Development and optimization of high intensity solid state - excimer hybrid laser systems and their application for short pulse material processing

PhD thesis

by

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Szeged

2003

I. Scientific background, aims

Thanks to the advantages of the well-known chirped pulse amplification (*CPA*) technology, solid state laser systems operating in the infrared (*IR*) region are widely used as compact high brightness sources, and in fact dominate this field. Only moderate efforts have been made to develop compact and reliable short-pulse laser systems operating in the ultraviolet (*UV*) part of the spectrum. In numerous applications, however, high intensity fs pulses in the UV wavelength region have ultimate advantages compared to longer wavelengths. In most of the high intensity experiments, it is the minimal achievable focal spot size, which is considered to be the major figure of merit for the performance of the laser system. This is where the better focusability of short wavelength gas lasers plays a very important role. The short wavelength results in an around 3 times smaller theoretically achievable minimal focal spot size, compared to that of solid-state systems operating around 800 nm. The gaseous active medium of excimers also introduces less optical distortions. As a result of the combination of these effects, a typically 10-100 times smaller focal spot area is reachable for excimer lasers compared to high intensity solid-state systems.

Since there is no effective way to generate ultrashort pulses in the UV, it is necessary to amplify frequency-converted beams, most effectively in specially designed excimer modules. Different front ends can be used, e.g. frequency tripled seed beams from a Ti:Sa solid-state laser or frequency doubled pulses of a short pulse dye laser system. In earlier experiments the dye laser front-end was used and successfully applied in numerous laboratories all over the world. However there are some features (reliability, simplicity, possibility of high frequency operation), which make the solid-state front-ends more preferable. Because of their enormous development in the last few years compact, reliable, easy-to-handle systems with high average powers and repetition rates are available. The above-mentioned developments opened the way for the development of solid state - excimer laser systems. This line was followed in this work combined with the study of possible applications.

A very important application field of such systems is material processing, especially if high precision is required. The most important advantages of such a system for submicron machining are the small penetration depth of the radiation, the small thermal diffusion length and the high optical resolution. The high peak power ensures a small penetration depth through multiphoton absorption and consequently well defined ablation depths. Additionally the short pulse duration provides a small thermal diffusion length and through that prohibits the lateral spreading of heat into the surrounding regions, ensuring the creation of small lateral structures. Operation with UV pulses provides the necessary high spatial resolution because the minimum achievable irradiated spot size scales with the wavelength. Further advantage of the short wavelength is the increased absorption of most of the materials in the UV region. This relaxes further the system requirements even for hardly machinable, in the IR and VIS wavelength region transparent materials. As a result of all these advantages practically all kind of materials can be machined with submicron precision, applying UV femtosecond pulses.

My PhD work focuses on the development of reliable and compact high power femtosecond UV laser systems, optimizing the operational conditions and optical setups. My aim was to improve the output parameters like pulse energy, repetition rate or output power and to adapt the system to different applications. Among the investigated applications one of the most important field was micromaterial processing. In this part of my work I focused on the development of time effective material processing techniques, allowing the fast machining of large surfaces, also in industrial environment.

II. Investigation methods and tools

In the experiments a big variety of investigation methods and tools have been used. For the development of high intensity solid-state-excimer laser systems a solid state Ti:Sa laser source has been used (*Spectra Physic Inc., USA*) to generate seed pulses at 745 nm wavelength. For the frequency upconversion a frequency-tripling unit was built in a linear arrangement based on BBO crystals (*CASIX*, *China*) as nonlinear medium. For the amplification of these pulses at 248 nm different excimer modules (*Lambda Physic GmbH, Germany*) were modified and optimized for the different experimental conditions. The investigated optical setups are based on the principles of the off-axis amplification and interferometric multiplexing schemes.

To measure the pulse energies of the amplified and seed pulses at 248 nm and to measure the pulse energies at 745 nm right before frequency tripling Gentec ED100, ED200, ED500 and RJP735 type piezoelectric energy meters were used. To measure the output power of the queasy cw Tsunami master oscillator Spectra Physics power meter (*model 407a*) was applied. To characterize the output pulse length at 745 nm a Spectra Physic single shot autocorrelator, at 248 nm a multiple shot UV autocorrelator based on multiphoton ionization of NO was used. For the spectral measurements a homemade grating spectrometer in Littrow condition was applied and the spectrum was visualized with a diode array (Hamamatsu) as a detection unit. To control the timing between the different laser units active delay control boxes from Lambda Physik (*LP*) were used. To adjust the optimal delay between the different laser components fast photodiodes (*Hamamatsu R1193U, Alphalas UPD-200-UD*) in combination with a 1 GHz bandwidth analog oscilloscope from Tektronix (*Tektronix 7104*) were used. For the measurements of the beam profile of the amplified UV beams a homemade image processing system was used based on a modified UV sensitive CCD camera and a beam analyzing system (*MrBeam, LLG, Germany*).

In the material processing measurements alternative laser sources as ns mode excimer lasers operating at 157 nm, 193 nm, 248 nm and 308 nm, AR^+ ion lasers at 488 nm, Nd-YAG lasers at 1064 nm and 532 nm were used together with the short pulse UV and IR systems operating at 248 and 745 nm, respectively. Different imaging techniques were tested and compared based on achromatic UV objectives (*Spindler&Hoyer, DUV Retro*), Schwarzschild-type reflective objectives (*Ealing 25x, 36x*) in combination with simple amplitude masks or specially designed and generated diffractive phase elements. For the precise positioning of the samples xyz-translators from PI (*Physik Instruments, M-510.11*) were used in combination with home made online monitoring systems. The morphology of the irradiated samples was investigated with light microscopes (*Zeiss Axioskop*) and with a scanning electron microscope (*Zeiss, DSM 962*), the depth of the ablated structures was measured by a profilometer (*Dektak 3030 Auto II Stylus*) and by atomic force microscopy (*AFM*).

III. Results

The aims described in the 1st point were investigated with the help of methods and tools detailed in the 2nd point, and the new experimental results of these investigations are listed in the following:

1. A new solid state – excimer hybrid laser system has been developed, amplifying the frequency tripled output of a solid state Ti:Sa system set to 745 nm delivering up to over 50 mJ energy at 248 nm in the UV wavelength region. For the amplification a specially designed wide-aperture KrF excimer module was used in a three-pass off-axis arrangement [4].

In the experiments a solid state Ti:Sa system was used to generate the short optical pulses in the infrared (*IR*) wavelength region. The applied solid-state system (*from Spectra Physics*) is based on a diode-laser (*Millenia*) pumped queasy cw Ti:Sa oscillator (*Tsunami*) set to 745 nm, and a Nd-Ylf laser (*Merlin*) pumped regenerative amplifier (*Spitfire*). The system was capable to deliver ~ 150 fs IR pulses with ~ 400 μJ output energy up to 1000 Hz. The required 248 nm wavelength range was reached by frequency tripling. For the frequency upconversion a tripling unit was built in a linear arrangement applying BBO crystals as nonlinear medium, delivering 30-40 μJ energy fs pulses at 248 nm. Using this arrangement ~8% conversion efficiency has been achieved providing an absolutely hands off, “no tweak” operation of the tripling box. Then the UV pulses were sent through the KrF amplifier, which was specially modified for short pulse amplification. After the necessary electrical and mechanical modifications of the KrF module its capability for short pulse amplification was characterized by measuring the momentarily stored energy of the device. It has been found and experimentally demonstrated that applying a carefully optimized 3-pass off-axis arrangement it is possible to reach output energy of 50 mJ of the UV femtosecond pulses.

2. By applying a 2-beam variant of the polarization-multiplexing scheme in combination with the same excimer module used in point 1, amplification of UV femtosecond pulses up to the 100 mJ region has been realized. Seed pulses with 2-3 mJ energy were used in order to reach optimized operation [4].

Experimental investigations showed that using the Sagnac interferometer’s principle to build a polarization multiplexing arrangement it is possible to extract 100 mJ energy out of a single KrF amplifier. The applied scheme was a 2-beam variant; each of them using 2 off-axis passes. Numerical calculations and the experimental results showed that proper recombination of the partial beams is possible with interferometric precision, resulting in a single output beam without the presence of any pre-pulse having a divergence ~2 times above the diffraction limit. To saturate the amplifier it was necessary to use input beams with pulse energies in the mJ region. Because of the lack of an additional power amplifier stage for the Ti:Sa system - allowing to generate some mJ energy frequency tripled pulses – a Szatmári-type dye laser - excimer system, based on a distributed feedback dye laser (DFDL), was used in these experiments to seed the excimer module. To clean the input pulses from the unwanted background radiation (ASE) a 3-meter long spatial filter for high power beams has been built based on home made high damage threshold CaF_2 pinholes. After recombination of the two partial beams at the output, 100 mJ energy subpicosecond UV pulses has been detected at the output of the excimer module used and described in the 1st point. The key component of the arrangement was the polarization sensitive reflector (PSR). By constructing the PSR three very important features had to be considered. The stability of the substrate material defined by the thickness, the multiphoton absorption of the substrate defined by the thickness and type of the material and the possible appearance of pre-pulses influenced by e.g. the positioning of the PSR. By investigating these possibilities a thick PSR on CaF_2 substrate has been developed to provide low loss operation without the appearance of - in many experiment unacceptable – pre-pulses.

3. Based on a high repetition rate solid-state front-end a Ti:Sa - excimer system has been developed operating up to over 300 Hz repetition rate, delivering 30 mJ output energy UV pulses. This way an average power of 9 W at 300 Hz repetition rate has been reached in the UV wavelength region [7].

In numerous applications it is not the high output energy but the high average power which is the most important parameter of the given laser system. It is usually the case in applications where big number of shots are required or large surfaces are necessary to machine. The systems described in the 1st and 2nd points were able to operate at some 10 Hz repetition rate, and so the delivered average power was rather low. Using a Ti:Sa system – running up to 1000 Hz repetition rate – to deliver the seed pulses for a modified excimer module capable to operate up to 300-350 Hz (*NovaLine100, Lambda Physik*), we developed a system running up to over 300 Hz repetition rate, delivering 9 W average power at 248 nm. After the necessary modifications for short pulse amplification, different electrical and operational parameters - like capacitance of the peaking capacitors, gas pressure etc. – has been optimized by measuring the momentarily stored energy of the module. It has been found that maximum stored energy can be measured at 3.3 bar pressure with 28.5 nF peaking capacitance. At these operational conditions 30 mJ output pulses has been measured at 300 Hz, using a carefully optimized 3-pass off-axis arrangement, similar to that described in the 1st point.

Making use of the unique possibilities of the above described laser systems various results in different application fields like the generation of efficient XUV pulses [5], investigation of short pulse ablation properties of metals, semiconductors [1] and nanoparticle gold films [2], metal deposition on silicon and polymer surfaces [13,14], application of diffractive phase elements at 248 nm [8], generation of gratings with grating periods down to 300 nm [1], and drilling of submicron-size holes on practically all kind of solids [1,3,5,6,8] etc. can be presented. The most important results are concluded in the following points.

4. By investigating the ablation properties of materials with high heat conductivity in the picosecond and subpicosecond region. Morphology changes as a function of the pulse duration have been studied, and the result show that the quality of the ablated structures strongly depends on the pulse duration. For metals and semiconductors optimal pulse duration under 5 ps was found to be necessary in order to eliminate undesired melting effects. Subpicosecond ablation properties (like thresholds, rates and morphology) of novel nanoparticle layers has also been investigated and compared with that of conventionally evaporated films. For nanoparticle films five times higher ablation rates have been measured. The ablation thresholds and rates were also found to be dependent on the particle size and evaporation pressure [2,4]

In the experiments a short pulse KrF laser system has been used delivering 10 mJ pulses at 248 nm with pulse durations varying between 500 fs and 50 ps. In the ablation experiments materials with high heat conduction has been investigated. In order to be able to follow the morphology changes sensitively, submicron size structures has been ablated in the samples. After irradiating the probes a melt zone has been observed with a depth defined by the pulse duration, the electron-phonon relaxation time and the thermal diffusivity. It is well-known from theoretical considerations, that the size of the heat affected zone can be minimized, if $L_{th} = \sqrt{2\kappa\tau} \leq \alpha^{-1}$, where L_{th} is the thermal diffusion length, κ is the thermal diffusivity of the material, τ is the laser pulse duration and α^{-1} is the penetration depth of the radiation. In the case of metals and semiconductors with high heat conductivity this condition is only fulfilled for pulses shorter than a couple of picoseconds. By recoil of the molten and evaporated material formation of rims and droplets were observed. The morphology of these structures also depends on the pulse duration. For short pulses the rapid resolidification causes sharp burrs and small droplets, and broad, smooth rims for longer ones. Both for metals and semiconductors the quality of the resulting structures deteriorates at pulse durations exceeding 5 ps.

Subpicosecond ablation properties of thick gold films of ultra-fine particles has been studied and compared of that of conventionally evaporated gold layers, applying 500 fs UV pulses of a 50 mJ solid state - excimer laser system. Ablation thresholds, ablation rates at different fluences, and morphology of the ablated structures has been investigated. While the ablation thresholds and the details of the ablated structures are similar, for nanoparticle films an ablation rate five times higher was found. The ablation thresholds and rates are supposed to be also dependent on the particle size of nanocrystals and on the evaporation pressure.

5. Micromachining of different type of metals and semiconductors has been investigated. It has been demonstrated experimentally that even in the case of materials with high thermal diffusion length (like metals and semiconductors) it is possible to create submicron period gratings or sub-μ diameter holes on the surfaces of different materials applying a mask projection setup based on a Schwarzschild-type reflective objective. Using high fluences and some thousands of shots it was possible to perforate 5 μm thick stainless steel foils with 600 nm diameter holes [3,5,6].

To demonstrate the possibilities of short UV pulses for micromaterial processing individual holes of submicron diameter on stainless steel sample surfaces has been investigated. As it is described in the 4th point, creation of submicron structures on metal surfaces is only possible by applying shorter than a few ps pulses, caused by melting effects. Using a mask projection setup and femtosecond laser pulses in combination with high numerical aperture reflective optics (Schwarzschild-objective, maximal optical resolution at 248 nm is ~ 360 nm), 500 nm diameter holes were generated on the surface of steel samples. It has also been shown that perforation of steel foils is possible preserving a sub-μm diameter, applying high number of pulses up to a material thickness of around 5 μm. Perforation of foils above this limit – presumably because of the high numerical aperture and small confocal parameter of the projection setup – is possible only by applying complicated pre-drilling processes. By applying a high precision mechanical pre-drilling process, generation of submicron diameter holes (output diameter) in 1 mm thick steel plates has been demonstrated

6. It has been demonstrated experimentally that using a high power femtosecond laser source, the experimental conditions can be optimized for various applications like: a)

generation of submicron size periodic surface modulation like normal grating or sinusoidal crossed grating structures, b) simultaneous drilling of large number of holes with diameters down to 300 nm using a special interferometric technique or c) generation of micron size hole matrices on different metal surfaces applying specially designed diffractive phase elements (DPE). Applying different experimental conditions and optimized optical arrangements for the application, significant increase in efficiency can be reached [1,6,8].

For efficient texturing of large surfaces different optical methods were developed and experimentally tested. In a relatively simple arrangement, by imaging an amplitude grating with a high numerical aperture Schwarzschild objective, submicron size grating structures can be generated even on highly conducting metal or semiconductor surfaces, dielectric or waveguide materials. Using two amplitude grating mask in a crossed position crossed grating structures can be created. Combined this technique with a Fourier filtering, hole matrices on metal surfaces with individual hole diameters down to 300 nm were generated on different metal surfaces, applying only a few numbers of shots for the generation of several thousand holes. For the creation of surface patterns with small filling factor a second method was tested, based on diffractive phase elements (DPE's). Two level DPE´s were used to distribute the laser light only in the desired directions minimizing the energy losses significantly increasing the efficiency of the machining process. Applying this technique 10×10 hole matrices with spot diameters in the μm region were fabricated.

7. **For the fabrication of the diffractive phase elements a one-step method was developed based on ns excimer laser ablation using ArF operating at 193 nm. I also developed and successfully tested a new method to decrease the stringent restrictions for the pixel-depth of the DPE. Immersion liquid has been used to set the refractive index difference of the mask substrate and the ambience and to reach optimized operation of the DPE for the given surface relief step height [8].**

Design and fabrication of DPE's has been investigated. Using an Iterative Fourier Transform Algorithm (IFTA) for the design and a simple one-step ablation process for the fabrication it was possible to create diffractive phase masks on quartz plates. The fabrication of quartz is usually quite complicated, especially if shallow holes with a few nm depth precision and very smooth bottom are required. For efficient operation of the phase mask at 248 nm (on fused silica substrate in air), the surface relief step height has to be

~244 nm. In order to relax the stringent restrictions for the hole depth a new technique has been developed and successfully applied using immersion liquid. Changing the refractive index difference of the substrate material and the immersion liquid can control this way the pixel depth. This technique was successfully tested on the phase elements fabricated with ArF excimer laser pulses at 193 nm. Applying distilled water immersion 900 nm step size is required for the efficient operation, this height was reached applying 2 laser pulses of ~19 J/cm^2 fluence for each pixel.

IV. Outlook, possible applications

The above listed scientific achievements contribute to a better understanding of the physical properties of KrF amplifiers. The combination of the advantages of solid state lasers with the ones of UV excimer power amplifiers open new perspectives by providing a simple tool for the investigation of a wide range of physical phenomena. The developed new sources and imaging techniques can open new ways in material processing of practically all kind of materials even for mass production in industrial environment. Alternatively, such a system opens the way for the investigation of high intensity processes even in small laboratories, using a reliable, easy-to-handle small-scale table-top solid state-excimer laser system.

V. List of publications

List of publications supporting the thesis points

1. P. Simon, J. Ihlemann, J-H Klein-Wiele, and **J. Békési**: *„Ablation of solid targets with UV femtosecond pulses”,* SPIE **3822**, 118-124 (1999).
2. **J. Békési**, R. Vajtai, P. Simon, and L. B. Kiss: *„Subpicosecond excimer laser ablation of thick gold films of ultra-fine particles generated by a gas deposition technique”,* Appl. Phys. A **69** [Suppl.], 385-387 (1999).
3. **J. Békési**, P. Simon, and J. Ihlemann: *„Femtosecond UV-laser processing of sub-micron holes in steel foils”, 2000* IEEE, Conference Digest 390 (2000).
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5. P. Simon, **J. Békési**, C. Dölle, J.-H. Klein-Wiele, and S. Szatmári: *„Ultraviolet femtosecond pulses: Key technology for sub-micron machining and efficient XUV pulse generation”,* Appl. Phys. B **74** [suppl.], 189-192 (2002).
6. **J. Békési**, J.-H. Klein-Wiele, and P. Simon: *„Efficient submicron processing of metals with femtosecond UV pulses”,* Appl. Phys A **76**, 355-357 (2002)
7. **J. Békési**, S. Szatmári, P. Simon, and G. Marowsky: *„Table-top KrF amplifier delivering 270 fs output pulses with over 9 W average power at 300 Hz”,* Appl. Phys. B **75**, 521-524 (2002).
8. **J. Békési**, J.-H. Klein-Wiele, D. Schäfer, J. Ihlemann, and P. Simon: *„Surface texturing of metals with sub-micron precision using a short pulse UV laser”,* SPIE 4830, 497-500 (2003) (Student Award of the Conference)

Other publications

9. **J. Békési**, K. Kordás, K. Bali, R. Vajtai, Cs. Beleznai, and L. Nánai: *„UV-laser-induced etching and metal seeding on polymers; a surface characterization”,* App. Surf. Sci. **138-139**, 613-616 (1999).

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12. K. Kordás, **J. Békési**, R. Vajtai, L. Nánai, S. Leppävuori, A. Uusimäki, K. Bali, Thomas F. George, and G. Galbács: „*Laser-assisted metal deposition from liquid-phase precursors on polymers”*, Appl. Surf. Sci. **172**, 178-189 (2001).
13. K. Kordás, **J. Békési**, R. Vajtai, M. Jauhianen, J. Remes, A. Uusimäki, S. Leppävuori, T. F. George, and L. Nanai: „*Laser-assisted via hole metallization in PCB materials”*, J. Electron. Mater. **30**, 21-24 (2001).
14. K. Kordás, S. Leppävouri, **J. Békési**, L. Nánai, J. Remes, R. Vajtai, and S. Szatmári: „*Nickel deposition on porous silicon utilizing lasers”*, Appl. Surf. Sci. **186**, 232-236 (2002).
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