

Contrast improvement of intense ultraviolet pulses

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PhD thesis (summary)

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The remarkable progress of laser technology in the last decades allowed breaking scientific results in various fields including physics, medicine, biology and material science. The industrial application of lasers has an increasing importance like in laser based micro/nano machining. The driving force behind many of these applications is the improvement of the peak intensity of laser systems and the extension of the wavelength range of secondary sources. The investigation and improvement of the spatio-temporal quality of ultrashort laser pulses plays an ever increasing role in high intensity physics. Most of the applications of high-intensity laser pulses require extremely high temporal and spatial quality. Temporal prepulses are especially detrimental for high intensity laser-matter interactions, as the lower intensity prepulse may generate a preplasma preventing direct interaction between the main pulse and the material to be studied.

During my research I investigated the contrast improvement of high-brightness ultraviolet short pulses, which are advantageous for numerous interactions, due to the shorter wavelength therefore higher photon energy and

better focusability. Ultraviolet high-intensity lasers can be regarded as complementary sources of the most commonly used infrared ultrashort pulses. In my thesis the construction and the main features of high-intensity lasers systems are reviewed with emphasis on the available contrast improvement techniques. Many techniques for contrast improvement of high-intensity pulses have been developed recently, however, the temporal quality of ultrashort pulses are still not satisfying regarding the available and targeted peak intensities.

High-intensity KrF excimer laser systems can produce 10^{19} W/cm² intensity with $\sim 10^9$ intensity contrast. Plasma mirror technique was successfully demonstrated for UV short pulses reaching $\sim 50\%$ reflectivity and 2 orders of magnitude contrast improvement. Nonlinear Fourier-filtering was also proposed to temporal and spatial filtering of UV short pulses. With this technique 40% energy throughput and 3 orders of magnitude contrast improvement was demonstrated. The motivation behind my scientific research was to develop a high-intensity and high contrast light source in the ultraviolet. For this reason,

I aimed to increase the maximum reflectivity of the plasma mirror and characterize the spatial features of the reflected pulses. I also wanted to identify the limiting factor of the achievable contrast improvement of the nonlinear Fourier-filtering technique and improve the filtering experimental arrangement. Based on these results, my ultimate goal was to integrate the nonlinear Fourier-filtering into a short pulse KrF laser system. My scientific results are summarized in the following 4 sections.

1. It has been experimentally demonstrated that 70% reflectivity of plasma generated by ultraviolet short pulses can be reached on quartz target, which is the highest published value at 248 nm by this time. Based on this plasma mirror effect an experimental arrangement has been developed for high-intensity KrF laser systems, offering contrast improvement of more than two orders of magnitude. It has been demonstrated that the focusability of the filtered beam is unchanged, moreover the reflectivity is practically independent of the polarization of the beam and of the

**pulse duration (measured for 500 fs and 220 fs pulses)
[1].**

The reflection of the plasma was measured on an anti-reflection coated quartz target with f/30 focusing. The reflectivity approached 70% for 12° of angle of incidence both for s and p polarized beam at the $1 \cdot 10^{15}$ - $5 \cdot 10^{15}$ W/cm² intensity range, when the pulse width was 500 fs. The plasma mirror effect was also demonstrated for shorter (220 fs long) pulses, where the maximum reflectivity was somewhat moderate ~55%. This is likely connected to the less optimum temporal and spatial quality of the beam. The focal distribution of the beam was measured before and after reflection; the diameter of the focal spot was 1.5 and 1.75 times of the diffraction-limited size, respectively. Regarding the achievable contrast improvement of > 2 orders of magnitude - defined by the ratio of the high and low intensity reflection - and the good spatial distribution of the reflected beam this approach can be regarded as an efficient filtering technique for high-intensity KrF laser systems.

2. A theoretical model has been developed to investigate the achievable contrast improvement of the nonlinear Fourier-filter. The numerical simulation showed that the limitation is imposed by the spatial contrast of optical imaging; restraining the maximum contrast in our experimental case to 10^3 . It was suggested that an object is better suited to high contrast imaging if the spatial frequency spectrum of the object is modulated by a low numerical aperture pre-imaging and this image is completed by a second aperture. It was shown by numerical calculations that such an object can further be imaged with higher than 10^8 spatial contrast [2].

The spatial contrast of optical imaging was studied by the use of frequency analysis of linear systems. This simulation can handle spatially coherent and incoherent illumination. For an imaging system of 500 cm^{-1} cut-off frequency the simulation showed good agreement with the experimentally obtained value for the spatial contrast of imaging. It was shown that with increasing solid angle (or resolution) of imaging the spatial contrast increases.

However, the eventually increasing phase front error of aberrations decreases the spatial contrast. It was demonstrated that the apodization of the exit pupil of the imaging system can improve the available spatial contrast by several orders of magnitude. For practical reasons another approach - based on the controlled modulation of the spatial frequency spectrum by decreasing the role of the high spatial frequency components - is more preferable. We realized this by applying a low numerical aperture imaging and a secondary beam-block in the image plane of this pre-imaging to exclude the areas of normalized intensity below 10^{-3} . An object of such spatial frequency distribution can be imaged with a spatial contrast greater than 10^8 .

3. It was experimentally demonstrated that the imaging system of the nonlinear Fourier-filter operating the visible wavelength range can be improved by the spatial frequency modulation of the object, resulting in a spatial contrast as high as 10^7 . The improvement of the contrast of imaging was also experimentally demonstrated for ultraviolet short

pulses by measuring the intensity dependent throughput of the nonlinear Fourier-filter. Using an object of controlled spatial frequencies the small signal throughput decreased from 10^{-3} to 10^{-5} - determined by the limited dynamic range of the measurement. Based on this results an experimental arrangement is suggested to improve the intensity contrast of high-intensity UV pulses by more than 5 orders of magnitude. The spatial features of the filtered pulses are also characterized [2].

The new object of modulated spatial frequency was created by a combination of a low numerical aperture pre-imaging and a secondary beam-block in the image plane which excluded the areas of normalized intensity below 10^{-3} of the intermediate image. This modulated object could be imaged with a spatial contrast of 10^7 . This corresponds to 5 orders of magnitude improvement in the visible wavelength range. The achievable contrast improvement of the nonlinear Fourier-filter was determined by measuring the intensity dependent throughput. By the use of the frequency modulated object

the low intensity transmission decreased below 10^{-5} . This value was limited by the dynamic range of the measurement; pulses of focused intensity below $\sim 4 \cdot 10^{12}$ W/cm² could not be detected. As far as the spatial features of the filtered beam are concerned, the far field distribution of the beam has a regular Gaussian shape with 1.75 times larger spot diameter than the diffraction-limit.

4. A special light source has been developed for the investigation of laser-matter interaction at high-intensity and high temporal contrast. An UV short-pulse amplifier chain - containing three KrF excimer amplifiers - has been constructed where the nonlinear Fourier-filtering technique is integrated into the system after the first preamplifier. Characterization of the output pulses showed that with sufficiently low F-number focusing 10^{19} W/cm² intensity can be achieved with 10^{12} intensity contrast in the focal plane [3].

The short pulses - generated by the sub-picosecond dye laser system - after frequency conversion are amplified in a KrF amplifier chain consisting of three gain

modules of increasing cross-section, up to an output beam size of $\sim 4 \times 4 \text{ cm}^2$. The two preamplifiers are used in a double pass arrangement and the nonlinear Fourier-filter is integrated between them. To promote effective pulse filtering the pre-imaging is done during the second amplification pass of the first preamplifier. The high contrast pulses of sub-mJ energy are then amplified in the second preamplifier and in the final amplifier - which uses a two beam interferometric multiplexing setup - up to 100 mJ. The temporal characterization of the pulses were based on second order autocorrelator and spectral intensity measurement. Both the near and far field spatial intensity distribution of the beam was also recorded. The expected focused intensity - with $f/2$ focusing - of the 2 times diffraction limited and 700 fs long pulses is in excess of 10^{19} W/cm^2 . Based on our experiments the temporal background of the output is determined by the amplified spontaneous emission generated solely after the nonlinear Fourier-filter. The energy of the ASE strongly depends on the gain of the amplifier chain following the filter. For different values of this gain an intensity contrast between 10^{11} and 10^{12} is obtained in the focal plane.

My scientific results were published in the following papers:

[1] B. Gilicze, A. Barna, Z. Kovács, S. Szatmári, I. B. Földes, “*Plasma mirrors for short pulse KrF lasers*,” Rev. Sci. Instrum. 87, 8, 083101 (2016).

[2] B. Gilicze, R. Dajka, I. B. Földes, S. Szatmári, “*Improvement of the temporal and spatial contrast of the nonlinear Fourier-filter*,” Opt. Express 25, 17, 20791 (2017).

[3] B. Gilicze, Z. Homik, S. Szatmári, “*High-contrast, high-brightness ultraviolet laser system*,” Opt. Express 27, 12, 17377 (2019).