

DAMAGE THRESHOLD IN ABLATION REGIME INDUCED
BY FEMTOSECOND LASER IRRADIATION ON
TRANSPARENT MEDIA

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Abstract

The current work is a case study of ongoing in-depth extensive fundamental research on the plasma formation and relaxation dynamics in transparent media induced by mid- and short-wavelength infrared laser pulses. The experimental investigation is performed on a 35 fs-laser system setup. The working parameters such as laser energy and the number of pulses are varied. The experimental measurements are applied on borosilicate glass samples doped with gold nanoparticles. The effects of the applied laser pulses and their consequence on laser-induced ablation damage thresholds are investigated and discussed. The results from this case study provide valuable information and a deeper understanding plasma formation and dynamics induced by femtosecond infrared laser pulses in solid transparent glass media.

Key words: laser ablation, damage threshold, femtosecond lasers, transparent media

Introduction. The motivation of the research is based on the aim of contributing to a better understanding of the mechanism of interactions between fem-

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tosecond laser irradiation and solid transparent materials leading to new technological applications in e.g., non-linear optics, nanostructure generation, bioimaging, and 5D data storage.

In general, the occurrence of surface damage on optical materials is due to the presence of residual defects of sub-micron size in the glass media, such as inhomogeneities in the volume or on the surface, and metallic or dielectric inclusions [1]. It is known that these defects absorb partially the laser energy and lead to the formation of craters following the laser pulse. The research on such laser-induced damage is of great importance for improving the requirements for glass quality and polishing, resulting in higher damage resistance.

To study the initiation mechanism of laser-induced damage in optical materials, starting with the nano- to femtosecond regime, enormous research and scientific effort has been done over the last decades and numerous studies are available in the literature. Among these, many studies are devoted and aim to investigate the laser-induced damage threshold of a wide range of optical materials [2-4]. In recent years with the fast development of the chirped amplification techniques many investigations have been focused on the study of ultrafast absorption in dielectrics. A variety of applications are based on the interaction of femtosecond pulses with transparent materials, such as basic scientific research, micro-fabrication with nano-scale features, optical communication, biomedicine, bioimaging, environmental monitoring, glass processing for consumer electronics, data storage, and transfer. The knowledge of the optical properties of the material exposed to laser irradiation is thus of paramount importance for precise and fine control of the laser absorption process. To achieve the best control of such processes, it is important to provide in-sign not only to the amount of laser energy necessary to the material surface, to limit the laser-induced stress and to avoid degradation. Studies of the transient optical properties of dielectric materials irradiated by femtosecond laser pulses dealing with the ablation processes, ablation dynamics, damage thresholds, and mechanisms are reported, as well as experimentally and theoretically proven [5-9]. Unlike optical damage caused by nano- and pico-second pulses, the damage mechanism of the femtosecond laser is mainly due to the accumulation of electrons rather than thermal accumulation. In the case of femtosecond laser interaction with the surface of glass samples, several interaction modes including ablation and damage occur. The laser-induced damage initiated by ultrashort laser irradiation is initiated and includes the multiphoton ionization process, energy absorption, collisional ionization, and plasma formation. The material exhibits optical damage when the plasma energy exceeds the damage threshold of the material [7,9-11], and the damaged processes are strongly affected by the material properties, laser parameters, and homogeneity of the surface area.

In this work, we report on research devoted to the study of laser-induced damage and ablation thresholds of optical material as a function of laser and material parameters. The investigation is performed on native borosilicate glass

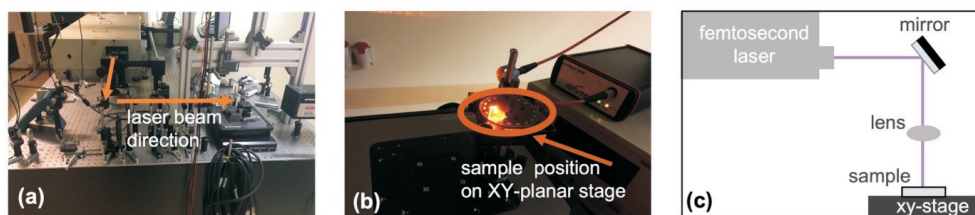


Fig. 1. Image of the experimental setup with the laser propagation direction (a), the sample illuminated at 600 nm for better visualization (b), and a schematic drawing of the setup configuration (c)

and with additions of Au nanoparticles irradiated by femtosecond laser pulses. The main findings are presented and discussed, followed by concluding remarks.

Experimental configuration and materials. *Experimental setup.*

The experimental investigation is performed on a femtosecond laser system setup including regenerative Ti Sapphire amplifier emitting at 800 nm central wavelength with a pulse duration of 35 fs and 1 kHz repetition rate, with maximum energy per pulse 6 mJ. An autocorrelation system (GRENOUILLE 8-20-USB) is applied to monitor and control the main output parameters of the fs laser radiation. The femtosecond laser is further equipped with a high-precision planar XY stage (Aerotech, Two-Axis Motor Stage), a power meter (Ophyr, Nova II), which covers a range of (300–1100 nm) wavelengths, and (20 μ J–10 J) impulse energies, and optical and optomechanical elements. The experimental setup configuration is presented in Fig. 1.

Figure 1(a) shows an image of the femtosecond laser system with arrows demonstrating the laser beam direction as well as showing the realistic scale of the setup configuration. The samples are placed on the XY-planar stage which provides precise control of the sample's area to be irradiated. Figure 1(b) shows an illuminated at 600 nm sample of borosilicate glass for better visualization and representative reason. The laser source is guided via an ultrafast dielectric mirror and focused on the sample surface using a doubly convex achromatic lens with a focal length of 25 cm. The sample is placed on the XY-translation table that allows changing the position of the sample while changing the laser source power. A power attenuator is used to regulate the power level.

The Laser-induced breakdown spectroscopy (LIBS) analyses are performed on a LIBS-6 Applied Photonics laser system with a pulse duration of 10 ns, pulse energy of 197 mJ, and repetition rate of 20 Hz.

Materials. The investigated borosilicate glass samples are a composition of Wt 50% SiO₂, 20% Al₂O₃, 20% B₂O₃, 5% CaO, 2% Li₂O, 3% MgO. Hydrogen tetrachloroaurate (III) hydrate (HAuCl₄) is added to the basic oxide mixture to ensure the gold concentration, apart from the samples of the basic glass material, without noble Au metal.

The size of the samples is approximately 12×15 mm and a thickness of 3 mm. Previously performed optical transmission measurements [12] indicate that the samples are transparent above a wavelength of 350 nm and the spectra do not depend on the noble metal content as well as the zones with modified optical properties expand all over the sample thickness due to the glass transparency at 800 nm and the higher Rayleigh lens range than the sample thickness of 3 mm. The samples under investigation are divided into three groups, native borosilicate glasses, borosilicate glasses doped with Au nanoparticles, and doped ones after annealing. A conventional melt-quenching technique is applied for the synthesis of the samples, the annealing procedure is performed in Pt crucible at 1450 °C. More detailed information on the sample concentration and composition is reported in our previous works [12–14].

Results and discussions. The laser damage threshold measurements in the ablation regime are applied to glass samples, initially divided into three groups. The first group corresponds to glass samples without doped particles, the second one with doped, and the third one with doped and annealed. The used ion concentration of the samples is 1% wt. The femtosecond laser processing is performed by the femtosecond laser system emitting at a wavelength of 800 nm. The applied laser energy is varied in the range of 5 to 250 μ J. It is found that below energies of 20 μ J no surface modification is observed. The number of applied laser pulses is 1000. In the case of multiple pulses acting on the glass surface, the former pulse strengthens the energy absorption of the latter pulse. And when defects are generated in the glass sample, the energy is deposited into the material, so that the defect energy level appears in the forbidden band, which strengthens the absorption of the latter pulse and leads to the modification of mechanical or chemical damage.

LIBS measurements and analyses are performed to verify the presence of gold additions in the glass material. The spectrum of LIBS measurements is based on the irradiation of a sample with a focused laser beam that results in the ablation of a very small quantity of material and the formation of a plasma plume, containing atoms and ions of the elements present in the sample. Analyzing the light emitted by the plasma plume information of the elemental sample composition is obtained. The spectra are measured for each sample by the LIBS laser system. Figure 2 presents an example of two samples – the one denoted by the red line corresponds to Au-doped, and the one denoted with the black line corresponds to the Au-doped and annealed sample. The figure demonstrates that in the case of the annealed sample (black line) the presence of new gold lines appears in the wavelength range of 316–340 nm, confirming the presence and formation of Au nanoparticles in the media volume at different measured positions. It is found that the presence and formation of golden nanoparticles is observed in the entire volume after annealing and not only in the irradiated by the femtosecond laser zones.

The experiment configuration allows the diameter and the depth of the ab-

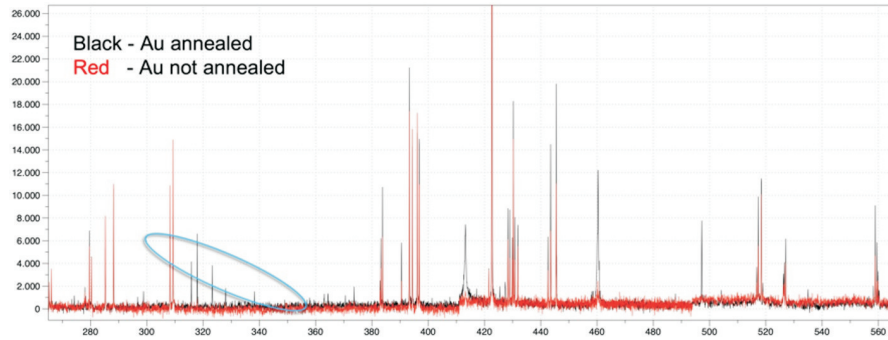


Fig. 2. A LIBS spectrum of two investigated samples doped with Au. The red line corresponds to the sample with Au addition and the black line corresponds to an annealed sample. The spectrum proves the appearance of formed gold nanoclusters by the presence of new Au lines in the range of 316–340 nm

lated crater, which has been created by irradiation using different laser powers and impulse lengths to be measured. The glass response to the laser radiation is studied below and above the ablation threshold. It is defined by extrapolation of the dependence of the ablation depth and crater diameter versus Laser fluence. The experiments are performed in the air. The ablation depth profile is measured by an optical microscope which observes the crater diameter at fixed depth steps between the sample surface and the crater bottom. Further, the damage threshold is calculated by a linear regression algorithm. In its origin, the linear regression method measures the damage threshold energy of the material to the linear relationship between the damage crater diameter and the applied laser energy. The relationship between the laser energy, the damage threshold energy, and the damage diameter is calculated from the experimentally measured dependence and can be presented as follows:

$$(1) \quad D^2 = 2\omega_0^2 \ln(E_{\text{pulse}}/E_{\text{th}}),$$

where D is the crater's diameter on the surface, and E_{pulse} is the laser energy per pulse. The laser beam diameter is calculated from the experimentally measured dependence of the crater's diameter, obtained on the irradiated surface, on the laser beam diameter using the relation above.

The laser energy E_{pulse} can be calculated according to the equation:

$$(2) \quad E_{\text{pulse}} = PR,$$

where P is the laser's average power and R is the repetition rate of the laser. Through the linear fitting curve between D^2 -LnE and the intercept x_0 of the curve and the x-axis, the laser damage threshold flux $E_{\text{th}} = \exp(x_0)$ corresponding to the optical material can be obtained.

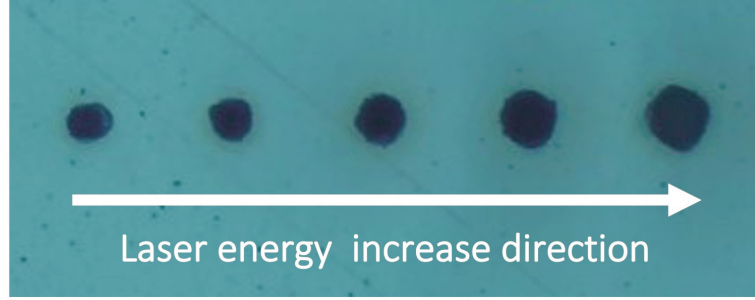


Fig. 3. An example from a microscope image of formed craters on the borosilicate doped with Au glass surface irradiated at different laser energies varied in the range from 50 to 250 μJ

The ablation threshold (F_{th}) is then calculated using the method described in [7,15] and is written in terms of the energy density as:

$$(3) \quad F_{\text{th}} = 2E_{\text{th}}/\pi\omega_0^2,$$

where E_{th} is the minimal energy in the impulse, that leads to ablation, and ω_0 is the diameter of the laser beam at the focal point of the lens. The laser pulse energy is measured and obtained by the laser pulse energy meter.

Figure 3 presents images of craters ablated at energies varied from 50 to 250 μJ with a step of 50 μJ , where each of the four energies corresponds to a crater (the black dots). The images show a smooth centre surrounded by an elevated rim from which very thin strips of melted material are extending away. The elevated rim seems to form as a result of a pressure-driven flow of the melt from the centre to the edge of the crater followed by resolidification at the rim. The morphologies of damaged craters are uneven with increasing laser power. The increase in the crater size follows the increase of size dependence as expected by the accumulated energy increases as well.

The reported experimental data in Fig. 4 represent a plotting D^2 versus the measured fluence and a numerical fit enables us to find the damage threshold, determined by the fluence for which the linear fit goes down to $D^2 = 0$. The black line corresponds to the native borosilicate glass sample; the red line corresponds to Au-doped sample, and the blue line corresponds to the Au-doped and annealed glass sample. It can be seen from the graphic that the native glass sample and the one doped with Au follow similar dependence and have close damage threshold values. The observed deviation in the case of Au-doped and the annealed sample could be explained due to the presence of gold nanoparticles over the entire volume of the sample and the presence of the plasma mirror effect that leads to a higher reflection coefficient, which has an impact on the laser energy balance and could affect the interaction dynamics by strongly localizing the deposition of the incident energy in the skin depth [6]. In the case of Au-doped samples, the

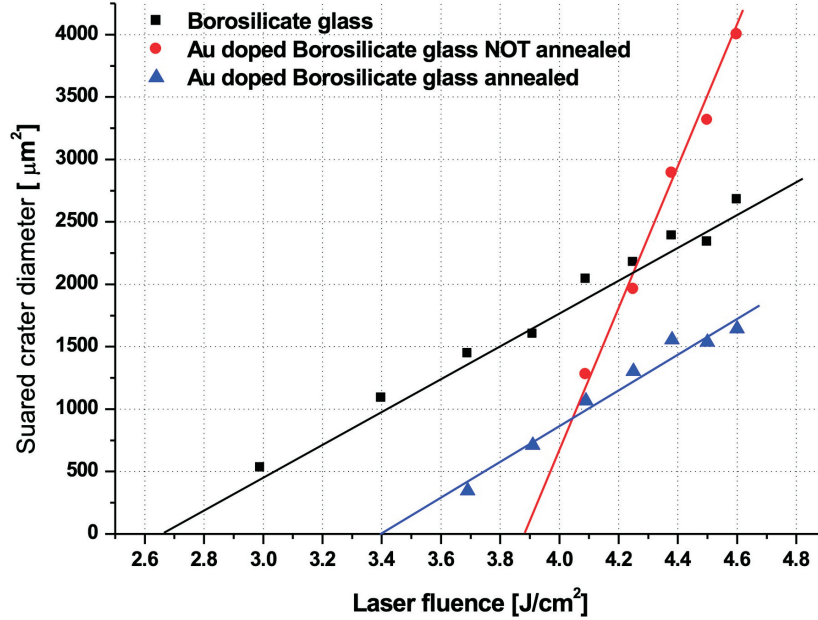


Fig. 4. Graphical representation of the squared crater diameter versus laser fluence (logarithmic scale). The black line corresponds to the native borosilicate glass sample; the red line corresponds to Au-doped sample, and the blue line corresponds to the Au-doped and annealed glass sample

formation of nanoparticle clusters occurs during laser irradiation and only in the processed zone. In other words, in the case of the native sample, less energy is absorbed compared to the cases of Au-doped annealed and not annealed samples as demonstrated from the observation presented in Fig. 4.

Conclusions. In the present study, a detailed analysis of laser-induced damage mechanism in borosilicate glass doped with Au nanoparticles, which is of key interest for fundamental investigations in the framework of non-linear optics, nanostructure generation, bioimaging, 5D data storage, and femtosecond laser micromachining, is performed.

The damage characteristics of borosilicate glass samples are investigated by using a femtosecond laser system with a pulse duration of 35 fs and a central wavelength of 800 nm. The laser-induced damage threshold of the glass is systematically studied by varying the applied laser energy. The results show that the increase (annealed and not annealed case) and presence of gold nanoparticles can effectively decrease the laser-induced damage threshold in the borosilicate glass. It is found that all the samples have a similar decreasing tendency on LIDTs at wavelengths of 800 nm. It is observed that the effect of the number of the laser pulses on the damage threshold follows the expected defect accumulation model.

This investigation is of particular importance in the context of the fundamental investigations in the framework of non-linear optics and nanostructure generation, for which better control of the material excitation is required. The main findings highlight the crucial role of transient optical properties during the laser-matter interaction in the ablation regime and contribute to the design of dedicated user-defined temporal excitation profiles and their applications by improving the requirements for glass quality and polishing, resulting in higher damage resistance.

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