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Damage mechanisms of ultrashort pulsed laser processing of glass in dependency of the applied pulse duration

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Abstract

We report on the transmission of ultrashort pulsed laser radiation for glass processing with single laser pulses. The applied pulse duration is varied from 80 fs to 9 ps and the intensity from $5*10^{11}$ to $5*10^{14}$ W/cm², respectively. Above a certain intensity threshold, the transmission of the laser radiation continuously decreases. This indicates an increased absorption of the laser pulse due to the generation of a denser free-electron plasma. Additionally, we observe visible damage of the glass by means of optical microscopy imaging. The intensity threshold decreases by more than one order of magnitude from $22*10^{12}$ W/cm² for 80 fs to $2*10^{12}$ W/cm² for 9 ps. This can be explained by different dominant ionization mechanisms, which depend on the applied pulse duration and intensity. By comparing our experimental results with simulations performed by other groups, we find that for a pulse durations of 80 fs, the damage of material is ascribed to the generation of a free-electron plasma by multiphoton ionization. For longer pulse durations 1 ps and 9 ps, the generation of the free-electron plasma and the damage of material is induced primarily by impact ionization.

Keywords: Ultrashort laser pulses, processing transparent materials, absorption, ionization mechanisms

1. Introduction

The processing of transparent materials with an wide electronic bandgap can be achieved by the application of laser pulses with high intensities in the order of 10^{12} to 10^{14} W/cm² realized by focused ultrashort pulsed laser radiation. Due to the complex superposition of linear and nonlinear excitation and interaction effects for these high intensities, an improved understanding of the role of the different

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ionization processes and the laser-plasma interaction is crucial for processing transparent materials as reported in our recent study (Kalupka et al., 2017). The initial excitation of electrons from the valence to the conduction band takes place by nonlinear photoionization processes like tunnel and multiphoton ionization. Subsequently, the free-electrons gain kinetic energy due to inverse bremsstrahlung and excite more electrons from the valence band by impact ionization mechanism. The subsequent avalanche ionization leads to a dense free-electron plasma with quasi-metallic optical properties. Therefore, a strong absorption of the laser pulse energy can be achieved, which leads to strong heating of the lattice and eventually melting and vaporization.

According to numerical simulations, nonlinear multiphoton ionization and impact ionization exhibit different dependencies on the intensity distribution (Rethfeld, 2006). The generated free-electron density ρ is described by a simple rate equation:

$$d\rho/dt = \sigma I(t)^k + \alpha(\rho)\, I(t). \tag{1}$$

The first term represents the multiphoton ionization rate, where I is the laser intensity, σ the multiphoton coefficient and k the number of photons needed for excitation of one electron into the conduction band. The second term describes the ionization rate due to impact ionization, which depends on the intensity I and on the impact coefficient $\alpha(\rho)$. Here, to focus on the ionization processes only, we neglect recombination processes in equation (1). The intensity distribution $I(t)$ is a function of time, that is described by a Gaussian intensity distribution with pulse duration τ and a peak intensity I_0. Because the generated free-electron plasma affects the intensity distribution due to reflection and absorption and vice versa, there is no analytical solution of the rate equation. In general, equation (1) is solved by numerical methods to calculate the interaction of plasma and laser pulse stepwise (Sun et al., 2013).

In this study, we present an experimental approach to investigate the ionization mechanisms for transparent glass. With regard to the rate equation (1), the contribution of multiphoton and impact ionization to the total number of free-electrons depends on the intensity distribution. Furthermore, in dependency on the pulse duration τ, the interaction time of laser pulse and the generated free-electron plasma can be adjusted. According to this model, the absorption of the laser pulse depends on the intensity on the one hand side and on the pulse duration on the other hand side. Absorption *A*, reflection *R* and transmission *T* are correlated by $A + R + T = 1$. Here, we neglect scattering of the laser pulse. Studies by Varkentina et al. show that the change of transmission is several times larger compared to the changes of reflection (Varkentina et al., 2013). Thus, we measure the transmission *T* of ultrashort pulsed laser radiation for glass processing in dependency on the intensity and pulse duration. Changes of the transmission *T* of the laser radiation are associated to a change of the absorption *A* due to absorption by a generated free-electron plasma.

2. Experimental

A Ti:Sapphire chirped pulse amplification laser system (Libra, Coherent) provides linearly polarized single laser pulses with a central wavelength of 800 nm, a pulse energy of 2 mJ and a beam quality factor of 1.2 to carry out the experiments (Fig. 1). The pulse duration τ can be selected between 80 fs and 9 ps (Gaussian distribution, FWHM) by the adjustment of the propagation distance of the spectrally broadened pulse inside the compressor unit of the laser system. The pulse energy E_P is adjusted by reflective filters and a combination of a half-wave plate and a polarizing beam splitting cube. The laser radiation is focused on the surface of the glass sample by a 10x microscope objective with a numerical aperture of 0.25. This results in a focal diameter of approximately $2w_0$ = 3 µm. The intensity *I* on the glass surface is calculated by

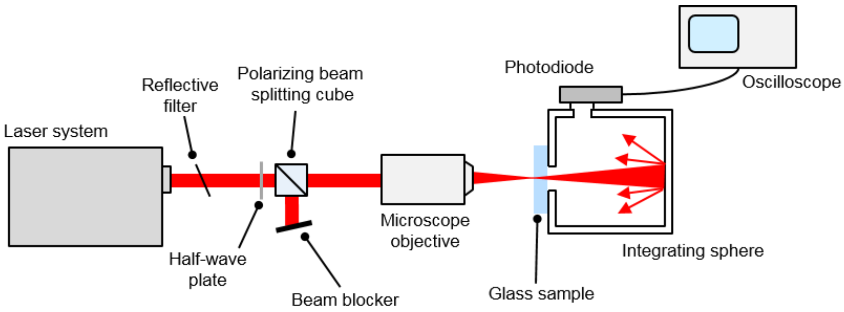

Fig. 1. Scheme of the experimental setup for the transmission measurements.

$$I = 0.94E_P/(\pi w_0^2\tau). \quad (2)$$

In this study, we use 700 µm thick alkali-aluminum-silica glass (Gorilla®, Corning®). For the transmission measurements, the glass sample is mounted on the entrance side of an integrating sphere.. The integrating sphere is mounted on a linear xy-axis in order to irradiate an unmodified are of the glass sample for each measurement. A fast photodiode is mounted on one exit hole of the integrating sphere for the measurement of the transmitted optical signal and the conversion into an electronic signal. The electronic signal is recorded by a digital oscilloscope. To calculate the normalized transmission, we perform measurements under the same conditions without the glass sample. For one selected intensity *I*, the transmission *T* is calculated by

$$T = U/U_{\mathrm{w/o\ glass}} \quad (3)$$

with *U* the measured voltage with the glass sample mounted on the entrance side of the integrating sphere and $U_{w/o\ glass}$ the measured voltage without the glass sample. To avoid deviations e.g. due to surface impurities or defects, we repeat the measurements 50 times for each intensity and calculate the mean value and the standard deviation of the transmission.

3. Results and discussion

In the following, the results are presented and discussed in two sections. First, the transmission of a single laser pulse through the glass sample is shown in dependency of the applied intensity for three different pulse durations. Furthermore, we compare the results to simulations performed by Rethfeld, to investigate the impact of the underlying ionization mechanism on the transmission of the laser pulse (Rethfeld, 2006). Second, we present microscope images of the glass surface for the shortest and longest pulse durations to examine the correlation of the underlying ionization mechanisms with visible damage morphology.

3.1. Transmission of ultrashort laser pulses

In Fig. 2 (a), the transmission of single laser pulses through the glass sample is shown in dependency of the peak intensity in the range from $5*10^{11}$ to $5*10^{14}$ W/cm² for three different pulse durations 80 fs, 1 ps and 9 ps, respectively.

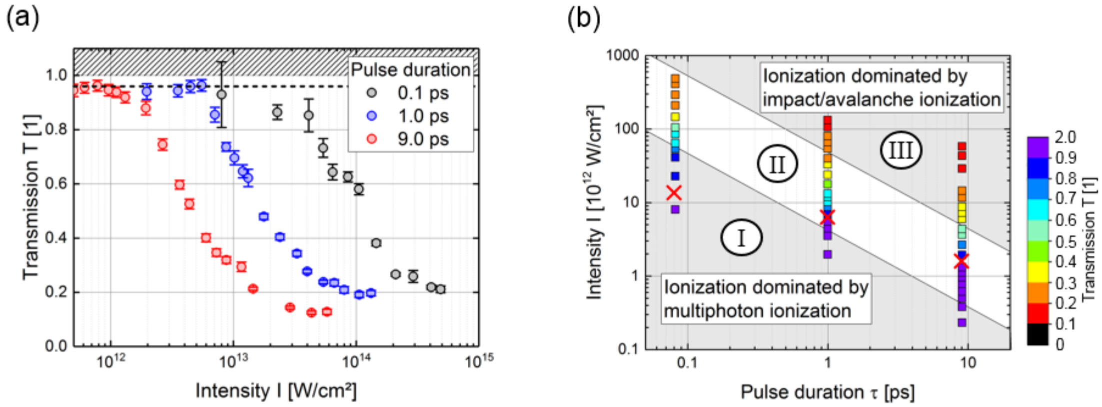

Fig. 2. (a) Measured transmission of single laser pulses for three pulse durations (80 fs, 1 ps, 9ps) in dependency on the intensity. The dashed line represents the intrinsic transmission of the glass according to ultraviolet-visible spectroscopy for a wavelength of 800 nm and normal incidence. (b) Comparison of the measured transmission with different ionization regimes (I), (II) and (III), that are identified by simulations performed by Rethfeld (Rethfeld, 2006).

The overall qualitative dependency of the transmission on the intensity is comparable for each investigated pulse duration. For intensities below a certain intensity threshold I_{thr}, the transmission is constant and estimated to approximately 95 ± 2 %. This corresponds to the intrinsic transmission of the glass sample for the wavelength of 800 nm, which is measured by linear ultraviolet-visible spectroscopy (indicated by a dashed line in Fig. 2 (a)). Moreover, this value is in accordance with the transmission of 93 % calculated by the Fresnel equation for two glass-air interfaces with a refractive index of the glass sample of approximately $n \approx 1.5$ for normal incidence. Thus, for intensities below the intensity threshold I_{thr}, absorption of the laser pulse by a free-electron plasma has a vanishing contribution to the measured transmission. Therefore, nonlinear ionization processes play a minor role.

The intensity threshold I_{thr} marks the onset of a decrease of the transmission for an increase of intensity. This indicates that nonlinear ionization processes and an accompanying absorption of the laser pulse by a generated free-electron plasma have to be taken into account for $I > I_{thr}$. For higher intensities, the plasma density increases which leads to a stronger absorption and therefore a lower transmission. For the highest investigated intensities, the transmission reaches a minimum value in the range from 12 to 20 %. Thus, the absorption remains constant for a further increase of the intensity, which can be explained by a complete first ionization of the material (Gamaly et al., 2014).

The value of the intensity threshold I_{thr} strongly depends on the pulse duration and decreases by more than one order of magnitude for an increase of the pulse duration by approximately two orders of magnitude. The thresholds are estimated to $I_{thr} = 1.5 \pm 0.7 * 10^{13}$ W/cm² for a pulse duration of 80 fs, $6.3 \pm 0.7 * 10^{12}$ W/cm² for 1 ps and $1.2 \pm 0.1 * 10^{12}$ W/cm² for 9 ps, respectively.

To find physical explanations for the varying intensity thresholds I_{thr} for different pulse durations, we compare our results to simulations performed by Rethfeld (Rethfeld, 2006). The simulations allow for the prediction of the dominant ionization mechanism for a selected combination of intensity and pulse duration. For a comparison, in Fig. 2 (b), the measured transmission is shown in a color map as a function of intensity and pulse duration. The estimated intensity thresholds I_{thr} from our measurements are indicated by red crosses. Moreover, three distinct regions (I), (II) and (III) are shown. According to Rethfeld, in the first region (I), the ionization of the material is dominated by the multiphoton mechanism, which means that more than 95 % of the total number of free-electrons are generated by multiphoton ionization. In the second region (II), the contributions of multiphoton and impact ionization to the total number of generated free-electrons are comparable. In the third region (III), multiphoton ionization plays a minor role because more than 95 % of the free-electrons are generated by impact ionization.

For a pulse duration of 80 fs and intensities slightly larger than the intensity threshold I_{thr}, absorption takes place by a free-electron plasma, which is generated primarily by multiphoton ionization. This is indicated by a measured transmission < 90 % in region (I). In contrast to this, for pulse durations 1 ps and 9 ps and intensities near the threshold intensity, absorption only takes place when a comparable amount of the free-electron plasma is also generated by impact ionization, which is indicated by a measured transmission < 90 % in region (II). The role of impact ionization is more pronounced for 9 ps compared to 1 ps. Therefore, multiphoton ionization only is not capable to induce a dense absorbing free-electron plasma for pulse durations 1 and 9 ps. The different contributions of multiphoton and impact ionization mechanism for different pulse durations provide an explanation for the observed variation of the intensity threshold. It is important to mention, that in the numerical simulations by Rethfeld, a temporal uniform pulse shape is assumed. This differs compared to our experimental studies, in which the temporal shape of the laser pulse exhibits a Gaussian distribution. Thus, for a detailed quantitative comparison, the simulations should be redone with a temporal Gaussian intensity distribution. Moreover, material parameters have to be chosen to fit the glass we use in our experiments. Additionally, the simulations are performed for one discrete intensity value. In our experiments the spatial intensity profile is a Gaussian distribution and therefore the transmission is measured for a superposition of several different intensity values.

3.2. Visible damage and modifications

In order to examine the correlation of dominating ionization mechanism with visible damage morphology, microscope images of the irradiated glass surface for pulse durations 80 fs and 9 ps are shown in Fig. 3. Images are shown for intensities close to the intensity threshold I_{thr} and for intensities approximately up to one order of magnitude larger than I_{thr}.

For pulse durations 80 fs and 9 ps, visible modification is observed for intensities exceeding the intensity threshold, $I > I_{thr}$. Therefore, the intensity threshold is identified with the modification threshold of the glass sample. For the shortest pulse duration 80 fs, an ablation crater with a rim structure is observed. According to the discussion in section 3.1., the ionization process is dominated by multiphoton ionization for intensities below $6*10^{13}$ W/cm². Therefore, primarily the multiphoton ionization mechanism generates a dense free-electron plasma, which results in visible damage of the material. An increase of the contribution of impact ionization to the total number of free-electrons by increasing the intensity above $6*10^{13}$ W/cm² leads to larger diameter of the modified region at the glass surface.

For a pulse duration of 9 ps, a dark central region and a surrounding bright ring structure are observed. Compared to section 3.1, for intensities below $0.4*10^{13}$ W/cm², multiphoton and impact ionization provide a comparable contribution to the total number of generated free-electrons. For these intensities, a slight modification of the glass surface can be detected. For intensities exceeding $0.4*10^{13}$ W/cm², ionization is dominated by impact ionization and modification can clearly be observed. For a further increase of intensity, the role of impact ionization increases even more and the diameter of the structure increases and additional bright and dark structures occur. The diameter of these structures is much larger than spot diameter of approximately 3 µm and therefore also several times larger than the ablation crater produced by 80 fs for comparable intensities.

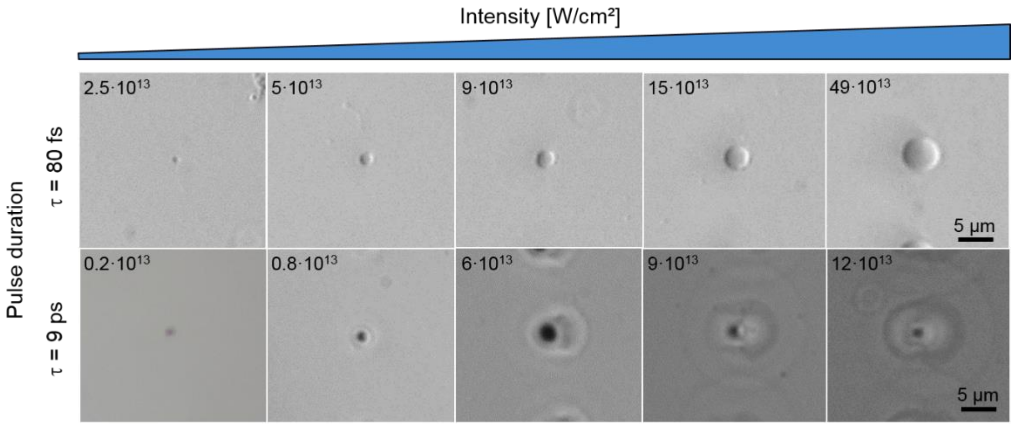

Fig. 3. Transmission microscope images of an irradiated glass surface with a single laser pulse with the indicated intensity for two pulse durations 80 fs and 9 ps, respectively.

A direct comparison of the modifications for 80 fs and 9 ps for one selected intensity of $9*10^{13}$ W/cm² in Fig. 3 shows a significant difference of visible material damage. With regard to Fig. 2 (b) this can be explained by a different contributions of impact ionization to the overall ionization process. Moreover, we suggest that the modifications for 80 fs correspond primarily to an ablation of the glass surface. In addition to this, for 9 ps, a significant large amount of the glass volume is supposed to be modified, e.g. due to changes of the local density of material as a consequence of heating, which eventually leads to changes of the local refractive index. Thus, for an extensive investigation of the ionization and the laser-plasma interaction process, the transversal and longitudinal spatial dimensions have to be considered for instance by 1D and 2D beam propagation, respectively.

4. Conclusion

In summary, we measured the transmission of ultrashort pulsed laser radiation of alkali-aluminum-silica glass for pulse durations 80 fs to 9 ps and for intensities in the range from $5*10^{11}$ to $5*10^{14}$ W/cm². For intensities below a specific intensity threshold, the transmission is approximately 95 %, which corresponds to the intrinsic transmission of the glass sample for a wavelength of 800 nm. Therefore, absorption of the laser pulse by nonlinear ionization processes plays a minor role. An increase of the intensity above the threshold intensity leads to a decrease of transmission which is explained by nonlinear ionization and the subsequent generation of a free-electron plasma, which absorbs and reflects the laser pulse. For the highest applied intensities, the transmission reaches a minimum that is explained by a complete first ionization of the material, which corresponds to a maximum free-electron density. The threshold intensity decreases for an increase of the pulse duration from 80 fs up to 9 ps by more than one order of magnitude. The comparison of our results with simulations shows that for intensities in the range of the threshold intensity, for 80 fs the generation of a free-electron plasma primarily takes place by multiphoton ionization. In contrast to this, for 1 ps and 9 ps, then plasma can only be generated by a combination of multiphoton and impact ionization. Multiphoton ionization only is not capable to generate a dense free-electron plasma, which affects the laser pulse by reflection and absorption. In addition to the transmission measurements, we record optical microscopy images of the irradiated glass surface. Modification of the surface is detected for intensities exceeding the intensity threshold, therefore the intensity threshold is identified with the modification threshold. For the pulse duration 80 fs, modification of the glass surface can be generated by multiphoton only, whereas for 9 ps, impact ionization plays a major role in the overall ionization process. The morphologies of the produced ablation craters differ significantly, which can be explained by different contributions of the underlying ionization mechanisms. Moreover, in contrast to 80 fs, we observe a modification of the glass volume for 9 ps. This gives evidence that for a full investigation of the laser-induced excitation process and the laser-plasma interaction, spatially and time-resolved measurements have to be applied to investigate the linear and nonlinear beam propagation inside the glass volume and the excitation process of the free-electron plasma.

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