SUMMARY OF THE PhD DISSERTATION

Ablation and investigation of transient-reflectivity enhancements of Borofloat, BK7 and B270 optical glass samples by ultrashort laser pulses

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1.Introduction

The process of high-intensity laser-matter interaction going hand in hand with material removal (i.e. ultrashort laser pulsed ablation [1]) have been in the focus of the studies for decades since the first ultrashort, high-energy pulses were generated [2]. The material processing with fs pulses inducing ultrafast processes can be characterized with some special features making it desirable for the industrial applications. Two of such features are the deterministic behaviour of the ablation [3] and the pure ablated area [4].

Since the first generation of the high-energy ultrashort laser pulses, due to their stronger unwanted nonlinear effects, intensity enhancement has remained a challenge. There are two approaches to enhance the peak intensity of the pulses: increase the energy of the pump of the amplifier, or decrease the area of irradiation. The latter is more effective, reliable and easier method. However, the sample produces different ablation responses and parameters in case of different spot diameters [5]. It is mandatory to know the geometrical sizes of ablated craters to determine the ablation response functions. With knowing the changes of diameter and depth, one can determine the ablation threshold. Furthermore, the morphology of the ablated craters is an important aspect: the shape and surface roughness characterize well the quality of the material processing. However, a huge scattering of these parameters can be observed in the literature.

The appearance of the plasma mirror devices used for contrast-enhancement of ultrashort pulses [6] created a high demand for transparent dielectric materials, which can be used as plasma mirror targets. The plasma mirror target must be a material on which one can generate a high-reflecting plasma on the surface of the target by irradiating it with ultrashort laser pulses. Because of the small number of experimental results published on ablation of multi-component optical glasses in the literature, there is a high need for investigation of plasma mirror reflectivity enhancement and ablation behaviour with systematically varied laser parameters and samples with different compositions.

2. Goals

Due to the huge scattering in the literature data I aimed to understand that how changing of the spot diameter effects the reflectivity of the plasma mirror and ablation parameters. In order to investigate this, I aimed to irradiate BK7 glass sample with fs laser pulses at different irradiated spot diameters. I intended to determine geometrical sizes of the ablated craters etched into the sample and ablation thresolds at different spot diameters. Furthermore, my aim was to monitor the energy reflected from the irradiated area with a photodiode. Along with the investigations, I aimed to explain the observed phenomena.

In the second phase of my research work I aimed to study that how the chemical composition effects plasma reflectivity and ablation parameters. In order to perform this, I choose three different glass types and irradiated with 34 fs laser pulses. My goal was to find the glass type with which I can generate the highest reflecting plasma mirror on the surface and to explain the observed differences in the reflectivity values found in the literature for different glass types. Finally, I aimed the comparison of the ablation parameters and morphology of the etched craters and giving explanations for the observed similarities, differences.

3. Applied methods

I used single, 34 fs, 800 nm laser pulses to process the samples, which are provided by the TeWaTi laser system in the Department of Optics and Quantum Electronics. During the experiments I changed the pulse energy between 10 μJ and 420 μJ by a polarization-based beam attenuator. The experiments were carried out in athmospheric air.

I choose the Borofloat, BK7 and B270 glass sample from Schott to investigate. The samples were mounted in 45° angle related to the processing laser pulse in a sample holder adapter. I clamped the sample holder to *x*-*y* direction translation stage to move it horizontally and vertically proportional to the sample surface. The sample was moved in order to the irradiated spots on the target surface do not overlap with each other. A *z* direction translation stage was used to move the parabolic mirror focusing the beam along the beam propagation direction, which ensured the changing in the beam diameter on the sample surface. The beam reflected from irradiated spots was focused onto the sensor of a photodiode, which enabled the monitoring of the reflectivity changes on the irradiated spots initiated by the processing laser pulses by measuring the signal of the photodiode.

I ablated 11 craters on the surface of the samples at each pulse energy during my material processing experiments. Along with these, I recorded the photodiode signals generated by reflected beam. After this, the etched craters were undertaken by profilometric studies.

A DEKTAK8 stylus profilometer was used to investigate the craters. I measured profiles of each crater by this device. From the profiles I determined the diameter and depth of the ablated craters.

4. Thesis points

T1.1 I analyzed the energy density dependence of the diameter and depth of the ablated craters. According the results I showed, that when the spot diameter changes from 52.2 μm to 27.0 μm, the ablation threshold of the sample changes from 6.5 J/cm^2 to 5.1 J/cm^2. However, I observed, that further decreasing the spot diameter to 23.4 μm and 20.2 μm, the ablation threshold increased to 6.7 J/cm^2 and 10.6 J/cm^2, respectively. The statements of this thesis are explained based on the results of the following thesis point.

T1.2. I pointed out, that at 40.0 μm spot diameter the diode signal monitoring the transient reflectivity can be characterized by fitting 3 linear lines with different slopes at different pulse energy domains. These slopes are: 1.: s_1=0.30 ± 0.05 mV/µJ, 2.: s_2=0.90 ± 0.10 mV/µJ, 3.: s_3=0.25 ± 0.05 mV/µJ. Only 2 linear sections appear at the greatest spot diameter (52 µm), which were identified as the 1. and 2. categories. At spot diameters below 40.0 µm I observed 2 linear sections, too, however, these sections were identified as the 2. and 3. categories. The ratio of the slopes characterizes the changes in the reflectivity.

T1.3. I showed, that the ablation thresholds and the breakpoints, where the lines of 1. and 2. categories join agree well at 52.2 and 40.0 µm spot diameters. Based on this observation, I concluded, that the energy range of the section of the 1. category could be associated to the range of permanent reflectivity of the glass sample. As the ratios of the slopes of the 2. and 1. sections were found to be higher than one at every spot diameter, meaning that the reflection increases. This was explained as the plasma mirror induced reflectivity enhancement.

T1.4. I showed, that the ablation thresholds (T1.1) change with the increasing spot diameter similarly as the s_1/s_1 ratios change with the increasing spot diameter. When the spot diameter was set from 52.2 µm to 40.0 µm the s_2/s_1 values changed from 3.2 to 3.0. With further decreasing the spot diameter to 27.0 µm, 23.4 µm and 20.2 µm, the s_2/s_1 values increased to 3.2, 3.5 and 4.3. Based on the results mentioned here, I concluded, that the increase of the ablation threshold can be accounted for the increased reflection due to the plasma generation at the lowest investigated spot diameters.

T1.5. I observed, that the slopes of the sections of the 3. category is always smaller, than the 2. category at spot diameters smaller than 52.2 μm, meaning that at such pulse energies the reflectivity values decrease. The decrease of the reflectivity was explained through the air-ionization effects at the energy densities in the section of the 3. category. The ionized air-layer above the sample scatters the incoming beam causing the observed increase of the spot size and the diameter of the etched crater, and the saturation of the ablated depth values.

T2.1. I determined the plasma-mirror initiated reflectivity changings on the irradiated area of three investigated glass samples as a function of energy density between 0.68 and 28 J/cm^2. I concluded, that at 25 J/cm^2 energy density the reflectivity enhances with 400%, 200% and 135% respect to the permanent values in case of Borofoat, BK7 and B270, respectively.

T2.2. I pointed out, that the average electron numbers per 1 mol material, which can be excited to the conduction band by irradiation changes in the same way, as the reflectivity enhancement values for the three glass samples. The correlation of the reflectivity and the electron number points out, that the deviations in the reflectivity could be explained by the stochiometric differences of the investigated materials.

T3.1. I determined the diameter and depth of the ablated craters etched into the investigated glass samples as a function of the energy density. Furthermore, I showed, that above ablation threshold the diameters follow a logarithmic dependence for all three investigated glasses. I determined from the diameters by means of regression method the ablation thresholds for the three investigated glasses, which lie between the 5.85 - 6.43 J/cm2 values. Fitting the multiphoton absorption based fit I showed, that the evolution of the depth as a function of the energy density can be very well described by the three-photon absorption form of the model for all three investigated glasses. I determined the ablation thresholds from the fitting parameters of the curve fitted to the depth values, as well. The resulting ablation threshold values lie between the 5.65 – 6.65 J/cm2. I found, that the thresholds determined by the two techniques agree well with each other.

T3.2. I investigated the morphology of the ablated craters by means of analysing the recorded profilometric traces. I found, that B270 exhibits the smoothest surface (Ra = 3.0 nm). Borofloat has a surface roughness lying between the two other investigated glasses (Ra = 4.7 nm) and BK7 exhibits the largest surface irregularities (Ra = 10.1 nm) in the crater profiles.

T3.3. From the functions fitted to the depth I determined the three-photon absorption coefficients of the investigated glasses: I got 5.45

$\times 10^{-25}$ cm^3/W^2 for Borofloat, 6.35 $\times 10^{-25}$ cm^3/W^2 for BK7 and 8.28 $\times 10^{-25}$ cm^3/W^2 for B270. Using these values, I explained the differences observed in the reflection enhancements and morphology of the glasses.

T3.4. I pointed out, that for all glasses investigated the ablated volumes increase linearly with the energy density until 20 J/cm2, however, above this energy density, the increase of the volume is slower. From the similar behaviour of the ablated crater parameters and ablation thresholds I showed, that, unlike their very different reflectivity enhancements, the investigated glasses do not differ from each other from point of view of their ablation response. This apparent discrepancy was interpreted as the result of different mechanisms determining reflectivity enhancement and the material removal: the reflectivity increase of the plasma is influenced by the excited average electron numbers and three-photon absorption coefficients being different for the glasses, while the ablation is governed by the thermal parameters being nearly the same for the three glasses.

5. Publications

Publications related to the thesis points

[T2.1, T2.2, T3.4] A. Andrásik, R. Flender, J. Budai, T. Szörényi, B. Hopp, Time integrated transient reflectivity versus ablation characteristics of Borofloat, BK7, and B270 optical glasses ablated by

34 fs pulses, Optical Materials Express **10**(2) pp 549-560 (2020), MTMT identifier: **31137745**

[T3.1, T3.2, T3.3] A. Andrásik, R. Flender, J. Budai, T. Szörényi, B. Hopp, Processing of optical glasses by single, 34 fs pulses in the strong field ionization domain: ablation characteristics and crater morphology, Applied Physics A – Materials Science and Processing **126**(12) 936 (2020), MTMT identifier: **31664065**

[T1.1, T1.2, T1,3, T1.4, T1.5] A. Andrásik, J. Budai, T. Szörényi, Cs. Vass, R. Flender, B. Hopp, Spot size dependence of the ablation threshold of BK7 optical glass processed by 34 fs pulses, Laser Physics (2022) MTMT identifier: **33062901**

Other publications

[4] E. Nagy, **A. Andrásik,** T. Smausz, T. Ajtai, F. Kun-Szabó, J. Kopniczky, Z. Bozóki, P. Szabó-Révész, R. Ambrus, B. Hopp, Fabrication of Submicrometer-Sized Meloxicam Particles Using Femtosecond Laser Ablation in Gas and Liquid Environments, Nanomaterials, **11**(4) 996 (2021) MTMT identifier: **31971131**

[5] K. Ludasi, O. Jójárt-Laczkovich, T. Sovány, B. Hopp, T. Smausz, **A. Andrásik,** T. Gera, Zs. Kovács, G. Regdon jr, Anti-counterfeiting protection, personalized medicines - Development of 2D identification methods using laser technology, International Journal of Pharmaceutics, **605** 120793 (2021), MTMT identifier: **32101093**

Conference papers and posters

[6] A. Andrásik, R. Flender, J. Budai, T. Szörényi, B. Hopp**,** Single-shot surface ablation and transient reflectivity changes of optical glasses induced by 34 fs laser pulses, In: P. Bakule, CL Haefner, Short-pulse High-energy Lasers and Ultrafast Optical Technologies, Bellingham (WA): International Society for Optical Engineering (SPIE), Paper 110340T (2019) (Proceedings of SPIE 0277-786X 1996-756X;11034) MTMT identifier: **30823508**

[7] A. Andrásik, R. Flender, J. Budai, T. Szörényi, B. Hopp, Surface processing of optical glasses with 34 fs pulses: ablation thresholds and crater shape, In: P. Földi, I. Magashegyi Kvantumelektronika 2021: Szimpózium a hazai kvantumelektronikai kutatások eredményeiről, Szeged: Szegedi Tudományegyetem TTIK Fizikai Intézet, pp 1-6 (2020), MTMT identifier: **31832820**

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[9] A. Andrásik, Sz. Toth, R. S. Nagymihály, P. Jójárt, R. Flender, Á. Börzsönyi, K Osvay, Development of few cycle Ti:Sapphire and NOPA amplifiers at 80 MHz repetition rate, SPIE Optics +

Optoelectronics, Konferencia helye, ideje: Prága, Csehország 2017.04.24 - 2017.04.27. pp 143-143 (2017), MTMT identifier: **3262077**

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6. References

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