UNIVERSITY OF PÉCS

Doctoral School of Physics

Laser physics, nonlinear optics and spectroscopy program

Interaction of ultrashort laser pulses with dielectric media

Summary of the PH.D. thesis

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Background to the research

The most prominent result of the enormous progress in pulsed laser technology over the past 60 years has been the production of ever shorter laser pulses with ever higher pulse energy. These parameters have improved by orders of magnitude compared to the characteristics of the first lasers. Q-switched lasers, which initially produced nanosecond pulses, were quickly overtaken by mode-locked picosecond and then femtosecond lasers, complemented by increasingly sophisticated optical amplifiers. In science, such a rapid progress leads to the emergence of new areas of research and development.

With the decrease in laser pulse duration, new fields of research have emerged, from attosecond physics to medical applications and laser micro- and nanostructuring. Solid-state lasers and laser amplifiers capable of generating pulses between 5 fs and 50 fs (building on the excellent laser physics properties of the titanium-doped sapphire crystal) appeared in the early 1990s, paving the way toward the study of the interaction of laser light with matter at higher intensities than ever before and its applications [1] [2].

Research on the laser induced damage threshold of optical components

One of the emerging areas of research is due to the increase in the energy of laser pulses. A key role in the future development of solid-state laser technology is to study and increase the damage threshold of optical elements in laser systems. In order to scale the pulse energy and to properly size such lasers, larger and larger optical apertures have become necessary to ensure that the increased power is well tolerated by individual mirrors, beamsplitters, amplifier crystals, gratings etc. As the size of the optical components increases, the cost of production increases in a highly nonlinear way. Thus, if high damage threshold components can be produced, the overall cost of the whole laser system can be drastically reduced by not requiring the use of large aperture beams.

One of the topics of this thesis is therefore related to the field of high-intensity light-matter interactions. I have investigated the laser induced damage threshold of different optical elements from several points of view simultaneously. The literature available at the time of this work lacked papers covering multiple mirror characteristics simultaneously in the visible/near-infrared femtosecond pulse length range, so I performed systematic investigations in this respect. In my experiments, I measured absolute damage thresholds under conditions that closely approximate real-world use, applying a method that is also a fast procedure for comparing multiple optics [3].

Thanks to our German industrial cooperation (Layertec GmbH), I was able to perform experiments on a large number of samples. Some of these mirrors were experimental pieces, and some were real mirrors already available commercially. What they all have in common is that they are all designed and manufactured for femtosecond applications. I was able to find correlations between the manufacturing technology, the number of layers, the mirror material, the designed maximum field strength within the layers, the reflectivity, the group delay dispersion and the damage threshold. These aspects were all considered to evaluate the measurement results. The measurements were performed at a repetition rate of 1 kHz with 42 fs pulses at a wavelength of around 800 nm.

Optically induced current generation in dielectric media

Another interesting phenomenon of the interaction between laser light and dielectric media is the so-called optically induced transient metallisation. This newly discovered phenomenon follows the process predicted earlier by Jacob Khurgin [4], whereby it is possible to optically induce current in bandgap materials without using voltage bias. This new phenomenon was first experimentally demonstrated in 2013 [5]. Since then, more and more studies have been published on this topic. Although research into the origin of optically induced currents in insulators is still an active area, many application ideas are now being brought to life. The phenomenon itself is that a dielectric medium, illuminated by a high-intensity laser pulse with few optical cycles, becomes conductive for the time the pulse is transmitted and the electric field of the pulse causes a current to flow between metal electrodes placed on the surface of the dielectric. The magnitude and direction of this induced current is related to the shape of the pulse.

I have taken the application of optically induced currents further into more accessible application directions. I was the first one to implement this process with a compact titanium-sapphire oscillator that had not been used to demonstrate this phenomenon before. A unique oscillator, manufactured by Venteon GmbH, was available for this purpose, capable of providing pulses of <6 fs duration with an average power of about 270 mW and a repetition rate of 80 MHz. With these boundary conditions, I designed and built an experimental setup of reflective optical elements for measuring optically induced currents with strong focusing.

The difficulty of the experiment is that the beam expansion can only be achieved with reflective elements due to the extremely short pulse length, so that the required intensity can be achieved with a custom optical system with a focal spot size of 2 μm. Since the intensity required for optically induced current is close to $\sim 10^{13}$ W/cm^2 and at the same time it has to be ensured that the pulse duration at the sample surface remains around 5.5 fs, the optimal operating range with the available oscillator is narrow for all experiments performed.

By omitting optical amplifiers and using pulse energies of the order of 1 nJ and below, I was the first to demonstrate that it is possible to induce currents optically in bandgap materials by means of an external electromagnetic field, which can be measured by electrodes properly placed on the surface of dielectrics. From there, it is possible to move on to the optimisation and control of current generation and its applications, thanks to the high repetition rate of the pulses.

Theses

Based on my own research findings, I formulate the following thesis items.

1. Within high refractive index dielectric layers of highly reflective femtosecond laser mirrors, nonlinear processes, e.g. multiphoton excitation processes, can occur. By a comprehensive experimental investigation of 29 different layer systems, I have shown that by using materials with 1-2 eV higher bandgap (Ta_2O_5, ZrO_2, HfO_2, with bandgaps of 4.2 eV, 4.7 eV and 5.5 eV respectively) and simultaneously reducing the strength of the standing wave field in the mirror structure, the damage threshold can be increased significantly. Combining these two methods, the damage threshold can be increased up to 2.4 times for <50 fs pulses of most interest today [T1].

2. I investigated the behaviour of a new type of metal-dielectric hybrid mirrors for pulses <50 fs. Such mirrors typically consist of a silver layer deposited on a substrate and evaporated (or sputtered) onto it with only 3-7 layer pairs in contrast to the 20-30 layer pairs used in conventional high reflector (HR) mirrors. I have shown that in metal-dielectric hybrid mirrors, the presence of the silver layer does not inhibit femtosecond usage, i.e. a high damage threshold (comparable to conventional HR mirrors). In addition, a low group delay dispersion in the 730-880 nm range [T1, T2] can be achieved simultaneously. I have found that the reflection bandwidth is also increased by the presence of the silver layer. All these properties make metal-dielectric hybrid mirrors suitable for use with pulses shorter than 10 fs. Thus, I have also shown that metal-dielectric hybrid mirrors can meet the requirements for femtosecond applications with a significantly lower number of layers than HR mirrors [T1].

3. By testing 8 innovative femtosecond dielectric and metal-dielectric hybrid mirrors, I have shown that the number of shots used in the damage threshold experiments has only a small effect on the damage threshold in the 10-10^5-shot range, and that the shot-number-damage threshold characteristics are nearly identical regardless of the type of mirrors [T2]. I also found that the decrease in the damage threshold in the 10-10^5-shot range was not greater than 20% for any of the mirrors, indicating the negligible presence of incubation effects, which are crucial for damage and therefore intensively studied [T2].

4. I have shown that in dielectric media (fused silica and HfO_2), transient metalization of the medium can be induced by laser pulses with energies below 1 nJ [T3], a recently discovered ultrafast optical phenomenon that is in the focus of international research efforts. It is thus possible to optically induce currents in dielectrics with a large bandgap, such as fused silica, in addition to HfO_2, which is being tested as a new material (in this respect). By using the carrier-envelope phase of the pulses, the direction of the current induced can be controlled in the direction of one electrode or the other. Previously, this phenomenon could only be detected with pulses of at least 30-40 nJ using complex, amplified laser systems. I was the first one to construct a corresponding experimental setup based on a compact laser oscillator with a repetition rate of 80 MHz, optimizing a reflective parabolic reflection system to achieve both tight focusing and short pulse length in focus [T3]. Thus, I have improved the parameters required to implement current control by 2 orders of magnitude in terms of both repetition rate and pulse energy, paving the way toward the realization of PHz optoelectronic devices based on compact lasers.

Own publications

[T1] **V. Csajbók**, L. Szikszai, B. J. Nagy, és P. Dombi, "Femtosecond damage resistance of femtosecond multilayer and hybrid mirrors," Opt. Lett. 41, 3527-3530 (2016)

[T2] **V. Csajbók**, Z. Bedőházi, B. J. Nagy, és P. Dombi, "Ultrafast multipulse damage threshold of femtosecond high reflectors," Appl. Opt. 57, 340-343 (2018)

[T3] V. Hanus, **V. Csajbók**, Z. Pápa, J. Budai, Z. Márton, G Z. Kiss, P. Sándor, P. Paul, A. Szeghalmi, Z. Wang, B. Bergues, M. F. Kling, G. Molnár, J. Volk, és P. Dombi, "Light-field-driven current control in solids with pJ-level laser pulses at 80 MHz repetition rate," Optica 8, 570-576 (2021)

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[3] B. J. Nagy, L. Gallais, L. Vámos, D. Oszetzky, P. Rácz és a. D. P., „"Direct comparison of kilohertz- and megahertz-repetition-rate femtosecond damage threshold",” *Optics Letters 40,* 2525, (2015)

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