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Ultrafast time-resolved photography of femtosecond laser induced modifications in BK7 glass and fused silica

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ABSTRACT The dynamics of absorption after excitation of fused silica and BK7 glass with femtosecond laser radiation are visualized by transient absorption spectroscopy. Focusing laser radiation with pulse durations in the picosecond time regime in BK7 glass generates free electrons with relaxation by emission of radiation or by formation of defects. The temporal and spatial emission characteristics are observed by high-speed photography in the streak mode. The beam waist moves within the pulse duration towards the incoming laser radiation by self-focusing and with the laser radiation absorbed by multi-photon processes. The dynamics of the long lasting stress formation is visualized by time-resolved Nomarski-Photography. The modification of the glass is investigated during and after irradiation with ultra-short pulsed laser radiation ($100\,\text{fs} < t_\text{p} < 3\,\text{ps}$) at the wavelength $\lambda = 810\,\text{nm}$. The formation of a sound wave in fused silica and BK7 glass is observed and the mechanical stress, depending on the excitation pulse duration, is measured.

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1 Introduction

Puzzling effects of optical breakdown, avalanche ionization, multi-photon ionization or laser-induced color center formation occupy research until now [1, 2]. The interaction of laser radiation with dielectrics can be subdivided into three steps: absorption, optical emission and modification.

The phase shift and the absorption coefficient of MgO, SiO_2, and diamond have been measured by time-resolved pump&probe in order to understand the processes in the photon-matter-interaction during and after irradiation with pulsed laser radiation in the femtosecond regime [3, 4].

The formation of color centers has been investigated with different types of radiation. Irradiation of insulators like alkali halides results in free electrons and holes with subsequent recombination to self trapped excitons (STEs). The recombination takes place by formation of non-bridging oxygen hole centers (NBOHCs), which decay to STEs by emission of 2 eV photons [5]. The recombination process to STEs of quartz is much faster than of the alkali halides showing at low temperatures a strong luminescence at 2.8 eV [6, 7].

The electron-hole formation process has been investigated by transient absorption spectroscopy after excitation of BK7 glass and fused silica with pulsed laser radiation ($t_\text{p} = 80\,\text{fs}$, $\lambda = 810\,\text{nm}$, $f_\text{p} = 1\,\text{kHz}$).

The temporal and spatial evolution of the optical emission after excitation with pulsed laser radiation ($t_\text{p} = 40\,\text{ps}$, $\lambda = 1064\,\text{nm}$, $f_\text{p} = 500\,\text{Hz}$) has been investigated for BK7 glass by high-speed photography. The spatial distribution of the emission can be described by a multi-photon absorption process.

The modifications like cracking and refractive index changes have been investigated by time-resolved Nomarski-photography via the pump&probe method within two different excitation time regimes ($t_\text{p} = 80\,\text{fs}$ and 3 ps, $\lambda = 810\,\text{nm}$). Refractive index changes and cracking in fused silica and BK7 glass have been observed from the femtosecond to the nanosecond time regime.

2 Laser and measurement tools

The investigations on absorption and modification in different dielectrics are performed using a femtosecond laser system with a repetition rate $f_\text{p} = 1\,\text{kHz}$, wavelength $\lambda = 810\,\text{nm}$, 25 nm spectral bandwidth (FWHM), and a pulse duration t_p in the range 70 fs – 3 ps by positively chirping the spectral phase of the pulse. The laser system has a nearly Gaussian beam of $M^2 = 1.5$.

Laser radiation of a picosecond laser system with a repetition rate $f_\text{p} = 500\,\text{Hz}$, wavelength $\lambda = 1064\,\text{nm}$ and pulse duration $t_\text{p} = 40\,\text{ps}$ has been applied for the investigation on emission. The laser radiation with a beam quality of $M^2 = 2.2$ was focused by a microscope objective (NA 0.4) within the substrate 500 μm below the surface.

The initial process of electron excitation by femtosecond laser radiation in fused silica and BK7 glass has been observed by transient absorption spectroscopy (TAS).

Using Nomarski-Photography the heating of the glass and the expansion of a sound wave has been observed. The setup for pump&probe-Nomarski and transient absorption spectroscopy with a variable delay ($t = 0$–$120\,\text{ns}$) are described elsewhere [8–10]. The output beam is divided into two beams by a separator. The exciting beam is focused by a microscope

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objective (NA 0.55) within the substrate 300 μm below the surface.

The spatial- and time-dependent evolution of the optical emission of BK7 glass irradiated by picosecond laser radiation has been measured by high-speed photography in the streak mode operation with a CCD camera (Imacon 500, Hadland Photonics) [8]. The used streak time was about 500 ns (time resolution 10 ps, spatial resolution 10 μm).

3 Results and discussion

3.1 *Absorption*

Within the interaction zone electron-hole-pairs are generated, which can recombine radiatively or by charge exchange and ion transport forming self-trapped excitons (STEs). STEs can also recombine by emission of photons. Depending on the electron relaxation time of the material a dense plasma is formed subsequently.

By exciting fused silica an absorption band from the UV to VIS can be detected, and the absorption band width persists for only a few ps. After 8 ps no absorption band can be detected anymore. As shown in [8] the duration of absorption can be calculated for the absorption of fused silica by evaluating the FWHM width of the absorption band. The absorption duration τ_{abs} shows no dependence of the wavelength (Fig. 1) and is attributed to free electron and hole formation [11]. These free electrons and holes are formed within a few femtoseconds, which then recombine or form STEs.

After 2.5 ps an absorption band from UV to IR can be registered for BK7 glass [9]. Contrary to fused silica BK7 glass is a multicomponent glass and the absorption takes place over 4 nanoseconds, decaying with exponential behavior [10]. After 7 ns no absorption is detectable anymore. Possibly the initial formed electrons relax quickly to STEs, which then relax through different channels slowly, e.g., by the formation of non-bridging oxygen holes (NBOH).

3.2 *Emission*

The spatial intensity, describing the emission along the beam propagation has a bell-like spatial distribution

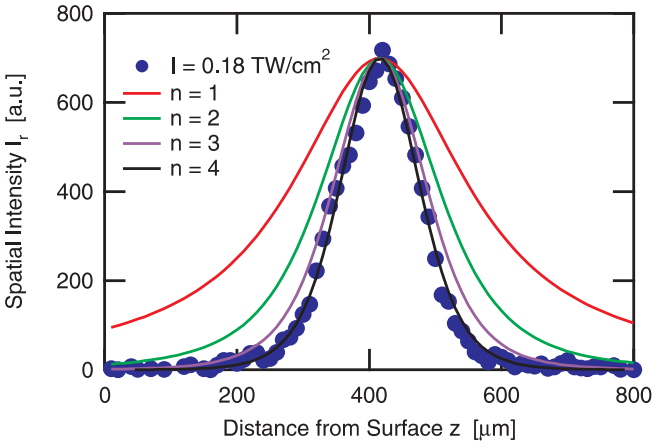

FIGURE 2 Spatial Intensity along the beam propagation for different multi-photon coefficients in BK7 glass ($t_p = 40$ ps)

(Fig. 2). The optical emission is a consequence of the absorbed energy in the glass. The distribution of the spatial intensity along the propagation of the laser radiation can be calculated by

$$I(z) = I_0 \left(\frac{1}{1 + \left(\frac{z}{z_R}\right)^2} \right)^n = I_0 \left(\frac{1}{1 + \left(\frac{z\lambda}{w^2 M^2 \pi}\right)^2} \right)^n , \qquad (1)$$

with z_R the Rayleigh length, w the beam waist, M^2 the beam quality, λ the central laser wavelength and n the multi-photon coefficient. BK7 glass has a band gap of about 4 eV and the IR-laser radiation ($\lambda = 1064$ nm) is absorbed by a 4-photon process and the relaxation has been detected by optical emission.

Because of the Kerr-Effect the position of the maximum of the emission

$$z_{I_{max}} = \frac{\pi^2 R w^4}{\pi^2 w^4 + R^2 \lambda^2 - \frac{P}{P_c} R^2 \lambda^2} \qquad (2)$$

changes with the laser pulse peak power (Fig. 3) as derived from the self-focusing theory [12] with R the phase curvature of the radiation and P_c the critical power for self-focusing.

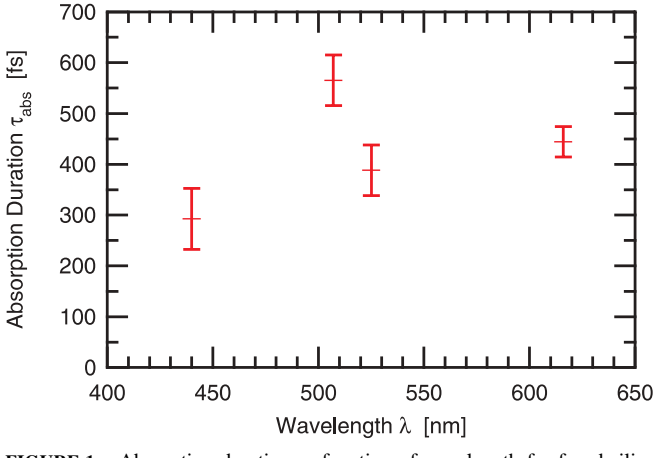

FIGURE 1 Absorption duration as function of wavelength for fused silica ($\lambda = 810$ nm, $t_p = 80$ fs, $I = 10\,\mathrm{PW/cm^2}$)

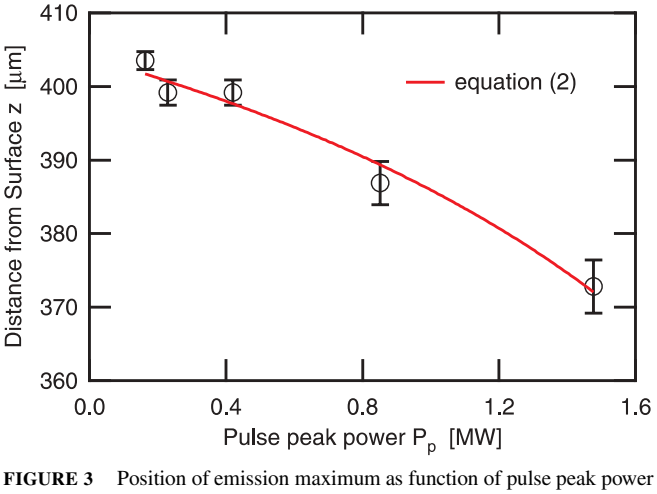

FIGURE 3 Position of emission maximum as function of pulse peak power in BK7 glass ($\lambda = 1064$ nm, $t_p = 40$ ps)

The adaptation of (2) to the measured values results in a critical power $P_c^{mea} = (3.9 \pm 0.2)$ MW, which coincides with the calculated one $P_c = c\lambda^2/(32n_2\pi)$ with the non-linear refractive index $n_2 = 2.8 \times 10^{-20}\,m^2/W$.

3.3 *Modification*

BK7 glass and fused silica have been excited with laser radiation at fixed pulse energy ($E = 6\,\mu J$) and the position and the amplitude of the sound wave can be measured [8]. Strong cracking is observed for both the materials in this excitation regime. The sound wave propagates with sound velocity (about 6 km/s) cylindrically to the beam propagation direction. The amplitude of the sound wave decreases with increasing radial distance from the irradiated zone (Fig. 4).

The mechanical stress, measured as the amplitude of the sound wave, is for both the glasses larger when excited by picosecond laser radiation. Free electrons are generated by multi-photon and field-effect ionization. Using femtosecond laser radiation the intensity is 40 times greater and a larger number of free-electrons are generated. After the femtosecond excitation the free electrons heat up the glass and dissipate out of the interaction zone. Different to femtosecond laser radiation the optical energy of the picosecond laser radiation is absorbed thereupon by the free electrons increasing the kinetic energy of this free electrons and initializing an avalanche ionization. More optical energy is absorbed and a much higher electron concentration in the interaction zone can be reached which consequently heats up the glass.

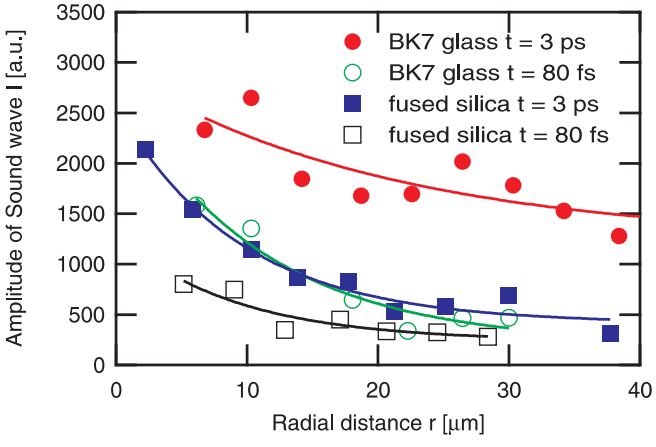

FIGURE 4 Amplitude of sound wave as function of radial distance ($\lambda = 810$ nm)

The amplitudes of the sound waves in fused silica are smaller than in BK7 glass and results from a smaller free electron concentration in fused silica after irradiation. This is due to a larger band gap and a smaller relaxation time of the free electrons.

4 Conclusion

The ultra-fast pump&probe investigations of BK7 glass and fused silica showed many similarities because of the common presence of SiO_2 in the materials investigated. But, of course, there are strong differences, especially between the chemical pure fused silica and BK7 glass: Transient absorption spectroscopy excited by a 80 fs laser pulse shows for fused silica the formation of a spectrally broad (> 400 nm) absorption band, which decays within about 300 fs. BK7 glass exhibits also a broad absorption band, which persists over 5 nanoseconds.

The optical emission of excited BK7 glass is driven by four-photon absorption. The beam waist changes the distance to the surface towards the incident radiation with changing pulse peak power and can be described by the self-focusing theory.

The development of sound waves in BK7 glass and fused silica is measured. The sound waves have a velocity of about 6.45 km/s and are expanding cylindrically and perpendicular to the beam propagation. The mechanical load of excited fused silica and BK7 glass is smaller for irradiation with femtosecond laser pulses ($t_p = 80$ fs) than with picosecond laser pulses ($t_p = 3$ ps).

REFERENCES

1 R.R. Alfano: *Supercontinuum Laser Sources* (Springer-Verlag, Berlin Heidelberg 1989)
2 R.R. Alfano, S.L. Shapiro: Phys. Rev. Lett. **24**, 592 (1970)
3 P. Martin, S. Guizard, Ph. Daguzan, G. Petite, P. D'Oliveira, P. Meynadier, M. Perdrix: Phys. Rev. B **55**, 5799 (1997)
4 F. Quere, S. Guizard, Ph. Martin, G. Petite, O. Gobert, P. Meynadier, M. Perdrix: Proc. SPIE **3578**, 10 (1998)
5 P.N. Saeta, B.I. Greene: Phys. Rev. Lett. **70**, 3588 (1993)
6 W. Joosen, S. Guizard, P. Martin, P. Agostini, A. Dos Santos, G. Grillon, D. Hulin, A. Migues, A. Antonetti: Appl. Phys. Lett. **61**(19), 2260 (1992)
7 S. Guizard, M. Meynardier: J. Phys.: Condens. Matter **8**, 1281 (1996)
8 A. Horn, H. Khajehnouri E.W. Kreutz, R. Poprawe: Proc. LIA, ICALEO 2002 **94**, 1577 (2002)
9 A. Horn, H. Khajehnouri, E.W. Kreutz, R. Poprawe: Proc. SPIE, HSPP2002 **4948**, 393 (2002)
10 A. Horn: PhD thesis (RWTH-Aachen 2003)
11 G. Petite, P. Daguzan, S. Guizard, P. Martin: Appl. Surf. Sci. **109&110**, 36 (1997)
12 J.H. Marburger: Prog. Quantum Electron. **4**, 35 (1975)