the 2 ps pulses ($T_{\text{hot}} \sim 1.5$ keV). This result is comparable to the predicted hot electron temperature given by Eq. (3.33). The calculation states $T_{\text{hot}} \sim 6.5$ keV for the 200 fs pulse and $T_{\text{hot}} \sim 2.8$ keV for the 2 ps pulses. It was also observed that the number of
hard x-rays increased significantly, by factor of 7, for the 2 ps pulses (20 mJ) as soon as a prepulse with several hundred ps delay was introduced. These results will be needed in the discussion of the simulations (next chapter).

4.4 Solid extended targets

It is interesting to compare results that have been obtained with the mass-limited droplet targets with a completely different and more common target type - an extended solid target. The geometry is not spherical, but rather planar, thus the plasma cannot expand equally in all directions and most of all there is a (principally) unlimited number of target atoms which can interact with the laser pulse. As will be shown, this leads to a different behavior of the EUV CE with respect to the laser pulse energy and pulse duration, enabling higher conversion efficiencies compared to mass-limited targets. However, due to the much higher emission of debris, the industrial use of non mass-limited targets will be extremely difficult [Berglund et al., 1998, Richardson, 1998, Rymell & Hertz, 1993].

4.4.1 The ice target

The first attempt to compare the laser-plasma interaction of mass-limited water droplets to a solid type target was performed with an ice target. Water was frozen on top of a water-cooled Peltier element, capable of cooling the ice to -15° centigrade. Since the droplet and the ice target had the same chemical composition, the only difference was the target geometry. Preliminary experiments showed a similar EUV spectrum (see Fig. 4.16) but a ÷1.5 times higher EUV yield for equivalent laser parameters. Detailed investigations, however, could not be performed with the ice target, due to the high vapor pressure of ice, which is 2 mbar for ice at -15° centigrade [Weast, 1980]. To avoid absorption of the EUV radiation, the chamber had to be pumped down to < 10⁻³ mbar, leading to evaporation of ice. The initially flat ice surface evaporated with a speed of ÷ 0.1 mm/min in the vacuum, leaving a rough undefined chunk of ice, thus preventing well defined experiments. Therefore, a different target material which contains oxygen had to be found, to perform carefully controlled experiments.

4.4.2 EUV CE of an extended quartz target

For this purpose a quartz (SiO₂) target was chosen. A bulk target (4 × 4 cm ; 0.5 cm thick) with an optically polished surface was used. According to the Kelly atomic database [Kelly] there are some weak lines from Si⁹⁺,Si¹⁰⁺ and Si¹¹⁺ ions within the bandwidth of 2.5% around 13 nm
which could influence the CE. Fortunately, the ionization potential for these highly charged ions (300 eV to 400 eV [Weast, 1980]) is considerably higher than for the O$^{5+}$ (138 eV, see Tab. 3.2). Thus, their contribution to the 13 nm radiation is very small. However, several additional Si lines outside the small spectral window around 13 nm (e.g. at 14 nm) have been observed for the quartz target compared to ice or water droplets (see Fig. 4.16).

**Figure 4.16:** The figure shows the EUV spectra obtained for a quartz target in comparison to water droplet and ice spectra (pulse duration: 120 ps, pulse energy: 50 mJ). The spectral lines of the solid targets are almost merged to a continuum, due to the blurring of the spectrometer image by a larger EUV source size.

### EUV CE dependence on the laser pulse energy

Fig. 4.17 shows the conversion efficiency as function of the laser pulse energy for two different pulse durations, namely 6 ns and 120 ps. The CE was calculated assuming a Lambertian distribution of the EUV radiation ($I_{\text{EUV}}(\phi) \propto \cos \phi$, where $\phi$ is the angle to the target normal) emitting in one hemisphere [Spitzer et al., 1996]. The spectrometer was placed at an angle of 45$^\circ$ degrees to the laser axis. Since EUV is only emitted within a solid angle of $2\pi$, all CE values for the solid extended target will be given in % per $2\pi$ and 2.5% BW. Comparing this figure to the CE measured for water droplets (Fig. 4.4) the results are quite different. First of all the absolute value of the CE is much higher. The highest CE measured for the quartz target, when scaled to $4\pi$ sr, (Fig. 4.17) is about a factor of ten higher than the best value obtained with the water droplets without prepulse. Whereas similar conversions were achieved with water droplets by introducing an optimized prepulse. This high number for the CE compares well with other experiments with solid glass and metal targets (e.g.[Dunne et al., 2000, Spitzer et al., 1996]). The maximum CE of 1.1% in $2\pi$ and per 2.5% bandwidth was obtained with 6 ns pulses at a laser pulse energy of 150 mJ, corresponding to an intensity of $I_{\text{laser}} = 8 \times 10^{12} \text{ Wcm}^{-2}$. Due to the essentially unlimited number of particles in the region of the laser plasma interaction, the CE dependence on the laser pulse energy is not limited by the number of particles of the target, but the increase of the CE is limited instead by the intensity. For the long (ns) pulses, a maximum in the conversion efficiency is observed by varying the energy of the laser pulse (squares in Fig. 4.17), similar to results reported by Schriever et al. [Schriever et al., 1998b]. The maximum can be qualitatively explained [Schriever et al., 1998a] by
assuming a Planckian emission for the plasma and a temperature scaling of $k_B T \propto I^{4/9}_{\text{laser}}$ according to Eq. (3.15). Using these two equations a maximum in the conversion efficiency is predicted. The heating becomes less effective for high intensities and the CE falls. However, these arguments to be valid only for long laser pulses in the ns regime. The CE should decrease further for higher laser intensities, but the measurement for a pulse duration of 120 ps shows a rising conversion efficiency.

![Figure 4.17: The 13 nm CE was measured for an extended quartz target as function of the laser pulse energy for two different pulse durations (120 ps / 6 ns).](image)

To compare the CE of the quartz and the droplet target, energies of 50 mJ or below seem to be better suited. The differing behavior of the two target schemes for high laser pulse energy ($> 100$ mJ) and ns long laser pulses has already been discussed. Focussing on the measured CE for 50 mJ laser pulses and a pulse duration of 120 ps which yields an optimum CE for the droplet target, the CE of the quartz target is only a factor of 3.6 higher than the CE obtained for the same laser parameters with the droplet target (see also Fig. 4.19). The higher CE measured for the quartz target can be explained as follows. First of all, the influence of the Si ions cannot be neglected. Due to the large number of outer electrons in the Si ions compared to the H ions, the average charge state $\bar{Z}$ is higher, providing a higher absorption (see Eq. (3.19)). In addition the higher mass slows the expansion down, due to their inertia, allowing more efficient heating. Secondly, the extended target expands almost one-dimensional in contrast to the three-dimensional expansion of the droplet plasma. The three-dimensional expansion dilutes the plasma faster and the cooling rate is higher. Therefore, the region of optimum plasma conditions for the highest CE (see Sect. 3.3.3) is traversed faster than in the one-dimensional case.

There are several experimental hints supporting these assumptions. First, the CE of ice is significantly higher than the CE of the water droplets. This cannot be attributed to the differing chemical composition. Furthermore, the expansion of droplets and extended targets has been visualized by K.-U. Amthor taking time-resolved shadowgraph images [Amthor, 2002]. The expansion was studied within the first 3 ns after the laser pulse. An almost one-dimensional cylinder-like plasma expansion was observed for $\sim 30$ mJ fs-laser pulses impinging on an extended target, whereas the
droplets expanded spherically. In addition, the source size of the EUV emission was found to be a symmetric plasma sphere (for sub-ns laser pulses) in the case of the droplet target (Sect. 4.6.2).

### EUV CE dependence on the laser pulse duration

Fig. 4.18 shows the conversion efficiency as a function of the laser pulse duration from 200 fs to 6 ns. The general trend is again quite different from the one obtained with the droplet target (see Fig. 4.7). Since there is always enough target material present counteracting the plasma expansion, ns laser pulses can still be efficiently absorbed. As expected from the increasing scale-length and absorption (see Sect. 3.2.2), the CE is rising from fs to ns pulse durations without maximum. However, the conversion efficiency increases logarithmically over almost five orders of magnitude.

![Figure 4.18](image)

**Figure 4.18:** The dependence of 13 nm CE on the laser pulse duration is shown for an extended quartz target ($E_L = 250$ mJ). The inset plots the same data in a double logarithmic scale. The red line indicates the scaling of the CE with $\tau_L^{0.33}$ (see Sect. 3.2.2).

Similar measurements, detecting $\sim 1$ keV radiation from laser generated plasmas, show a rise of efficiency with increasing pulse duration but cannot develop a logarithmic rule due to only small variations in their pulse duration [Altenbernd et al., 1997, Broughton & Fedosjevs, 1993, Eidmann & Schwanda, 1991, Garban-Labaud, 1982]. As already discussed in Sect. 3.2.2 an empirical scaling of the CE with the laser pulse length to the power of 0.3 was found by Ref. [Altenbernd et al., 1997] for short ps pulses. This law seems to agree well with the dependence of the short pulses, however it does not describe the CE measured for the longer pulses at all (see inset of Fig. 4.18). The laser-plasma interaction spans an enormous range from collective processes dominating the short pulse interaction to hydrodynamic equilibrium physics with ns pulses. To date, no theoretical model is able to cover this large parameter range [Gibbon & Förster, 1996]. Thus, the astonishing but very distinct rule of the logarithmic increase of the CE still waits for explanation.
4.4.3 Preplasma dynamics

It was shown in the previous sections that the plasma dynamics is quite different for the droplet target and the extended one, when the target is irradiated by the main pulse. The complete droplet is evaporated in such an interaction. The situation changes when only the prepulse impinges on the target. Laser pulse energies of several hundred $\mu$J up to 2 mJ only produce a plasma sheath at the target surface. To investigate this dynamics, the dependence of the EUV CE on the prepulse delay was investigated for identical laser parameters but different targets (see Fig. 4.19). The measured values of the CE are shown as full symbols. The CE for water droplets (red circles) is much lower than the EUV CE for the quartz target (black stars). However, the general trend of both curves seems similar. Multiplying the CE values obtained for the droplet target by a factor of 3.6 as described in the last section, the dependence of the EUV CE on the prepulse separation is indeed identical for the two distinctly different target types. The similarity is quite surprising, since it was assumed that the preplasma (in analogy to the ‘main plasma’ expansion) around the droplet expands equally in three dimensions, in contrast to the more one-dimensional plasma expansion in the case of the extended solid target. This result might imply that the preplasma at the surface of a droplet is not spherically expanding for prepulses with only few mJ, but rather planar, thus justifying the one-dimensional simulation of the preplasma.

4.5 Ion velocities

To learn something about the expansion velocities of the laser-produced plasma, about the initial temperature in the interaction region and about the number of ions which are emitted from the plasma the ion current was measured with a Faraday cup. The Faraday cup was located 16 cm away from the laser-plasma interaction, measuring the ions which escaped from the hot and dense plasma region without having recombined. The basic idea of the device, shown in Fig. 4.20, is to separate...
the ion from the electron current. All deviations of charge neutrality are quickly counterbalanced by fast electrons and no net current can be detected, unless a bias of -70 V is applied to hinder the electrons from penetrating the Faraday cup. The measured signal is proportional to the number of ions multiplied with their charge. The Faraday cup signals obtained for various laser parameters are plotted in Figs. 4.21, 4.22 and 6.2. The arrival of the laser pulse was detected by a photodiode, setting the time-origin. The signal, seen at negative times, can be attributed to the electromagnetic noise pulse of the laser Pockels cells, whereas the much higher random fluctuations at short positive times are due to the electromagnetic pulse generated by the laser-plasma as well as due to fast electrons with energies high enough to enter the Faraday cup and finally due to ionizing radiation from the plasma, creating secondary electrons in the Faraday cup.

Looking at Fig. 4.21, where the dependence of the Faraday signal on the prepulse energy is shown, two observations are obvious: The ion current increases as soon as a prepulse is introduced, whereas the mean velocity decreases (the ion peak arrives at later times). In contrast to the previous findings, the plasma seems to be colder when a prepulse is introduced. This contradiction can be resolved by the MEDUSA simulation results. The simulated laser-plasma interaction shows that the fraction of absorbed laser energy does not change significantly with the introduction of a preplasma, however the ratio of the energy consumed for thermal energy and kinetic energy of the expansion is substantially altered. The fraction of the kinetic energy decreases from 70% down to 20% as soon as a prepulse is employed (see Sect. 5.2). Thus, a prepulse causes a better heating of the plasma, but in turn a slower expansion. Indeed, the simulation predicts a 30% decrease in the velocity in good agreement with the measurement. Due to the expansion of the prepulse, the interaction region of the laser pulse increases, when a preplasma is applied and thus, the number of accelerated ions increases.

The increasing amplitude of the noise signal with the prepulse energy at short times can be attributed to a higher number of hot electrons and increased emission of ionizing radiation. The increase of the ionizing radiation has already been discussed in detail (see Sect. 4.3.4). The increase of the number of supra-thermal electrons with the plasma scale-length by enhanced resonance absorption and parametric instabilities has been theoretically predicted (see Sect. 3.2.3) and measured (see Fig. 4.15).

The Faraday cup signal does not distinguish between hydrogen (deuterium$^1$) and oxygen atoms.

$^1$Some measurements have been performed with heavy water (D$_2$O), as will be explained in Sect. 6.1.1.
Since the mass difference is considerable, 1 (2 for deuterium) to 16, oxygen ions will arrive later. In thermodynamic equilibrium the temperature of both species is equal, resulting in a velocity difference of $\sqrt{16/2} = 2.8$ between deuterium and oxygen. And indeed, such a signal was observed for high intensity laser pulses (see Fig. 6.2). The velocity of the oxygen ions accelerated by laser pulses exceeding $10^{19}$ Wcm$^{-2}$ is much slower (maximum at 1.7 µs) than the signals obtained with weak 1 ps pulses (maximum < 1.3 µs). Thus, it seems obvious that the signal measured with the Faraday cup for short pulses up to several ps and a laser pulse energy of 20-50 mJ is only due to hydrogen (deuterium) ions. Knowing the mass (2 atomic units) and the charge ($Z = 1$) of the ions the temperature in the region of the laser-plasma interaction can be obtained. A velocity of $1.6 \times 10^7$ cm/s implies a kinetic energy of 250 eV. According to Eq. (3.11), which is appropriate for 2 ps pulses, the electron temperature at the time of the laser absorption is calculated to be 300 eV. This value fits quite well with the simulation results (see Fig. 5.1).

Observing the Faraday cup traces for 120 ps long laser pulses impinging on water droplets (red curve in Fig. 4.22) the signal is quite similar to the measurement with 2 ps pulses without prepulse. Introducing a prepulse for the measurement carried out with 120 ps pulses enhanced the signal, but the shift to later times (as observed in Fig. 4.21) was not observed. This is also expected from the MEDUSA simulation, which does not see a change in the ratio of kinetic to thermal energy for the longer laser pulses. However, there is a big difference when the same experiment is carried out with the quartz target (blue curve in Fig. 4.22). The signal is much higher, but the maximum of the ion current occurs at even shorter times. This is indeed surprising, because hydrogen ions cannot account for the Faraday cup signal, thus O and Si ions must have been detected. These heavy ions arrive at even shorter times than the deuterium ions in Fig. 4.21. This implies a 10 to 15 times higher kinetic energy.

**Figure 4.21:** Faraday cup signal for water droplets (here: D$_2$O) hit by a 2 ps laser pulse with an energy of 25 mJ. The prepulse energy was changed from 0 to 2 mJ at a delay of 4 ns.

**Figure 4.22:** The Faraday cup signal is shown for a water (H$_2$O) droplet target as well as for a quartz target interacting with a 120 ps / 25 mJ laser pulse.
4.5. Ion velocities

Calculating the plasma parameters predominating in the laser-plasma interaction region, Eq. (3.13) seems to be adequate for the long pulse duration and heavy ions O and Si (compared to H) for which recombination cannot be neglected. Thus a plasma temperature of $T = 340$ eV was calculated for an initial ionization stage of $\bar{Z} = 6$ (see Tab. 3.1). This temperature is again consistent with the simulations (see Fig. 5.1).

The plasma temperature inferred from the Faraday cup measurements decreases again for ns long laser pulses\(^2\) (see Fig. 4.23). Again, Eq. (3.13) was applied leading to a temperature of 210 eV for 3 ns pulses (25 mJ) impinging on water droplets. Thus droplets are heated best and to the highest temperatures by pulse durations in the range of 100 ps. This was already concluded from the spectra shown in Fig. 4.13.

![Figure 4.23](image-url) **Figure 4.23**: The Faraday cup signals for various laser pulse energies (pulse duration 3 ns) are shown. The peak of the signal is shifted to shorter times with increasing energy.

![Figure 4.24](image-url) **Figure 4.24**: The figure displays the peak velocity, taken from Fig. 4.23, as function of the laser pulse energy (blue dots). The red line was fit to the data, resulting in $v_{\text{ion}} \propto I_L^{0.20\pm0.01}$. By increasing the laser pulse energy for a constant pulse duration, the peak of the Faraday cup signal is shifted to earlier times (see 4.23). Thus, the mean velocity increases from 1.5 to 3 $\times 10^7$ cm/s and the calculated temperature rises from 180 eV to 450 eV for 10 mJ to 300 mJ laser pulses, respectively. The peak velocity as function of the laser pulse energy is shown in the inset in Fig. 4.23. The measured data was fitted by a power law resulting in $v_{\text{exp}} \propto I_L^{0.20\pm0.01}$. This is in good agreement with the expected scaling of $v_{\text{exp}} \propto I_L^{2/9}$ according to Eq. (3.14). The same dependence was also found for a steady-state ablation model by Ref. [Gupta et al., 1986]. Thus, the plasma ablation is indeed a steady state equilibrium process for the nanosecond long laser pulses.

It is important to note that the measured ion signal does not necessarily represent the number of ions created in the interaction, since the measurement takes place far away from the initial

\(^2\) A Nd:YAG laser (Infinity from Coherent) instead of the TW-Ti:sapphire laser was used for this experiment. The pulse duration was 3 ns at a wavelength of 1064 nm.
4.6. Characterization of the EUV source

4.6.1 Pulse duration of the EUV emission

In order to find other applications for the laser-plasma produced EUV radiation, e.g. time-resolved EUV pump-probe experiments, it is necessary to know the EUV pulse duration [Nakano et al., 1996]. In addition, the simulations predict an increase of the EUV pulse duration with growing prepulse delay (see Sect. 5.4). To investigate this prediction, a streak camera was coupled to the transmission grating spectrometer to get time-resolved EUV spectra (the setup is described in Sect. 2.3.1). The photo cathode of the streak camera was insensitive to 13 nm, but radiation around 7 nm could effectively be detected. Since Si ions (Si$^{5+}$ to Si$^{8+}$) emit hundreds of lines in the vicinity of 7 nm
4.6. Characterization of the EUV source

[Kelly], the quartz target was used again. The result of the temporal evolution of the 7 nm radiation can be seen in Fig. 5.4 compared to the simulated 13 nm pulse durations (Fig. 5.5). Both figures show an increasing EUV pulse duration for increasing prepulse delay. The agreement between experiment and simulation is impressing.

4.6.2 Determination of the EUV source size

To use this kind of EUV source for lithographic applications, the source size has to be below a certain limit. This limit is currently set to a source diameter of 1.3 mm [Stamm et al., 2002]. To make sure that this condition is fulfilled, the source size was measured for short pulses (2ps with 25 mJ laser pulse energy) preceded by a 2 mJ prepulse (Fig. 4.25) and for 3 ns pulses\(^3\) with a laser pulse energy of 100 mJ to assure that the limits set by the industry are not exceeded by any laser parameter. Instead of using a rather complicated pinhole camera setup, a razor blade was utilized in order to determine the source size. The razor blade was positioned 1 mm apart from the plasma, between plasma and EUV detector (EUV monochromator device - Sect. 2.3.3), blocking the EUV radiation in the direction of the detector. By moving the blade slowly out of the line of sight of the detector, part of the EUV emission can reach the detector. Moving the blade even further, eventually all EUV light coming from the source can be detected. The EUV signal detected by the EUV monochromator as function of the blade position can be seen in Fig. 4.25. Differentiating the obtained curve gives a measure for the source size in one direction. The same procedure has to be repeated for the perpendicular direction to get a fully characterized source size. Due to the geometrical influence of the 4 mm diameter photodiode used in the detector, the actual source size is 12 \(\mu\)m smaller than the fitted FWHM in Fig. 4.25.

The 2 ps pulses resulted in a EUV source size of about 70 \(\mu\)m in diameter, spherically symmetric, whereas the 3 ns pulses produced a \(\sim 180 \mu\)m \(\times 260 \mu\)m diameter EUV spot. These sizes are quite comparable with the blurring observed in the transmission grating spectra as shown in Fig. 2.9.

\(^{3}\text{See footnote no. 2 on page 69}\)
4.6. Characterization of the EUV source

The spatial extent along the laser direction was found to be 40\% larger than in the perpendicular direction for the ns laser pulse duration. This can easily be explained by the expansion of the droplet during the laser pulse duration. The expanding plasma streaming along the laser axis can be heated during the pulse, whereas matter expanding perpendicular to the laser axis is not heated outside the focal region. In general the differing source size can be attributed to the larger plasma expansion for the ns pulses compared to the short ps pulses (see Sect. 4.3.3).

In summary, the source size was found to be small enough to fulfill the requirements needed for a high quality imaging source.

4.6.3 Angular distribution of the EUV emission

To built a high precision lithography tool capable of imaging nm size structures, the illumination has to be homogenous and isotropic. Thus the angular dependence of the EUV emission has to be measured to assure that the investigated source can be used for lithographic applications. The angular dependence was also needed for a more practical reason. The calibrated spectrometer detects only a very small fraction of the EUV photons emitted by the plasma. But in order to determine an EUV conversion efficiency for the laser-plasma interactions, the total energy emitted into the 13 nm line has to be evaluated. This is only possible when the angular distribution is known. To measure the EUV emission in various different directions, a goniometric type of mounting for the EUV monochromator was constructed. The EUV signal was recorded in an solid angle of $\pi/2$ in vertical and $\pi$ in horizontal direction. As expected from a symmetrically expanding plasma sphere, the emission was found to be isotropic within 15\% error for 3 ns pulse duration\(^4\) and a laser pulse energy of 100 mJ impinging on a 20 $\mu$m diameter droplet.

\(^{4}\)See footnote no. 2 on page 69
5 Simulation of the EUV measurements

As pointed out in the last chapter, only some of the experimental results can be understood by using simple models, like the scaling of the CE with the droplet size. In particular, the dependence of the CE on the prepulse delay for the various laser pulse durations has no obvious interpretation. In order to understand some of the underlying physics acting in the laser-plasma interaction, detailed simulations were performed.

The simulation of the EUV CE was performed in basically three different steps. The first step is to calculate the population density $n_u(n_e, T_i)$ of the upper 13 nm level as function of the ion temperature $T_i$ and the electron density $n_e$. This calculation has to be performed only once and the result is shown in Fig. 3.2. The second step is to simulate the laser plasma dynamics for the various laser parameters, yielding $n_e(x,t)$ and $T_i(x,t)$. These simulations were performed using the MEDUSA code (see Sect. 3.2.5). A computer program was written within the MatLab environment for the third step: the combination of the results derived by the two initial steps. The population density of the upper EUV level $n_u(n_e, T_i)$ was calculated for each point in space $x$ and each time step $t n_u(n_e(x,t), T_i(x,t)) \rightarrow n_u(x,t)$ resulting in time- and space-dependent population density. As shown in Eq. (3.43), the population of the upper level of the transition $\text{O}^{5+}(4d)$ is proportional to the 13 nm emission yield. Integrating over the spatial coordinate results in the time-dependent EUV emission as shown in Fig. 5.5. For most cases, the time-dependence is of no importance and therefore, $n_u(x,t)$ was integrated over both variables, providing a measure for the total EUV emission. By normalizing the calculation result by the laser energy used in the MEDUSA simulation, a measure for the EUV CE was obtained. Since the MEDUSA simulation is one-dimensional, the EUV emission provided by the simulation is calculated in arbitrary units, not in numbers of photons. The simulation result was scaled once to the experimental values (Fig.5.6 and Fig.5.7), and this scaling factor was used for all other figures as well. However, the aim of the work was not to reproduce the absolute CE value, but rather to simulate the qualitative behavior and learn from the simulation about the laser-plasma interaction.

5.1 Simulation of the electron density and the ion temperature

Laser pulse durations of 100 ps, 10 ps, 1 ps and 500 fs with a main pulse energy of 20 mJ and a constant prepulse of 2 mJ were simulated. 200 fs pulses, used in the experiment, could not be simulated by MEDUSA due to intensity limitations of the program. The calculation did not converge. By analogy with the experiment, the focus diameter was chosen to be 20 $\mu$m resulting in intensities ranging from $8 \times 10^{13}$ Wcm$^{-2}$ to $1.6 \times 10^{16}$ Wcm$^{-2}$. Prepulse delays up to 4 ns were simulated.
5.1. Simulation of the electron density and the ion temperature

Water plasma is essentially a plasma consisting of \( \text{H}^+ \) and \( \text{O}^{x+} \). Therefore, the atomic mass needed as an input parameter by the MEDUSA program was replaced by an average mass of 6 atomic units and the atomic charge was replaced by the average ion charge of 3.3 elementary charges. Plotting the ion temperature \( T_i \) and the electron density \( n_e \) as function of space, as determined by MEDUSA (see Fig. 3.1), does not help to reveal the substantial differences between the laser-plasma interaction of long (100 ps) and short (1 ps) laser pulses. However, plotting \( T_i \) as function of \( n_e \), as shown in Fig. 5.1, the difference is obvious. First of all, both long and short pulses exhibit a dependence on the prepulse delay. Generally speaking, the longer the delay, the hotter the plasma. However, the density at which the heating occurs is different. The simulation shows that the mean ion temperature after the main pulse produced by 1 ps pulses with or without a short (< 100 ps) prepulse delay is rather low (~ 100 eV). The simulations performed for longer prepulse delays of 500-700 ps showed that mean ion temperatures for short pulses are comparable to the temperatures obtained for the 100 ps pulses without prepulse (~ 300 eV). Using even longer prepulse delays ion temperatures exceeding 450 eV can be reached for 1 ps laser pulses. The same behavior can indeed be found in the measured spectra. Since a plasma with temperatures of several hundred eV emits at wavelengths considerably shorter than 13 nm, the magnitude of the radiation at short wavelength (< 8 nm) gives a measure of the initial ion temperature during and shortly after the laser pulse impinged onto the target. Looking at Fig. 4.13, 120 ps pulses heat the plasma to higher temperatures than 1.8 ps pulses do when no prepulse is introduced; whereas the spectrum acquired for 1.8 ps pulses preceded by a prepulse (see Fig. 4.12) looks similar or even hotter than the spectrum produced by the 120 ps pulses.

To understand the prepulse-dependent heating of the plasma, the laser absorption process has to be examined. As discussed in Sect. 3.2.2, the dominant absorption mechanism in the under-
critical plasma is certainly collisional absorption. Fig. 5.2 shows the absorption coefficient of the pre-formed plasma right before the arrival of the main pulse. The absorption coefficient was calculated by Eq. (3.30), taking the intensity-dependence into account. The parameters $n_e$, $Z$ and $T_e$ were taken from the MEDUSA output. The collisional absorption coefficient increased with the electron density until the critical density is reached, beyond which the laser light cannot propagate. The absorption coefficient is shown for different prepulse delays, showing that the absorption increases clearly with the prepulse delay due to the larger scale length, as expected from Eq. (3.26). However, the absorption coefficient for the preplasma produced with short pulses is lower than for the longer pulses. Short and therefore intense pulses are not as efficiently absorbed in the underdense plasma as the longer pulses are. Thus, short pulses can penetrate deeper into the plasma and heat denser plasma regions without losing too much energy, heating the thin preplasma (Fig. 5.1 a). By contrast, 100 ps pulses are already partially absorbed in lower density plasma (Fig. 5.1 b). As soon as the 100 ps long main pulse reaches the critical density layer, the plasma created by the main pulse itself dominates the absorption, so the ion temperatures at higher electron densities do not depend on the prepulse delay anymore. This can be understood by looking at the ion velocities in the plasma. According to Sect. 4.5, the ion velocity within the plasma is about $10^7$ cm/s, or in more practical units: $0.1 \mu$m/ps. There is almost no movement during the interaction of a 1 ps laser pulse. The interaction of such a short pulse is dominated by the preformed plasma extending over several $10 \mu$m. In contrast, the plasma cannot assumed to be motionless for 100 ps long laser pulses. The high density region at the critical density, where most of the laser absorption takes place, is altered significantly during the interaction with the main pulse. Therefore, only the heating of the thin preplasma several $10 \mu$m in front of the target is different for the various prepulse delays.

For 1 ps pulses and 20 mJ pulse energy, the plasma scale-length at the critical density, in units of the laser wavelength $L/\lambda$ ranges from $< 1$ with out prepulse increasing to 30 at 700 ps delay and finally decreasing again. In the case of 100 ps pulses, the main pulse creates its own scale-length of $L/\lambda \sim 50$. 

**Figure 5.2:** The inverse bremsstrahlung absorption coefficients of the preplasma, created by 1 ps (dashed lines) and 100 ps (solid lines), are plotted for different prepulse delays. The absorption coefficients are calculated according to Eq. (3.30).
So far, only collisional absorption was considered, but at the intensity of $8 \times 10^{15}$ Wcm$^{-2}$ as achieved with the 1 ps pulses, collisionless mechanisms have to be taken into account as well (see Sect. 3.2.3). Besides theoretical arguments for collisionless absorption at this intensity there is a large body of experimental evidence that collisionless absorption takes place in the measurements for 1 ps and shorter pulses (see Sect. 3.2.3, 4.3.6 and 4.5).

It was already shown that the collisional absorption is the dominant process for the prepulse-dependent heating, but what is the role of collisionless absorption?

![Figure 5.3:](image)

The collisionless absorption, namely for resonance absorption, is a function of the preplasma scale-length (see Sect. 3.2.3). The absorption increases with increasing scale-length for s- as well as for p-polarized light [Gauthier, 2001]. In addition, the angle of incidence at which the absorption reaches its maximum for p-polarized\(^1\) light approaches normal incidence with larger scale-length [Fedosejevs et al., 1990, Gauthier, 2001]. Due to the spherical geometry of the droplet, most of the laser beam impinges under angles close to normal incidence. Therefore, a prepulse enhances the collisionless absorption as well as the collisional.

By changing the fraction of the collisionless absorbed energy, which is an input parameter in the MEDUSA code, it can be seen that the collisionless absorption influences the density at which the ions are heated (see Fig. 5.3). This influence was observed in the simulations performed for short pulses ($\leq 1$ ps), while no changes at all were observed when the fraction of collisionless absorbed energy was varied for the 100 ps laser pulses, as expected.

In the case of short pulses, the fraction of the collisionally absorbed laser energy was varied from 2%, which is certainly too low to explain the observed amount of x-rays in the experiment, up to 50%, matching the maximum fraction of energy that can be absorbed by resonance absorption [Forslund et al., 1975]. The density of the plasma which is heated by the laser pulse changes greatly and thus the EUV yield. However, by changing the fraction of collisionless absorbed energy, it was possible to observe the effect on the ion temperature.

\(^1\)The maximum of the absorption of s-polarized light is always found at normal incidence [Gauthier, 2001].
energy in the range of 10% to 30% which is reasonable according to the discussion in Sect. 3.2.3, the EUV yield, calculated from the simulation result, was not strongly influenced. The fraction of collisionless absorbed laser energy was set to 20% and the fraction of energy absorbed by collisionless processes which is converted into suprathermal electrons was set to 10% (see Sect. 3.2.5).

The influence of the collisionless absorption component can be understood by taking into account that the ions are not heated by the laser pulse itself, but they are rather heated and excited by electron collisions (see e.g. Sect. 3.2.2 or 3.3.5). By increasing the fraction of energy deposited in collisionless processes, the number of suprathermal electrons increases. Since the cross section for electron-ion collisions decreases with one over the fourth power of the electron velocity (see Eq. (3.29)), these fast electrons can deeply penetrate into the overdense plasma and heat the ions at densities $\gg n_{\text{crit}}$ as shown in Fig. 5.3.

## 5.2 Laser energy deposition and EUV CE

The MEDUSA output provides the fraction of laser energy absorbed by the plasma. As expected from several experiments (e.g. [Garban-Labaune, 1982, Wood et al., 1988]), the absorbed energy is highest for the 100 ps pulse (40%) and decreases with the pulse length to about 20% for the 1 ps pulses [Price et al., 1995]. MEDUSA subdivides the energy absorbed by the plasma in thermal energy and kinetic energy of the expansion. The fraction of the absorbed laser energy does not change much (less than 30%) with the prepulse delay. The ratio of kinetic and thermal energy on the other hand is strongly dependent on the prepulse for the 1 ps pulses. The fraction of the thermal energy increases from 25% of the absorbed energy without prepulse, to almost 80% with 700 ps delay. Thus the plasma is heated more efficiently when a preplasma is present. The ratio of kinetic and thermal energy on the other hand does not strongly depend on the prepulse delay when the simulation is performed for 100 ps pulses and the fraction of the thermal energy was about 50%. This is not surprising since the main pulse interaction is not strongly influenced by the preplasma. Just this behavior was indeed found experimentally by the ion velocity measurements described in Sect. 4.5. Still, the 100 ps pulses couple more thermal energy into the plasma than the 1 ps ones, but to have efficient EUV production it is not sufficient to get as much laser energy as possible into the plasma: From Fig. 3.2, it is clear that the energy has to be absorbed in such a way as to maximize the volume with density close to the critical density and a temperature of about 30 eV. Right after the main pulse interaction there is no such plasma (see Fig. 5.1). Following the temporal evolution of the plasma (not shown), the initially hot plasma cools down and expands in a way that plasma with initial electron densities $> 10^{22} \, \text{cm}^{-3}$ and temperatures above 100 eV finally evolves into plasma with the optimum EUV conditions. Therefore, heating of high density plasma will result in an efficient EUV production. The higher the temperature and density initially,
the longer it takes for the plasma to cool down and expand to the optimum plasma conditions for 13 nm emission of $2 \times 10^{21} \text{ cm}^{-3}$ and 25 eV, respectively. The volume of the 13 nm emission region is also increasing. This enlargement, however, was found to be small in comparison to the temporal elongation of the EUV emission with increasing prepulse delay.

5.3 Simulated EUV pulse duration

The temporal behavior of the EUV emission is obtained from the simulation result by simply retaining the time-dependence of the population density $n_u(x,t)$. The introduction of a prepulse results in a longer EUV emission duration and a somewhat larger EUV emitting volume and therefore a higher CE compared to simulations without prepulse. An EUV emission duration between 200 ps and 1 ns, as plotted in Fig. 5.5, clearly shows that the EUV radiation is still emitted long after the laser impinged on the target. This fact was already inferred from the similarity of the EUV spectra in the range of 10 nm to 18 nm for extremely different laser parameters as shown in Fig. 4.13.

![Figure 5.4: Streak camera measurement of the EUV ($\lambda_{\text{EUV}}=7 \text{ nm}$) pulse duration for different prepulse delays. The laser pulses were 2 ps long with a pulse energy of 150 mJ, preceded by a 3 mJ prepulse.](image1)

![Figure 5.5: Calculated pulse duration of the 13 nm EUV emission, plotted for different prepulse delays. The simulation was performed for 1 ps pulses with 20 mJ energy and a 2 mJ prepulse.](image2)

In order to experimentally verify this rather surprising prediction, a streak camera was coupled to the transmission grating spectrometer (see Sect. 2.3.1). The experimental results for radiation around 7 nm, as plotted in Fig. 5.4, have already been discussed in Sect. 4.6.1. Comparing this figure with the simulated pulse duration (Fig. 5.5), a good qualitative agreement is achieved. The emission time of the 7 nm radiation, however, is shorter since the higher energetic photons result from a higher temperature plasma decaying faster (see e.g. Fig. 3.3).
5.4 Comparison of the simulated and the measured EUV CE

The EUV CE was simulated for each measured laser parameter combination. Plotting all these results in a one-dimensional graph would be confusing, since the laser pulse duration as well as the prepulse duration were altered. Therefore, the experimental and simulated results are mapped in a two-dimensional way as shown in Fig. 5.6 and Fig. 5.7. A good agreement between the simulated CE (Fig. 5.7) and the measurement (Fig. 5.6) is seen. Both simulation and experiment indicate an optimal EUV emission for pulse durations around 1 ps (preceded by a prepulse). The experimentally determined increase of the CE for 1 ps pulses without prepulse by a factor of 6-8 when a prepulse is introduced, can be found in the simulation, as well as the decrease of the CE for shorter and longer laser pulses. The lower EUV emissivity and the smaller increase of the CE with increasing prepulse delay for the 10 ps pulses compared to the 1 ps pulses is shown. The simulated CE for 100 ps pulses, however, is almost independent of the prepulse delay. This deviation from the experiment might be attributed to the three-dimensional plasma expansion during the laser pulse which is not included in the one-dimensional model. Such a one-dimensional model is sufficient as long as the plasma expansion is small compared to the transverse dimension of the plasma. The expansion velocity is on the order of $10^7$ cm/s (see Sect. 4.5) and, therefore, the plasma expansion during the 100 ps long laser pulse is comparable with the focus diameter. Thus, a pulse duration of 100 ps is at the limit of one-dimensional calculations.

Note that the interpolation between the measured or simulated data is only a guide to the eye.

---

**Figure 5.6:** The plot shows the measured dependence of the EUV CE on the prepulse delay as well as on the laser pulse duration. The prepulse delay was changed from 0 to 700 ps in 100 ps steps for laser pulse durations of 200 fs, 1.8 ps, 10 ps and 120 ps with a constant laser pulse energy of 20 mJ.

**Figure 5.7:** The simulated EUV CE is shown for the same laser pulse durations and prepulse delays as in the measurement (see Fig. 5.6). The absolute values of the CE are scaled to the experiment for best fit.
5.4. Comparison of the simulated and the measured EUV CE

The preplasma, by contrast, expands more slowly and remains almost one-dimensional even several ns after the prepulse impinged onto the target (see Sect. 4.4.3). Therefore, a good qualitative agreement of the measured and simulated EUV CE for 1 ps and 100 ps pulses can be achieved even for nanosecond long delays, as plotted in Fig. 5.8. The dramatic ($15 \times$) increase of the CE for 1 ps pulses as well as the roughly constant ($\sim \times 2$) CE for 100 ps laser pulses was well reproduced by the simulation.

For a higher pulse energy (100 mJ) the plasma is heated to somewhat higher temperatures, but the heating takes place at about the same electron densities as for 20 mJ pulses. Still, only higher temperatures at higher electron densities expand in a way that they end up in the region of the desired plasma parameters ($T_e = 30$ eV and $n_e \sim 2 \times 10^{21}$ cm$^{-3}$) as discussed in Sect. 5.2. The enhancement, therefore, does not depend on the laser energy, which in turn implies that the CE is lower for the 100 mJ pulses compared to 20 mJ pulses. In addition, the intensity is higher for higher laser pulse energies, leading to a lower absorption (Eq. (3.30)), which also limits the CE.

Figure 5.8: A comparison of the measured (taken from Fig. 4.10) and simulated CE is shown for laser pulse durations of 1 ps and 100 ps (20 mJ). Prepulse pulse delays of up to 4 ns were simulated.
6 Ultra-high intensity measurements

The Jena TW laser is certainly capable of producing much higher intensities than the several times $10^{16}$ Wcm$^{-2}$ as used in the EUV experiments. Shortening the pulse duration to 80 fs, boosting the energy up to 600 mJ and focusing the beam almost diffraction limited to 3 $\mu$m FWHM, results in $5 \times 10^{19}$ Wcm$^{-2}$. How these improvements were implemented is discussed in the Appendix A. As pointed out in the theoretical description of the ultrahigh intensity laser-plasma interaction (Sect. 3.2.4), the quiver velocity of the electrons situated in the laser focus becomes relativistic and the energy scale of interest is no longer eV but rather MeV (e.g. [Düsterer et al., 2001b, Düsterer et al., 2001c, Schwoerer et al., 2001]).

6.1 Laser-triggered fusion

6.1.1 Introduction

Fusion reactions initiated by laser-plasma interaction, were the domain of large scale, factory size lasers [Lindl, 1995], until Pretzler et al. [Pretzler et al., 1998] and Ditmire et al. [Ditmire et al., 2000] observed first fusion neutrons from tabletop short pulse lasers, using the CPA technique, a few years ago. The main reaction detected in these experiments is the fusion of two deuterium nuclei resulting in a $^3$He ion, a neutron and 3.27 MeV of excess energy in the center-of-mass system of the reaction [Pretzler et al., 1998]:

$$d + d \rightarrow ^3\text{He}(0.82 \text{ MeV}) + n(2.45 \text{ MeV})$$  \hspace{1cm} (6.1)

The fusion energy is subdivided between both reaction products according to the energy and momentum conservation. 50% of the fusion interactions undergo this reaction channel, while the remaining 50% of the fusion reactions end up with a tritium nucleus and a proton, which are hard to detect.

Again, a comparison of an extended bulk target (4 cm diameter 0.5 cm thick) consisting of deuterated plastic (CD$_2$) with a mass-limited 20 $\mu$m diameter droplet target was performed. In order to provide deuterium in the form of a mass-limited target, heavy water (D$_2$O) instead of ‘regular’ water was used. The relevant parameters of both liquids are close enough to ensure a stable droplet formation for D$_2$O, too.

Fusion can only take place if the thermal energy of the deuterium nuclei is high enough to overcome the Coulomb barrier rejecting them. The cross-section for the fusion reaction is shown in Fig. 6.1. Few fusions are already taking place at low kinetic energies of 10 keV, while the
6.1. Laser-triggered fusion

![Figure 6.1](image1.png) **Figure 6.1:** d-d fusion cross-section as function of the d-ion energy [FENDL]. 1 barn = $10^{-24}$ cm$^2$

![Figure 6.2](image2.png) **Figure 6.2:** The Faraday cup signal was recorded for a 20 µm droplet, hit by a 600 mJ, 80 fs pulse focused to $5 \times 10^{19}$ Wcm$^{-2}$. The separation between deuterium and oxygen ions is obvious.

cross-section increases tremendously for higher ion energies. The mean ion energy in the laser-produced plasma was measured with the Faraday cup (see Sect. 4.5). A typical ion TOF spectrum recorded for a 20 µm diameter heavy water droplet, hit by a $5 \times 10^{19}$ Wcm$^{-2}$ laser pulse, is shown in Fig. 6.2. By converting the flight time into the particle velocity and further into the kinetic energy this curve can be fitted by a Maxwellian distribution (Eq. (3.2)) providing a temperature of about 1 keV - much too low to achieve fusion. Even the high energetic tail of distribution does not lead to a significant number of fusion reactions. Thus, the majority of ions will not undergo fusion reactions, which is the big difference to inertial confinement fusion [Lindl, 1995].

Only a small fraction of ions is accelerated to considerably higher temperatures of several hundred keV by electro-static fields arising within the laser-produced plasma [Izumi et al., 2002]. The impinging laser pulse accelerates the high-energetic fraction of electrons to temperatures of up to 2 MeV according to Eq. (3.34). These electrons, mainly accelerated at the critical density$^1$, result in a lack of electrons within a ridge around the laser focus. This leads to electro-static acceleration fields with a field strength on the order of MV/µm [Karsch et al., 2002]. Some ions are thus accelerated by this field to energies of several hundred keV, compensating the space charge.

### 6.1.2 Energy distribution of fusion neutrons

Certainly, the fusion neutron has always same energy in the center-of-mass system (2.45 MeV) and the emission characteristic is almost isotropic. But one deuterium ion (resulting from the laser

$^1$Since the critical density depends on the mass of the electron (Eq. (3.7)), the critical density increases as the mean electron energy increases due to the relativistic energy-momentum relation.
plasma) is generally much faster than the other one (from the surrounding material) leading to a moving center-of-mass. A fusion neutron produced in such an interaction has a higher kinetic energy (‘blue-shifted’) if it is emitted in the direction of the center-of-mass motion or a lower energy if emitted backwards (‘red-shifted’). The initial motion of the ion is thus imprinted on the neutron. Therefore, the neutron energy distribution can be used to trace back the ion motion within the hot plasma [Hilscher et al., 2001]. No other method can measure the ion velocities in the hot center of the laser-plasma interaction without influencing the interaction.

The kinetic energy of the d-d fusion neutrons is about 2.4 MeV, plus or minus several hundred keV depending on the initial deuterium ion energies. The neutron velocity is therefore about $2.2 \times 10^9$ cm/s or in more practical units: 2.2 cm/ns. This is 100 times faster than the expanding plasma moves, but still slow enough to be measured by a time-of-flight (TOF) detection scheme. The scintillation neutron detector\(^2\) was placed 240 cm away from the point of the laser plasma interaction and at an angle of 145\(^\circ\) degrees to the laser axis. The scintillator material (NE110) contains hydrogen atoms (protons), which can be accelerated efficiently when hit by a neutron. The neutron kicks out the proton which subsequently leads to an ionization track. Light is emitted while the track recombines. This light flash is then detected by a photomultiplier. A typical signal obtained by the scintillation detector is shown in Fig. 6.3. The time origin is set by the x-ray flash produced by the laser-plasma interaction, whereas the second peak, typically 120 ns later, denotes the neutron signal. Additional peaks at later times result from scattered fusion neutrons. The probability of finding a neutron produced by cosmic radiation in the observed time window is negligible. The complete scintillation detector is shielded by 9 cm of lead to suppress scattered x-ray signals leading to additional peaks. Only unscattered MeV x-ray photons can penetrate the shielding, providing the time origin. The neutrons are only weakly influenced by the lead.

Fig. 6.4 (a) shows the time-of-flight spectrum from the laser-plasma interaction with the droplet target. The solid angle of the detector was chosen in such a way that on average, a neutron signal

\(^2\)The detector was provided and operated by S. Karsch from the Max-Planck-Institut für Quantenoptik in Garching.
was detected only every fourth laser shot. Thus, a measured neutron signal results from only one single neutron. No pileup effects have to be taken into account. Plotting all the measured neutron flight times for 40,000 laser shots, a neutron TOF spectrum as shown in Fig. 6.4 (a) was obtained. While the absolute position (mean flight time) yields only the 2.45 MeV fusion energy, the width of the neutron TOF spectrum provides information about the temperature of the deuterium ions. The number of accelerated ions can be inferred from the signal height, in addition. A three-dimensional Monte Carlo neutron transport code, written by S. Karsch [Karsch, 2002], was used to calculate the hot ion temperature as well as the approximate number of these highly energetic ions from the TOF spectrum. About $7 \times 10^{10}$ deuterium ions with a temperature of 250 keV were obtained by fitting the experimental spectrum. This implies that 0.2% of the ions contained in the droplet have been accelerated to such a high energy, carrying 3 mJ of kinetic energy.

These fast ions, with a temperature of 250 keV, leave the focal region with a speed of 4.4 $\mu$m/ps - too fast for the droplet to become ionized or expand significantly. Thus, the fast ions created at the frontside of the droplet, and which are accelerated into the droplet cannot penetrate very far. They are hindered by 20 $\mu$m of water. In fact just 4 $\mu$m of water is sufficient to stop a 250 keV deuterium ion, according to database values for the stopping range [Berger et al.]. If the droplet is ionized or expanded, the stopping will be significantly reduced [Karsch et al., 2002]. However, the ionization front and the expansion of the plasma are happening on a 40 times slower timescale since both processes are related to the ion sound speed [Hall et al., 1998, Young et al., 1998]. Ionization due to space charge effects induced by fast electrons can also be neglected [Karsch et al., 2002]. Thus, the droplets can be considered as solid extended target for the ions accelerated in the laser-plasma. Indeed the experiments performed with the extended deuterated plastic target shows a similar neutron TOF spectrum. However, the big difference between bulk and mass-limited target lies in the fact that fast (MeV) electrons can easily penetrate 20 $\mu$m of water without significant energy loss [Berger et al.] in contrast to the thick plastic slab. This enables the use of the droplet target for an even more exciting experiment, as discussed below.

![Figure 6.4: TOF neutron spectra, recorded without catcher (a) and for different catcher distances of 8.4cm (b), 14.8cm (c), 25.0 cm (d). The smooth curves represent the simulated spectra.](image-url)
The direct heating of the ions within the initially created laser-plasma, at the frontside of the droplet so to speaking, is not the only way to accelerate ions to high energies. Despite accelerating ions at the frontside, ions coming from the backside of the target have been observed [Mackinnon et al., 2001, Snavely et al., 2000] and theoretically explained [Wilks et al., 2001] as well. A sketch of this process is shown in Fig. 6.5. The mechanism is similar to the acceleration at the frontside, however, the electrostatic field builds up at the surface of the droplet rather that at the critical density within the laser focus. In analogy to published experiments, the ions accelerated at the surface of the droplet will be called backside accelerated ions. The fast (MeV) electrons, accelerated within the laser focus, can easily penetrate the droplet, in contrast to the ions. Such a bunch of electrons, leaving the droplet, creates a strong electric field at the surface of the droplet due to the growing space charge. This electric field, in the range of MV/µm [Mackinnon et al., 2001], rips off the lightest ions (deuterium in our case) from the backside and pulls them behind the electron bunch to neutralize the space charge. Only protons originating from oil or water contaminants of the target backside, were ripped of the backside of the target in the cited experiments. To avoid this undefined acceleration of contaminations and accelerate only deuterium ions, a ‘self-refreshing’ target had to be used. The fast moving droplets stay only for several tens of µs within the vacuum before they are hit by the laser. This time is too short for any contamination of the droplet surface. By choosing the droplets as laser target, it was possible to produce a defined deuterium ion beam from the backside of the droplet. In order to measure the backside ion beam, again fusion reactions were utilized.

![Figure 6.5](image-url)

Figure 6.5: This figure shows a schematic sketch of the neutron TOF experiment. The laser pulse hits the target producing ‘frontside’ fusion neutrons within the laser-plasma. d-ions from the back side are accelerated by the charge separation field induced by fast electrons (inset). When these fast ions hit the catcher target they undergo fusion, producing and undergo a fusion reaction in the catcher and lead to the ‘backside’ neutron peak.

A block of deuterated plastic (50mm diameter), so-called catcher, was placed several centimeters behind the droplet target, in laser propagation direction (see Fig. 6.5). The deuterium ions accelerated at the backside of the droplet eventually hit the deuterated catcher target, initiating fusion neutrons. These neutrons will generate a similar peak in the TOF neutron spectrum as the

---

3Under certain conditions, like very thin targets (< 2µm) [Machsimchuk et al., 2000] or very high proton energies (> 4 MeV) [Clark et al., 2000], it is possible that ions detected behind the target in fact penetrated the target and originate from the frontside.
neutrons resulting from the frontside of the droplet, but however delayed by the time the deuterium ions need to get from the backside of the droplet to the catcher.

Fig. 6.4 (b-d) shows the resulting neutron TOF signals. Indeed two peaks are observed. In order to be sure that the second peak is not due to neutrons scattered from the catcher, the catcher was covered by a 200 µm thick un-deuterated plastic shield, blocking the deuterium ions. Having the cover in front of the catcher, no second peak was observed. Thus the neutrons detected in the second peak originate indeed from fusion in the catcher target. The time difference between the peak due to the frontside neutrons starting from the droplet and from the peak resulting from the catcher yields a mean kinetic energy of 140 keV for the backside accelerated ions. The second peak is broadened by the velocity dispersion of the ions. Fast deuterons arrive earlier at the catcher than slower ones, leading to a broader peak. Again using the 3D Monte Carlo code [Karsch, 2002] shows that significantly more ions were accelerated at the backside of the droplet compared to the frontside. About $1.9 \times 10^{11}$ deuterium ions, accelerated at the backside of the droplet, hit the catcher and induced fusion neutrons. The catcher covers 1.6 sr behind the droplet (at a catcher distance of 8.4 cm). Nuclear track detectors (CR39) were used to determine the complete angular distribution of the ions coming from the surface of the droplet. The emission was found to be isotropic in $4\pi$ which is also in good agreement with the findings from Ref. [Snavely et al., 2000], claiming that the ions are always accelerated perpendicular to the surface. Scaling the observed number of ions within 1.6 sr to $4\pi$, about $1.5 \times 10^{12}$ deuterium ion were accelerated at surface of the droplet. The same number of ions was also calculated for the other measured catcher distances, as expected from a isotropic emission. The temperature of these ions is $100 \pm 30$ keV which is somewhat lower as compared to the frontside accelerated ions. More details about detection and modeling of TOF spectra can be found in Ref. [Karsch, 2002] and [Karsch et al., 2002].

The ions accelerated at the surface of the droplet contain about ten times more energy as compared to the frontside. About 30 mJ were converted to the surface accelerated ions, equivalent to $\sim 5\%$ percent of the incident laser energy. This number is consistent with other experiments [Fews & Norreys, 1994] and simulations [Wilks et al., 2001]. About 15-20% of the incident laser energy is converted into fast electrons (see Sect. 3.2.4). In turn, about one quarter of the electron energy is transferred into the hot ion population, according to Ref. [Wilks et al., 2001]. Thus one ends up with the measured 5% of the initial laser energy converted into fast ions.

### 6.2 Laser-triggered fission

As described above, a TW-ultra short pulse lasers can trigger fusion reactions leading to the emission of neutrons. However, the extreme conditions within such a hot plasma can also initiate fission of nuclei [Cowan et al., 2000, Düsterer et al., 2001b, Düsterer et al., 2001c, Ledingham et al., 2000, Schwoerer et al., 2001, Schwoerer et al., 2002]. The laser radiation itself is certainly not capable of
splitting nuclei\(^4\), but the high energetic bremsstrahlung produced by laser accelerated electrons can initiate photo-fission reactions. Bremsstrahlung exceeding a certain energy threshold can decompose nuclei, leading energetic fragments. The reaction under investigation is a \((\gamma,n)\) type reaction.

\[
{}^9\text{Be} + \gamma \rightarrow \alpha + \alpha + n \tag{6.2}
\]

The bremsstrahlung photon \(\gamma\) kicks out the photo-neutron. Subsequently, the instable \(^8\text{Be}\) nucleus decays into two \(^4\text{He}\) cores - \(\alpha\) particles. The energy of the \(\gamma\) photon has to exceed a certain threshold of 1.67 MeV to initiate the reaction. This threshold is very low compared to other elements and furthermore, the natural abundance of \(^9\text{Be}\) is 100%. The cross-section of the reaction almost constant at \(~0.5\) mbarn at the threshold and it increases slightly to 1 mbarn at \(\gamma\)-energies of 6 MeV [Utsunomiya et al., 2000].

As discussed in Sect. 3.2.4, fast electrons arising from the laser-plasma interaction produce bremsstrahlung when colliding with ions. The bremsstrahlung yield, according to Eq. (3.35), increases linearly with the atomic number \(Z\) and with the square of the electron density within the solid, which is roughly proportional to the density of the material. To achieve the maximum number of MeV bremsstrahlung photons, tantalum was used, which has a rather high \((Z=73)\) mass and a high density \((\rho = 16.6\ \text{g/cm}^3)\). Focusing a 250 mJ, 80 fs laser pulse to \(5 \times 10^{18}\ \text{Wcm}^{-2}\) on a Ta target, the temperature of the hot electron population is expected to be 420 keV according to Eq. (3.34). The bremsstrahlung spectrum was measured using thermoluminescence detector (TLD) stacks as described in [Düsterer et al., 2001b, Düsterer et al., 2001c, Schwoerer et al., 2001]. The electron temperature was found to be anisotropic, due to relativistic effects, which have to be taken into account for an electron temperature comparable to the electron mass. The highest bremsstrahlung yield was found close to the laser reflection direction, produced by electrons with a temperature of 700 keV. Integrating over the bremsstrahlung flux yields about \(10^5\) photons above 1.6 MeV per sr solid angle. A beryllium disk (12 mm thick, 70 mm diameter) was placed 4.5 cm away from the laser plasma in specular direction. The Be slab covered a solid angle 1.3 sr. Therefore, \(1.3 \times 10^5\) photons with energies higher than 1.6 MeV were penetrating the Be each shot resulting in about 100 photo-neutrons per laser shot, according to the reaction cross-section and the geometry of the Be slab. The expected neutron flux was much smaller than in the previously described fusion experiments and in addition the spectral distribution exhibits a broad continuum, unlike the fusion neutrons. Thus, it is hard to obtain a TOF spectrum which can deliver information about the plasma conditions. A different detection scheme had to be used in order to gain a large enough signal. Using nuclear activation reactions for the neutron detection is much more promising. The choice fell on gold, which can capture thermal neutrons undergoing the reaction

\[
{}^{197}\text{Au} + n \rightarrow {}^{198}\text{Au}^* \tag{6.3}
\]

\(^4\)This might be possible for much higher intensities than produced today [Tajima & Mourou, 2002].
with a very high cross-section of 99 barn [Firestone, 1999]. Fortunately the natural abundance of \(^{197}\text{Au}\) is 100\% [Weast, 1980], making the process even more effective. The excited gold nucleus subsequently undergoes a beta decay, with a half-life period of 2.4 days, promptly followed by a gamma decay with the characteristic energy of 411.8 keV:

\[
^{198}\text{Au}^* \rightarrow ^{198}\text{Hg} + \beta^- + \gamma(411.8 \text{ keV})
\]  

Thus, the laser interaction indirectly transmuted gold into mercury, acting to a certain extend as an inverse philosopher’s stone. The problem was that the enormously high cross-section of the neutron capture reaction (6.3) is only attained for thermal neutrons at around ‘room temperature’ (\(\approx 25 \text{ meV}\)). The fission neutrons had to be thermalized, which is effectively done by light nuclei e.g. as hydrogen and carbon. The Be disk was mounted on the surface of a polyethylene sphere of 12.5 cm diameter, so-called Bonner sphere. Due to geometrical reasons, about 40\% of the neutrons generated in the Be disk entered the Bonner sphere and were thermalized by inelastic scattering with hydrogen and carbon nuclei. The neutron thermalization efficiency of the Bonner sphere was absolutely calibrated for the relevant neutron energies by the Physikalisch Technische Bundesanstalt (PTB). A 16 g gold disk was placed in the center of the Bonner sphere, capturing thermal neutrons. Due to the long lifetime of the \(^{198}\text{Au}^*\), the activations produced by 20,000 laser shots were accumulated. The 411.8 keV emission of the irradiated gold disk was subsequently measured by a germanium x-ray detector located 1 km beneath earth’s surface. The background radiation in this laboratory, operated by the PTB [Neumaier et al., 2000] has an extremely low ambient radiation dose of <1 nSv/h in comparison to \(\sim 270 \text{ nSv/h}\) at earth’s surface [Veith, 2001]. It was possible to record the small \(\gamma\) signal (0.0006 counts per second !) resulting from the \(^{198}\text{Au}^*\) decay as shown in Fig. 6.6 (blue curve) due to the low background and additional thick shielding. The spectrum shown was accumulated over 2 days (!). The red curve in Fig. 6.6 results from a control experiment. The 411.8 keV radiation from a unirradiated gold disk was measured, providing the neutron activation rate produced by neutrons induced by cosmic radiation [Neumaier et al., 2000]. This small naturally occurring contribution was subtracted. Taking the detector efficiency into account, 925 activations have been initiated by the laser triggered fusion neutrons within 20,000 laser shots.

**Figure 6.6:** The figure shows a \(\gamma\)-decay radiation spectrum of the excited \(^{198}\text{Au}^*\) gold disk. The blue curve shows the emission of a gold disk, which was exposed to laser-induced fission-neutrons, while the red curve presents the \(\gamma\)-yield of a reference gold sample.
shots. Calculating backwards, this number of activations implies that \(3 \times 10^4\) bremsstrahlung photons above 1.67 MeV must have penetrated the Be disk for each laser shot. Within the error bars of the measurement this is in good agreement with the TLD measurement. Thus, the nuclear reactions induced by the bremsstrahlung can be used to measure the integrated absolute bremsstrahlung photon yield above the threshold of the reaction. In fact it is possible to measure the bremsstrahlung photon energies below 3 MeV with other detectors (e.g. the TLD detector), however, for photon energies exceeding 3 MeV nuclear reactions are the only detection mechanism. By activating various elements with different thresholds even the bremsstrahlung spectrum can be inferred up to energies of several tens of MeV [Cowan et al., 2000, Ledingham et al., 2000, Schwoerer et al., 2002].
7 Conclusion

Summarizing the work presented in this thesis, major contributions to the enhancement of laser-plasma based 13 nm EUV radiation production and the understanding of the underlying physics have been shown. Besides the scientific gain, the efficient production of radiation at 13 nm is of crucial importance for the feasibility of the next-generation lithography in the semiconductor industry.

A novel target concept of mass-limited droplets was chosen for the experiments. In contrast to extended solid targets, which are already intensely investigated, few experiments have been performed with such kind of target, providing the possibility of low debris operation. The design of the droplet production unit operating in vacuum has not been described before and was thus patented [Ziegler et al., 2001]. Varying the laser parameters over a wide range of laser pulse durations and laser pulse energies, significant differences in the interaction of the laser pulses with solid extended targets and with the droplet targets have been revealed for the first time. It was shown that the EUV production yields an optimum for exactly one laser pulse duration and one laser pulse energy using a fixed target size (see Fig. 4.8). To generalize the obtained results for other target dimensions, scaling laws were proposed and experimentally verified. As result, the laser energy demanded for the optimum EUV CE scales with the droplet volume, whereas the optimum pulse duration scales with the droplet diameter. Thus, laser pulses can be tailored for every given target size or vice versa. These results have been published in Ref. [Düsterer et al., 2001d]

Considering the interaction of only one laser pulse interacting with the target, the EUV conversion efficiency of droplets is significantly lower than the CE for extended solid targets, in agreement to all other published results. This seems to be the price paid for debris-free operation of the EUV source. However, introducing prepulses preceding the main laser pulse, conversion efficiencies for the 13 nm radiation of up to 2% in 2.5% bandwidth and $4\pi$, quite comparable to xenon and solid extended targets, were achieved.

Prepulses alter the plasma conditions interacting with the main pulse. This different plasma environment causes the optimum laser parameters to shift from rather long ($\sim 100$ ps) pulses with 50 mJ laser pulse energy which are best when no prepulse is used to short pulses ($\sim 1$ ps) with 20 mJ. This exciting and new result could only be explained by plasma dynamic simulations. These simulations revealed the different interaction and heating mechanisms occurring when the main pulse impinges onto the preplasma. The dependence of the EUV CE on the laser pulse duration and the prepulse main pulse separation have been simulated, reproducing the experimental data well. The main results of these simulations are summarized in Fig. 7.1. This figure shows the measured EUV CE as function of the laser pulse duration and the prepulse delay, demonstrating the effects influencing the CE at a glance. Using this knowledge, the laser parameters were carefully adjusted to tailor the preplasma. Thus, the EUV CE was enhanced by a factor of 10 compared
to most other published values. Furthermore, the debris (fast ions) emitted by the laser-produced plasma was significantly reduced by using 2 ps laser pulses instead of commonly used ns pulses. This work will be published in Ref. [Düsterer et al., 2002].

**Figure 7.1:** This figure summarizes the effects influencing the EUV CE. The measured data are taken from Fig. 5.6.

In future, using pulse shaping techniques [Stobrawa et al., 2001], and self-learning algorithms [Feurer, 1999] it will be possible to enhance the EUV CE even more by fine-tuning the laser-plasma interaction, allowing the production of the very high EUV average power, demanded by industry, with reasonable laser systems.

Furthermore, it was demonstrated that the developed droplet target can also contribute to the investigation of ultrahigh intensity interactions. Exchanging the ‘regular’ water by heavy water (D₂O) and focusing the laser pulses very tightly to achieve an intensity of \( 5 \times 10^{19} \text{ Wcm}^{-2} \), a fraction of the deuterium ions was heated to temperatures exceeding 100 keV. Deuterium ions within such a hot plasma undergo fusion reactions, resulting in fusion neutrons. Up to \( 5 \times 10^4 \) neutrons per laser shot were detected. Since the neutrons are emitted from a small \( \mu \text{m-sized volume} \) and within a short, sub-nanosecond time [Ditmire, 2002] this source is one of the most brilliant neutron sources world-wide. This source will enable a whole variety of experiments utilizing the pulsed high neutron flux. The mass-limited target concept made it possible not just to create a powerful neutron source, but also to investigate a widely discussed question within laser-ion...
acceleration. Neutrons from fusion reactions of deuterium ions were utilized as tracer for the ion energies. By means of time-of-flight neutron detection, it was possible to measure for the first time ions accelerated at the front as well as backside of the target within one experiment and compare the number and kinetic energy of accelerated ions from both sides. The results will be published in [Karsch et al., 2002]. Comparing these data to simulation results will enable theoreticians to reveal details of the ion acceleration.

Finally, a different method of initiating neutron emission by laser-plasma interaction was presented. Focusing the intense laser pulse onto a high Z, solid target (Ta), accelerates electrons to relativistic energies of several MeV giving rise to bremsstrahlung with photon energies of several MeV. It was shown that this hard MeV x-ray radiation can indeed induce fission of beryllium nuclei, decomposing into two helium nuclei and a fission neutron. Recording the number of fission neutrons, the MeV x-ray yield can be inferred. Thus, the photo fission was employed as x-ray detector for very energetic photons. This was the first demonstration of a laser fission experiment conducted with a small tabletop laser system and is published in Refs. [Düsterer et al., 2001b, Düsterer et al., 2001c, Schwoerer et al., 2001].
8 Bibliography


A Description of the laser system

A.1 The oscillator - the front end

The front end of the laser system is a titanium:sapphire oscillator with a center wavelength of \( \lambda_L = 795 \text{ nm} \) (‘Tsunami’ from Spectra Physics). The oscillator is pumped by an all-solid-state, diode-pumped, frequency-doubled Nd:YVO\(_4\) laser (‘VERDI’ from Coherent) providing a single-frequency green output of 5.5 W at 532 nm (Fig. 2.1). The oscillator produces a train of laser pulses with a repetition rate of 82 MHz. The laser pulses have an energy of \( \sim 7 \text{ nJ} \) and a spectral bandwidth of \( \Delta \lambda = 21 \text{ nm} \). According to Fourier theory the pulse duration \( \tau_L \) (FWHM of the intensity) is related to the spectral bandwidth of a laser pulse \( \Delta \nu \) (FWHM) via:

\[
\tau_L \Delta \nu \geq 0.441 \quad \text{(A.1)}
\]

for a Gaussian shaped intensity profile [Diels & Rudolph, 1996]. For the center wavelength of 795 nm of the used laser and \( \Delta \lambda_L \ll \lambda_L \). The pulse duration \( \tau_L \) can be also expresses as:

\[
\tau_L \Delta \lambda \geq 930 \; [\text{nm fs}] \quad \text{(A.2)}
\]

The shortest possible pulse which is achieved when the equality holds is called the bandwidth limited pulse. Thus the bandwidth limited pulse duration produced by the oscillator is 44 fs.

A.2 Laser pulse expansion and compression

Equation (A.2) gives a lower limit on the pulse duration, but the pulse can be longer. It is easy to think of several irreversible methods to make the pulse longer, however, there are only few methods stretching a pulse reversibly [Diels & Rudolph, 1996]. The basic idea is just to delay some frequencies with respect to the others, resulting in a frequency sweep over the pulse duration - the pulse is chirped. To express the chirp mathematically, the pulse shape is assumed to be Gaussian. The electric field is given by:

\[
\mathcal{E}(t) = E_0 e^{-2\ln(2)(1-iA)(t/\tau)^2} \quad \text{(A.3)}
\]

where \( A \) is the chirp parameter defining the frequency sweep over the pulse duration. \( A \) can be positive or negative depending on whether high or low frequency are leading within the pulse. A connection of \( A \) to physical quantities will follow later. Calculating the time bandwidth product (Eq. (A.1)) in the frequency picture and including \( A \) one gets:

\[
\tau_{\text{FWHM}} \Delta \nu = \frac{2\ln 2}{\pi} \sqrt{1 + A^2} \quad \text{or} \quad \tau_{\text{FWHM}} = \frac{2\ln 2}{\pi \Delta \nu} \sqrt{1 + A^2} \quad \text{(A.4)}
\]
for the pulse duration. As soon as a chirp is introduced, the pulse duration will be longer.

In the first realization of a CPA system [Strickland & Mourou, 1985], the laser pulse was chirped by a nonlinear interaction with matter. However, the elongation of the pulse was rather small. Some years later [Martinez, 1987], the stretcher as will be described here was introduced to CPA systems. Meanwhile, stretching factors of several 10.000 have been achieved. The experimental setup of the stretcher consists of a 1:1 telescope and two antiparallel gratings. The dispersion of the gratings is used to introduce the chirp, whereas the telescope inverts the laser beam spatially, thus introducing a positive chirp ($\mathcal{A} > 0$). The relevant parameters of the stretcher setup are the tilt angle ($\beta$), the line spacing of the grating ($d$), the focal length ($f$) of the two telescope lenses and the spacing between the gratings and the lenses ($s_1$ and $s_2$) as well as the center frequency ($\omega_0$). By using only geometrical considerations, the group velocity delay (GVD) that is defined as second derivative of the phase of the laser pulse with respect to the frequency is given by [Svelto, 1998]:

$$ \text{GVD} = \frac{d^2 \phi}{d\omega^2} \bigg|_{\omega_0} = \frac{4\pi^2 c}{\omega_0^3 d^2 \cos^2 \beta} (2f - s_1 - s_2) \quad (A.5) $$

with $c$ as speed of light. Knowing all these parameters for our setup, a GVD of $2.1 \times 10^{-24} \text{ s}^2$ was calculated [Ziener, 2001]. The chirp is then given by:

$$ \mathcal{A} = \frac{1}{2} \Delta \omega^2 \frac{d^2 \phi}{d\omega^2} \bigg|_{\omega_0} = 2300 \quad (A.6) $$

where $\Delta \omega = 48 \text{ THz} \ (\Delta \lambda = 21 \text{ nm})$. Higher orders $d^n \phi/d\omega^n$ for $n > 3$ are neglected here, however they play an important role for pulses considerably shorter 100 fs. This is a ten thousand times larger GVD compared with the GVD induced by typical transparent media, like BK7 glass. The chirp, calculated in Eq. (A.6) together with Eq. (A.4), results in a stretched pulse duration of 130 ps for the Jena Terawatt system. The long, chirped pulse can now be amplified safely, without inducing optical damage due to high intensity and without significant nonlinear interactions of the laser pulse with the amplifying media. Nonlinear interactions always produce higher order phase distortions which become a problem when the stretched pulse has to be recompressed.

The stretched pulses are compressed by a setup conjugate to that of the stretcher. The compressor consists of a sequence of two parallel grating pairs without inverting telescope. The resulting GVD is always negative, equivalent to anomalous dispersion [Diels & Rudolph, 1996]. Therefore, the GVD induced by the stretcher can be neutralized with properly chosen grating line width, angle of incidence and grating separation. The four grating combination was chosen in order to minimize the thermal load on each of the gratings. In the current setup, four holographically produced gold gratings with 1200 lines per mm and a separation of $L_0 = 689 \text{ mm}$ are used to compress the amplified pulse back to $\tau_0 = 80 \text{ fs}$. Due to losses into zero and higher orders of the grating and absorption of the gold surface, the transmission of the compressor is only 65%, but however, it is still the most efficient way to produce sub 100 fs pulses with an energy of several hundred mJ.
Certainly, for the ultra-high intensity experiments, the entire chirp induced by the stretcher has to be compensated. Therefore, the grating separation is kept at \( L_0 \) for this type of experiment, whereas when the compressor is somewhat detuned \( L = L_0 + \Delta L \), not all the chirp is compensated, meaning \( |A| > 0 \) in Eq. (A.4), and thus, the pulse duration (\( \tau_{\text{exp}} \)) of the pulse, leaving the compressor in direction to the experiment, will be longer than \( \tau_0 \). Combining Eq. (A.4) and (A.6) together with the fact the chirp depends linearly on the grating detuning (\( A \propto \Delta L \))[Diels & Rudolph, 1996] results in the pulse duration as function of \( \Delta L \):

\[
\tau_{\text{exp}} = \tau_0 \sqrt{1 + \frac{1}{4} \left( \frac{\Delta \omega^2}{d^2 \phi} \frac{\Delta L}{L_0} \right)^2}
\] (A.7)

The pulse duration resulting from Eq. (A.7) is plotted as a function of the grating separation in Fig. A.1. The experimentally determined values agree well with the calculation.

![Figure A.1](image_url)

**Figure A.1:** The pulse length as function of the compressor detuning. The stars represent the measured pulse length, while the line is calculated by Eq. (A.7) without free parameters.

### A.3 Amplification stages

It is simply not possible to amplify every one of the 82 million pulses, leaving the oscillator each second, up to several hundred mJ. Thus, an electro-optical shutter, a so-called Pockels cell, reduces the pulse repetition rate to 10 Hz. The stretched pulse is injected, again by a Pockels cell, into the first amplification stage. The so-called regenerative amplifier is a linear cavity that acts as resonator and has its own modal structure, therefore regenerating the spatial pulse shape. The Ti:sapphire crystal in the regenerative amplifier is pumped by a frequency-doubled (532 nm) Nd:YAG laser (‘Surelite ’ from Continuum) with \( \sim 50 \text{ mJ} \) in 8 ns. The laser pulse oscillates back and forth in the cavity, and after passing 44 times through the Ti:sapphire amplifier crystal, the pulse energy saturates at about 2.5 mJ. Now, the pulse is ejected from the cavity by another Pockels cell. Starting with nJ pulses, the regenerative amplifier increases the energy per pulse about \( 10^6 \) times which
A.4 The prepulse unit

makes the regenerative amplification the most efficient scheme to transfer energy from a ns laser pulse into a fs-pulse [Diels & Rudolph, 1996].

The next amplification stage is a four pass. After the laser beam diameter is expanded to about 5 mm, the pulse is sent 4 times through a 2 cm thick Ti:sapphire crystal pumped by two frequency-doubled Nd:YAG lasers ('Powerlite 8010' from Continuum), each providing $\sim 700$ mJ in about 10 ns. The laser pulse is amplified to a maximum of 400 mJ. Depending on the application the pulse can either be sent directly to the compressor or to the next amplification stage.

The last stage is only used for ultra-high intensity experiments, like for example for the fission and fusion experiments described in Chap. 6. The pulse is only passing twice through a Ti:sapphire crystal pumped by two beams with each having 2.5 J, again from a frequency-doubled Nd:YAG laser ('Macholite' from Continuum).

After passing all amplification stages, the pulse has an energy of up to 1.3 J, resulting in 900 mJ in 80 fs after the compressor. Even though the laser beam diameter is increased to about 45 mm (FWHM) before the compressor unit, the intensity of the fully compressed pulse is exceeding 200 GW cm$^{-2}$. This enormous intensity causes the laser pulse to interact with air. The laser beam freely propagating in air tends to focus itself, by inducing an intensity-dependent index of refraction. The beam breaks up into several small filaments, destroying mirrors in the beam path. However, being undesired in the laboratory, this effect can be extremely favorable for investigating the earth’s atmosphere via white light LIDAR [Wöste et al., 1997]. To avoid self-focusing and other interactions with air, the laser beam has to be guided in a vacuum beam line. Starting from the first compressor grating, the beam transport to the experiment and finally the entire experiment has to be in vacuum.

A.4 The prepulse unit

A special feature of the Jena Terawatt laser system is the prepulse unit, described in detail in Ref. [Ziener et al., 2000]. Limitation and minimization of prepulses, naturally occurring in the amplification process, is a major issue for all lasers of the Terawatt class and beyond. Because the interaction of the laser with the target becomes less and less defined, the more uncontrolled prepulses are present. However, the precisely controlled introduction of a prepulse enables additional degrees of freedom to influence the laser-plasma interaction dramatically (see Sec. 4.3.4).

The prepulse unit is located between the regenerative and the 4-pass amplifier. The basic setup is equivalent to a Mach-Zehnder interferometer. The incoming pulse is split by a 8:2 beam splitter into the main pulse and the prepulse. The main pulse passes along a fixed path whereas the prepulse

$^1$The critical intensity for self-focusing in air is only 47 MW cm$^{-2}$ [Menzel, 2001]
A.5. The characterization of laser pulses ranging from fs to ns

arm can be varied in length by a motorized translational stage. At the end of the prepulse unit, both beams, the main and the prepulse, are united again by a 9:1 beam splitter. The variable delay in the prepulse arm is constructed in a way that the prepulse has a shorter path than the main pulse, therefore leaving the prepulse unit prior to the main pulse. Limited by the length of the motorized translational stage, the delay between prepulse and main pulse can be varied from zero delay to 700 ps. Additional fixed delays up to 11 ns were introduced by delaying the main pulse with a 2 m long delay stage. In this work, a positive delay represents the case in which the prepulse impinges prior to the main pulse onto the target.

For experiments focusing the laser beam down to 3 µm focal spots, the angular overlap of both pulses has to be very well aligned. A computer controlled motorized mirror within the prepulse arm assured an angular deviation of < 15 µrad for any delay. In addition, a rotatable λ/2 plate was included in the prepulse as well as in the main pulse arm. In combination with a polarizer, the λ/2 plates act as variable attenuator. Main pulse and prepulse energy can be adjusted independently. This feature was valuable for the EUV measurements where different laser pulse energies had to be used while all other parameters (especially the focusing) had to be kept constant. The straightforward procedure to lower the laser pulse energy is to decrease the energy of the pump laser. However, a lower pump fluence will change the temperature distribution in the amplification crystal (in the four-pass amplifier) which leads to a different focal length of the thermal lens, since the temperature gradient in the crystal acts as a lens. This lens changes the divergence of the laser beam and subsequently the focus position will move. This undesirable effect was observed but could be circumvented by attenuating the laser beam with the λ/2 plate without changing the pumping power. The energy of the pulses could be varied leaving the laser focus position fixed.

The ratio of the unattenuated main pulse and the prepulse energy was measured by an autocorrelator (see next section) to be 1.8%. The fraction of the laser energy contained by the prepulse can be increased by attenuating the main pulse with the λ/2 plate. Starting with a 110 mJ main pulse and hence a 2 mJ prepulse, the main pulse can be attenuated to 20 mJ, but the prepulse is still 2 mJ or now 10% of the main pulse energy. Using this scheme, the prepulse energy was kept constant for varying the main pulses for the EUV measurements in Sect. 4.3.4.

A.5 The characterization of laser pulses ranging from fs to ns

One of the main challenges of the EUV project (see Chap. 4) was the measurement of the EUV conversion efficiency as function of the laser pulse duration. Laser pulses ranging from femtoseconds to nanoseconds had to be produced without changing any other parameter influencing the laser-plasma interaction. The laser pulse energy, the beam profile and the focusing as well as the laser wavelength had to remain unchanged.
A.5. The characterization of laser pulses ranging from fs to ns

A.5.1 80 fs to 10 ps pulses

Measuring pulse durations in the picosecond or even subpicosecond regime directly with electronic devices is not possible. Their response time is limited to several tens of picoseconds. However, Fig.A.1 shows that it is possible to measure fs pulse durations. The basic principle of an autocorrelator is that the measurement of the temporal duration is transformed to the measurement of a spatial path length. Therefore, the problem of measuring femtoseconds is mapped to the measurement of distances in the \( \mu \text{m} \) regime. A detailed description of the endless variations of fs-pulse length measurements can be found in Ref. [Diels & Rudolph, 1996].

The correlator in the Jena TW laser system is a third order autocorrelator that can distinguish between prepulses and ‘post-pulses’. The dynamic range of the correlator is \( 10^{-4} \) within a scanning range of 300 ps prior the main pulse. Only two pulses, 1.2 ps and 600 fs prior the main pulse, resulting from spectral modulations in the Pockels cell of the regenerative amplifier, with a contrast ratio of \( \sim 10^{-2} \), were detected. However, these prepulses occur on such a short time-scale that they cannot influence the EUV measurements significantly. No other prepulse was found with a contrast ratio higher than \( 10^{-4} \) within that range.

The amplified and compressed 80 fs laser pulse is essentially a thin slab of light with a diameter of about 5 cm, but a thickness of only 25 \( \mu \text{m} \). If this slab is slightly tilted with respect to the direction of propagation, the pulse duration in the focus will be elongated. The tilt of the pulse front arises from slight misalignment of the grating compressor. In order to correct the tilt, a device capable of measuring the pulse front had to be built. A modified Mach-Zehnder interferometer, containing an unequal amount of mirrors in both arms was constructed in close analogy to the device described in Ref. [Pretzler et al., 2000]. Using this pulse front sensor the gratings could be fine tuned to assure that the tilt of the pulse front elongates the pulse duration in the focus less than 15%.
A.5. The characterization of laser pulses ranging from fs to ns

The pulse length can be elongated by detuning the grating separation, as was already discussed in Sect. A.2. The separation can be changed \( \pm 10 \text{ mm} \) by translational stages below the gratings. This allows the variation of the pulse length between 80 fs and 2 ps (Fig. A.1 and A.2). To achieve pulse durations around 10 ps, the grating separation had to be changed by 50 mm, calculated via Eq. (A.7). Again, in this new position, the translational stages below the gratings were used to achieve pulse durations of 10 ± 2 ps.

The EUV experiments (see Chap. 4) took place in a vacuum chamber, separated from the main vacuum system to avoid the contamination of the main laser system with water. Two quartz (SQ1) windows were placed within the laser path. The linear interaction (group velocity dispersion) broadens a 80 fs short laser pulse only to some percent, whereas for a laser fluence reaching up to 20 mJ/cm\(^2\), the nonlinear temporal pulse broadening cannot be neglected anymore. Self-phase modulation causes the pulse to be elongated to about 200 fs (calculated with LabII [Feurer et al.]). Therefore, the shortest pulse duration used in all EUV experiments is 200 fs instead of 80 fs.

A.5.2 120 ps pulses

The pulse duration of the stretched pulse was calculated in Sect. A.2 to be 130 ps, knowing the stretcher parameters and the bandwidth of the laser. Still, measuring this pulse duration is a problem. The autocorrelation signal decreases with the third power of the laser pulse intensity and is thus not a useful tool to measure 130 ps pulses. Conventional electronic devices on the other hand are too slow. An elegant way to measure the pulse length of a linearly chirped pulse is spectral interferometry. When a short pulse (directly from the oscillator) is superposed with the long chirped pulse, oscillations occur in the spectrum. By changing the delay between both pulses, the pulse duration of the chirped pulse can be derived very accurately [Ziener, 2001]. The measurement resulted in a pulse duration of the stretched pulse in the Jena TW laser system of 120 ps in good agreement with the calculation.

To perform experiments with the stretched and amplified but not compressed pulse, the compressor had to be bypassed. Before the laser pulse entered the vacuum chamber of the compressor, the laser beam was redirected. Without entering vacuum the beam was sent through the laboratory, directly into the interaction chamber. The intensity of the uncompressed laser beam is much lower than the intensity of the compressed one, therefore nonlinear effects due to the propagation in air were not observed.

A.5.3 6 ns pulses

As pointed out in section A.3, the regenerative amplifier is not just a multi-pass amplifier, but more a seeded resonator. By blocking the seed pulses coming from the oscillator, the regenerative
amplifier acts like a Q-switched oscillator itself, controlled by the switch time of the Pockels cell. The regenerative amplifier provided 6 ns laser pulses which were amplified in the four pass amplifier. The pulse duration could easily be measured with a fast photo diode. The 6 ns pulses were, as well as the 120 ps, sent through air to the vacuum chamber of the experiment.

A.6 Focusing the laser beam

A.6.1 The focus

In analogy to the equation (A.7) describing the Gaussian pulse duration, a similar equation can be derived for the spatial propagation of the laser pulse. For a laser beam with a wavelength $\lambda$, a given 1/e-beam radius $w_F$ and the focal length $f$, the resulting 1/e-focus radius cannot be smaller than $w_0$, the diffraction limited focal radius defined by [Diels & Rudolph, 1996]:

$$w_F = w_0 \sqrt{1 + \left( \frac{f}{\rho_0} \right)^2} \quad \text{with} \quad \rho_0 = \frac{\pi w_F^2}{\lambda}. \quad (A.8)$$

In the case of $f \gg \rho_0$, $w_0$ can be expressed as:

$$w_0 \approx \frac{\lambda f}{\pi w_F} \quad (A.9)$$

The similarity of Eq. (A.7) and Eq. (A.8) is just one example for the close relationship between temporal and spatial transformations of Gaussian shaped pulses [Diels & Rudolph, 1996]. The parameter $\rho_0$ is called the Rayleigh range. Moving one Rayleigh range out of the focal plane the beam radius increases by a factor of $\sqrt{2}$, lowering the intensity by a factor of 2. Therefore, $\rho_0$ gives a constrain on the target design that the positioning of a target (including surface roughness) has to be better than one Rayleigh range. Using the parameters for the Jena TW laser: $\lambda = 795$ nm, $f = 119$ mm (used for all experiments) and a beam diameter $2w_F$ of $\sim 43$ mm, the minimum focus diameter ($2w_0$) is 2.8 $\mu m$ and the Rayleigh range $\rho_0 = 8$ $\mu m$ - close to the dimensions of the used wave length because of the tight focusing.

To focus the laser beam extremely tight, in order to produce intensities $> 10^{19}$Wcm$^{-2}$, one can certainly not use a focusing lens. First of all, spherical lenses always encounter aberrations for large opening angles ($w_F \sim f$) that have to be used to achieve a small focus (Eq. (A.9)). Furthermore, the lens material induces nonlinear interactions in the TW laser beam, leading to temporal and spatial distortions of the beam. One way to avoid these two problems is to focus the beam with a reflecting paraboloidal shaped device. By using reflective optics, the beam does not have to propagate through material. In contrast to a spherical surface, a paraboloidal shaped focusing mirror does not induce aberrations enlarging the diffraction limited focus diameter $w_0$. In
A.6. Focusing the laser beam

Figure A.3: Image of a focal spot recorded for a laser pulse energy of 700 mJ and 80 fs pulse duration, leading to an intensity of $5 \times 10^{19}$ Wcm$^{-2}$.

The described experiments, an off-axis paraboloid was used to focus the laser beam off centered, outside the incoming beam (see Fig. 2.2).

The minimum focal spot size $w_0$ is the theoretical limitation of the focal spot size - hardly achieved by lasers of the TW class. A parameter $M^2$ has been introduced to account for the deviation between the real beam and the theoretical optimum. The bigger $M^2$, the more distortions are present in the beam profile [Menzel, 2001].

$$w_0 M^2 = M^2 w_0$$  \hspace{1cm} \text{(A.10)}

The actually measured focal spots encounter a diameter at FWHM of $\sim 3 \mu m$ (or $\sim 4\mu m$ 1/e-diameter). Slight variations are due to small differences in the quality of different parabola. Therefore, the $M^2$ value is about 1.5, which is rather good for a laser of the TW class. By measuring the beam diameter for different distances from the focal plane, like in Fig. A.4, a Rayleigh range of $\rho_{exp} \sim 20 \mu m$ was found, in good agreement with a $M^2 = 1.5$. Scaling $\rho_0$ according to Eq. (A.10) results in a theoretical $\rho_{0M^2}$ of 17 $\mu m$.

Figure A.4: Images of the laser beam recorded at different distances from the actual focus. The circle denotes a diameter of 20 $\mu m$, equivalent to the size of the droplets, acting as target.

To observe the focal spot, it has to be enlarged. By placing a microscope objective (40 × magnification) slightly behind the focus, a greatly enlarged image of the focus can be produced, which is recorded by a CCD camera $\sim 60$ cm behind the microscope objective (Fig. A.3). By moving the microscope objective back and forth in laser direction, the beam profile before and after the focus can be observed. This gives important information about the size and quality of the
A.6. Focusing the laser beam

Due to the short focal length (f = 4mm) of the microscope objective, the depth of focus of the image produced is much smaller than the Rayleigh length of the laser. This assures that the image of the focus is indeed originating just from a thin slice around the focal plane and the image is not a averaged picture of several Rayleigh ranges.

![Sketch of the setup used to image the ~100 times enlarged focal spot onto a CCD camera.](image)

The CCD camera was placed outside the vacuum chamber, about 60 cm away from the focus, to observe the focal spot during the evacuation and in vacuum. The base plate of the vacuum chamber is 40 mm thick, thus one expects such a plate does not bend while the chamber is evacuated. However, considering that the surrounding air pressure is equivalent to an adult elephant sitting on the base plate of the vacuum chamber, of course even a 40 mm plate is slightly bent altering the position and tilt of the parabola in respect to the laser beam, causing aberrations. Thus, it is indeed of essential importance to adjust the parabola in vacuum to achieve a minimum focal spot size.

There are still two open questions. How can the length-scale of the CCD image of the focal spot be calibrated, and how can the focus of a TW laser, vaporizing everything in the vicinity of the focal region, be measured by placing a microscope objective almost directly in the focal plane? In order to calibrate the magnification of the imaging system, a simple and fast method was used.

![The magnification of the focus imaging system (see Fig. A.3) was calibrated by using a diffraction grating. Only for the reason of clarity, a lens instead of the experimentally used parabola was drawn.](image)

(Fig. A.6). A ‘macroscopic’ grating with a grating period (a) of 10 mm was placed in the laser beam in front of the parabola. The higher diffraction orders, slightly inclined to the 0th order, were focused to different locations. Not just one, but several focal spots appear with a constant spacing. A simple calculation shows that the spacing between the spots (d) is given by:

\[ d = \frac{\lambda f}{a} \]  

(A.11)
A.6. Focusing the laser beam

For the focal length \( f = 119 \text{ mm} \) and the wave length \( \lambda = 795 \text{ nm} \) for Ti:sapphire, the distance \( d \) can be calculated to be \( 9.5 \mu\text{m} \). Knowing the distance between the focal spots, the CCD image can be calibrated.

![Figure A.7: Scheme of the TW-laser beam attenuation.](image)

To image the focal spot onto the CCD camera, the intensity of the laser beam has to be tremendously reduced. Simply turning off the last amplification stages would reduce the intensity sufficiently enough that the microscope objective is not destroyed, but the laser beam shape will change as well. To assure that the recorded focal spot was equivalent to the high-power focus in the experiment, the amplifiers had to be operated at full power. The laser beam was attenuated using only the transmission of a highly reflecting dielectric mirror for the imaging (Fig. A.7). Two dielectric mirrors weakened the laser beam to \( \sim 10^{-5} \) of its original power, sufficient to avoid damage of the microscope objective. It was carefully checked that neither the direction of the beam nor the laser beam profile is changed by the attenuation.

A.6.2 Determination of the intensity

All the quantities described so far, laser pulse energy \( E_L \), laser pulse duration \( \tau_L \) and the 1/e radius \( w \) of the focal spot, have to be combined in order to determine the intensity in the interaction region. In good approximation, the intensity can be given as a radially symmetrical function:

\[
I(r) = \frac{E_L \tau_L}{\pi w^2} e^{-\left(\frac{r}{w}\right)^2}
\]  

(A.12)

For the Jena TW laser system the typical focal spots are indeed quite symmetrical (Fig. A.3) and can be fitted by a Gaussian distribution (Fig. A.8). The slight deviations from the Gaussian shape do not contain more than 15% of the laser energy.

Due to grooves on the focusing parabola which result from the manufacturing process, part of the laser light (\( \sim 15\% \)) is diffracted out of the focal spot. By placing a 50 \( \mu\text{m} \) diameter pinhole in the focal plane only 15% of the incident laser energy were blocked, the remaining 85% were focused in a region with a diameter of 50 \( \mu\text{m} \) around the focus. But this is exactly the region that can be imaged onto the CCD camera. Thus, for a maximum laser energy of \( E_L = 750 \text{ mJ} \) reaching the focal region, \( w = 1.7 \mu\text{m} \) and \( \tau_L = 80 \text{ fs} \) a peak-intensity of \( I(r = 0) = 10^{20} \text{ Wcm}^{-2} \) can be achieved.
A.6. Focusing the laser beam

Figure A.8: The averaged radial intensity distribution of the focus from fig. A.3. All pixels at a certain radius around the focus were averaged to get a direction independent representative intensity distribution for the focal spot. The blue region shows the mean intensity in an FWHM-intensity area around the focus (see text).

according to Eq. (A.12). However, the peak-intensity is only a mathematical construct which is defined for an infinitesimal small area around the center of the focus. Therefore, the intensities given for the experiments are mean intensities (blue region in Fig. A.8). The laser energy ($\sim 50\%$ of $E_L$), which is focused in a macroscopic region $\pi w_{FWHM}^2$ (typically $\sim 8 \mu m^2$), divided by this area and the pulse duration is taken as realistic intensity. This value is about 70% ($1/(2 \ln 2)$) lower than the peak-intensity for a Gaussian profile, but is much more adequate to describe the laser-plasma interaction. The same procedure was used for the pulse duration. The intensity was averaged over the FWHM of the pulse duration instead of taking the peak value.
Zusammenfassung der Arbeit


Für die Realisierung eines solchen Targets musste neben einem System mehrerer Vakuumkammern insbesondere eine Apparatur geplant und aufgebaut werden, die zuverlässigen Wassertröpfchen im Vakuum erzeugt. Diese Apparatur ist neuartig und wurde patentiert [Ziegler et al., 2001]. Erstmals wurde die Konversionseffizienz systematisch für Laserpulsdauern von ultrakurzen 200 fs bis zu 6 ns für jeweils verschiedene Laserpulsesenergien untersucht. DieseMessung wurde sowohl für die erwähnten 20 \micro m Tröpfchen als auch für ein ausgedehntes sauerstoffhaltiges Target (SiO \sub 2) durchgeführt. Es konnte gezeigt werden, dass für massenlimitierte Targets sowohl die Laserpuls-
energie als auch die Laserpulsdauer unabhängig voneinander optimiert werden müssen, um eine maximale Konversionseffizienz zu erreichen, unabhängig von der Intensität. Eine optimale Konversion (0,23\% in 4\pi und 2,5\% Bandbreite) ergab sich für genau eine Laserpulsenegie (\sim 50 mJ) und eine Laserpulsdauer (\sim 120 ps), bei gegebenen Targetabmessungen (20\mu m Durchmesser). Diese Entdeckung steht im Widerspruch zur Abhängigkeit der Konversionseffizienz bei ausgedehnten Targets, die wesentlich von der Intensität der Laserpulse bestimmt wird. Der Unterschied konnte auf die begrenzte Anzahl von Atomen in massenlimitierten Targets und auf die Plasmaexpansion zurückgeführt werden. Um die Ergebnisse nicht nur auf eine Targetgröße einzuschränken, wurden Skalierungsregeln für die Laserparameter aufgestellt und experimentell belegt. Die optimale Laserpulsennergie ist der Anzahl der Teilchen im massenlimitierten Target proportional, und die Laserpulsdauer skaliert mit dem Targetdurchmesser. Mit Hilfe dieser Skalierungsregeln kann ein ideal auf einen gegebenen Laser abgestimmtes Target entwickelt werden.

dingungen durch maßgeschneiderte Laserpulse die Konversion sehr deutlich zu steigern vermag. Es ist denkbar, durch Einsatz komplexer Laserpulsformung [Stobrawa et al., 2001], die die Produktion von mehr als 2 Pulsen (Vor- und Hauptpuls) erlaubt, und der zusätzlichen Anwendung von selbstlernenden Rückkopplungsmechanismen [Feurer, 1999] die Plasmabedingungen noch besser abzustimmen und so die Konversionseffizienz noch weiter zu steigern. Damit wäre die Möglichkeit gegeben, eine laserbasierte EUV-Quelle für lithographische Anwendungen wirtschaftlich einzusetzen.

Weiterhin wurde das, im Rahmen der vorgestellten Arbeit entwickelte Tröpfchentarget für Experimente eingesetzt, die sich mit grundlegenden Fragen der relativistischen Plasmaphysik beschäftigen. Dazu wurde die Laserintensität um mehr als drei Größenordnungen auf $5 \times 10^{19}$ Wcm$^{-2}$ gesteigert. Elektronen, die von solch einem intensiven Laserfeld erfasst werden, werden auf kinetische Energien beschleunigt, die ihre Ruheenergie übertreffen und einige MeV erreichen können. Diese Elektronen beschleunigen ihrerseits Ionen über Raumladungseffekte auf Energien von einigen 100 keV. Um die Charakteristika der schnellen Ionen messen zu können, wurden kernphysikalische Methoden benutzt. Das "normale" Wasser, das für die EUV-Messungen verwendet wurde, wurde durch schweres Wasser (D$_2$O) ersetzt. Bei Ionentemperaturen von 100 keV kommen sich die Deuteriumkerne nahe genug, dass zwei Kerne verschmelzen können und Helium ($^3$He) und ein Fusionsneutron erzeugen. Zunächst ist diese Art der Neutronenproduktion, die die Neutronen in einem sehr kleinen Quellgebiet ($\sim 100 \mu m^3$) für sehr kurze Zeiten ($\ll 1$ ns) und einer relativ monoenergetischen Energieverteilung ($2,4 \pm 0,4$ MeV) erzeugt, eine der brillantesten Neutronenquellen weltweit. Misst man die Energieverteilung der Neutronen, so kann man auf die ursprüngliche Ionenenergieverteilung zurückschließen. Man hat somit ein nicht-invasives Thermometer für Milliardengrad heißes Plasma. Dieser Gedanke wurde zwar schon an ausgedehnten Festkörpertargets angewandt [Hilscher et al., 2001], jedoch nicht an massenlimitierten Targets. Durch die Kürze der Wechselwirkung ($\sim 100$ fs) kann die Expansion vernachlässigt werden, und die Wechselwirkung findet nur in einem ca. $5 \times 5 \times 2 \mu m^3$ kleinem Volumen im Auftreffpunkt des Lasers statt - sozusagen auf der Vorderseite des Tröpfchens. Die Neutronen-Emmissionscharakteristik der Neutronen aus diesem Bereich ähnelt daher, trotz der geringen Tröpfchenabmessungen, der eines ausgedehnten Targets. Das Neue an der durchgeführten Messung ist jedoch, dass mit Hilfe eines "Ionenaffängers", bestehend aus einer Scheibe deuterierten Plastiks, auch Ionen nachgewiesen werden konnten, die auf der Rückseite des Tröpfchens beschleunigt wurden. Durch Vergleich von Simulationsrechnungen mit den gemessenen Neutronenspektren konnten in einem Experiment sowohl die Anzahl der rückseitig sowie der vorderseitig beschleunigten Ionen als auch deren jeweilige Temperatur bestimmt werden. Es zeigt sich, dass ca. 10-20 mal mehr Ionen auf der Rückseite beschleunigt werden als auf der Vorderseite im Laserfokus, die Temperatur der Rückseitenionen jedoch um Faktor 2 niedriger ist als die der Vorderseitenionen. Durch diese Größen lassen sich die Beschleunigungsfelder in den jeweiligen Beschleunigungsregionen bestimmen. Diese sind in guter Übereinstimmung mit theoretischen Modellen.
Danksagung

Ich möchte diese Arbeit nicht beenden ohne all denen zu danken, die einen mehr oder weniger großen Anteil zum Gelingen der Arbeit beigetragen haben und mir in den letzten Jahren viel über Physik und das Leben beigebracht haben. Zunächst gilt mein Dank Prof. R. Sauerbrey, der mich in der Arbeit nicht nur anleitete, sondern mich auch mit seiner Begeisterung über die erzielten Ergebnisse motivierte. Weiterhin danke ich Prof. Dr. O. Willi und PD Dr. P. Gibbon für die Übernahme der Gutachten.


Lebenslauf

Name: Stefan Düsterer
Geburtsdatum: 9.1.1974
Geburtsort: Dinkelsbühl (Bayern)
Statsangehörigkeit: deutsch
Familienstand: ledig
Privatanschrift: Fuchslöcher Str. 12 / 07749 Jena

Sept. 1980 - Sept. 1984: Grundschule Dinkelsbühl
Sept. 1984 - Juni 1993: Mathematisch-naturwissenschaftliches Gymnasium Dinkelsbühl
Juni 1993: Erlangung der allgemeinen Hochschulreife
Okt. 1994 - Juli 1997: Studium der Physik an der
Julius-Maximilians-Universität Würzburg
Aug. 1997 - Aug. 1998: Masterarbeit (‘Second harmonic generation from ferromagnets’) an der University of Texas at Austin
Seit Nov. 1998: Doktorand am Institut für Optik und Quantenelektronik der Friedrich-Schiller Universität Jena

Jena, 28. August 2002
Annotation

Minor modifications, compared to the original version of this thesis, have been implemented to achieve clearance from our industrial partner.

Hamburg, 4/8/2003