Time-Resolved Observation of Energy Deposition in Fused Silica by Ultrashort Laser Pulses in Single and Cumulative Regime

Cyril Mauclair, Konstantin Mishchik, Alexandre Mermillod-Blondin, Jörn Bonse, Arkadi Rosenfeld, Ingolf-Volker Hertel, Eric Audouard, Razvan Stoian

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Cyril MAUCLAIR[1,2], Konstantin MISHCHIK[1], Alexandre MERMILLOD-BLONDIN[2], , Jörn BONSE[2], Arkadi ROSENFELD[2], Ingolf V. HERTEL[2,3], Eric AUDOUARD[1] and Razvan STOIAN[1]

[1] Laboratoire Hubert Curien (UMR 5516 CNRS), Université de Lyon, 18 rue Pr. B. Lauras,42000 Saint-Etienne, France
[2] Max-Born-Institut, Max-Born-Str. 2A, D-12489 Berlin, Germany Stahnsdorf
[3] Freie Universität Berlin Fachbereich Physik, Arnimallee 14, 14195 Berlin, Germany
cyril.mauclair@gmail.com

Abstract: When femtosecond laser pulses are focused in the bulk of transparent materials (glasses), deposition of energy on a restricted volume can occur owing to the non linear character of the laser matter interaction. As a consequence, the possibility to generate micrometer-sized structural modifications arises. Those local changes are often associated with a minute variation in the refractive index which, when positive, enables the fabrication of light guiding components in three dimensions through simple laser translation. Although the first corresponding experimental demonstration approaches fifteen years of age, the complete picture of the dynamics and the processes leading to the local refractive index changes has still to be drawn to reach an optimal control of the laser-induced modification process. In this report, the laser-dielectric interaction is followed on an ultrashort time scale with the help of a unique time-resolved side-imaging technique allowing for absorption and phase contrast detection. Experimental observation of an absorptive electronic cloud in the first moments of the interaction along with the launch of a pressure wave after a few ns is reported. These physical objects are shown to be reliable indicators of the success of the energy transfer to the lattice which largely depends on the pulse temporal envelope.

OCIS codes: (320.7130) Ultrafast processes in condensed matter; (070.7145) Ultrafast processing

1. Introduction

The technique of ultrafast laser processing of bulk transparent materials knows a growing importance as reported by the numerous demonstrations of various photonic devices achieved in its frame [1]. Micrometer sized lattice modification are reachable due to the highly non linear nature of the interaction between the infrared ultrashort pulses and t he transparent material. Local optical properties are thus influenced by the femtosecond irradiation, resulting in possible variations of birefringence, absorption and refractive index [2]. As a consequence, optical components can be photodrawn in three dimensions by translation of the laser spot in the bulk of the sample [3].

From a fundamental point of view, a better understanding of the successive steps conducting to the permanent bulk modification is obviously an indispensable study to be carried out in order to exploit the full potential of femtosecond bulk processing. In the following, microscopic imaging of the material transient states under femtosecond irradiation on a ultrafast time scale is reported.

2. Experimental Methods

The experimental apparatus is based on a noncollinear pump-probe technique [4]. The radiation source is an ultrafast laser system delivering 160 fs pulses at 800 nm. The laser beam is divided into two parts, one being used to excite the material, while the second is spatially magnified, frequency doubled, and used to image the excitation region in a perpendicular geometry with a time resolution of 0.6 ps.

A microscope objective (numerical aperture NA = 0.45) focuses the pump beam into the bulk of the samples, which are polished parallelepipeds of 3 × 20 × 10 mm. The focal plane is fixed at a depth of 200 μm. The blue probe beam enters the illumination path of a modifie d phase contrast microscope (Olympus BX41). The final picture is recorded with the help of an intensifying CCD camera. This setup allows for ultrafast imaging in optical transmission microscopy (OTM) mode or in phase contrast microscopy (PCM).

3. Results and Discussion

PCM pictures of permanent bulk modifications subsequent to laser irradiations in fused silica are presented in Fig. 1.

In all cases, the pulse energy is 0.4µJ inside the material and the laser comes from left. However, the number of pulses per sites varies as well as scanning direction of the sample. As already reported, the high accumulation of light pulses leads to dramatic evolution of the modified area in fused silica accompanied with an increase of the affected volume [15]. Upon longitudinal translation of the sample guiding structures are readily achievable.

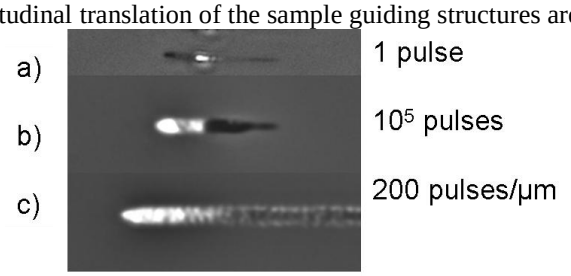

Fig. 1 : Permanent modifications in fused silica from femtosecond irradiation.
The laser comes from left. a) Single pulse, b) 10^5 pulses at 100kHz
c) Scanning of the sample along the laser propagation axis at 100kHz.

Examples of time resolved investigations at high energy are reported in Fig. 2. The strong electronic cloud clearly visible in Fig. 2 a) extents to a much larger region than the permanent damage. Interestingly enough, the absorption peaks are closely correlated to the presence of permanent changes (Fig. 2b)). Regular dots preceding the main focal point are also observable. Our first investigations permits to think that these modifications are simply related to the irradiation focal map according to simple Fourier propagation simulations (not shown here).

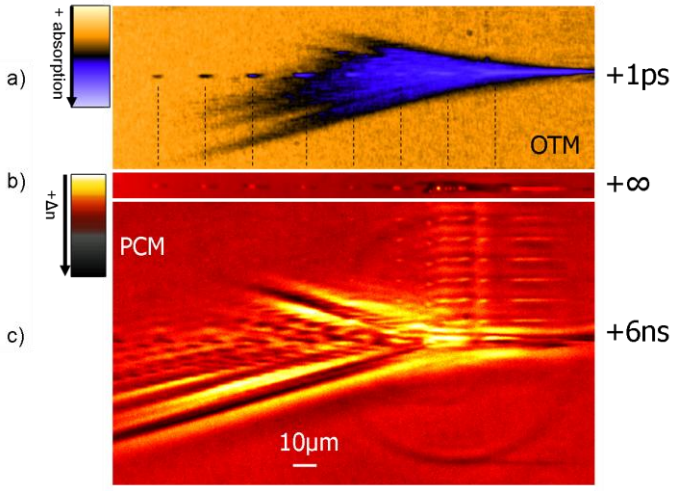

Fig. 2 : PCM and OTM time resolved pictures of ultrafast irradiation of fused silica.
The pulse energy is 43µJ and the laser comes from left.

For time delays in the order of the nanosecond, the launch of a relatively strong pressure wave takes place and requires the phase contrast imaging to be detectable.

4. Conclusion

The electronic cloud along with the launch of the pressure wave are pictured on a ultrafast time scale. At high intensities, regular structures preceding the main focus are observed presumably linked to the focal irradiation map as predictable from Fourier propagation simulations. Further investigations concerning the non linear effect are envisaged.

5. References

[1] K. Itoh, W. Watanabe, S. Nolte, and C. Schaffer: MRS Bull, 31, (2006) 620–625.

[2] E. Bricchi, B. G. Klappauf, and P. G. Kazansky: Opt. Lett., 29, (2004) 119–121

[3] S. Nolte, M. Will, J. Burghoff and A. Tuenner-mann: Appl. Phys. A, 77, (2003) 109-111.

[4] A. Mermillod-Blondin, "Analysis and optimization of ultrafast laser-induced bulk modifications in di-electric materials," Ph.D. Thesis Freie Universitat Berlin (2006).

Ultrafast

1. Introduction

Ultrafast laser

pulses and the transparent material enables local modification on the micrometer scale. The related structural changes are usually accompanied by alteration in the optical properties such as birefringence, absorption and refractive index [2]. The size of the local change is usually on the order of the focal volume, enabling the photodrawing of complex photonic devices by simple translation of the laser spot with respect to the sample [3].

Efforts to precisely control irradiation conditions are continuously conducted aiming at improvements of the achievable machining flexibility by femtosecond lasers. Techniques based on a precise tuning of the laser repetition rate, laser pulse duration, polarization and wave front structure recently emerged, thus greatly increasing the dimensions of processing windows [4-8].

From a more fundamental point of view, it is highly desirable to obtain a better understanding of the intermediate steps that lead to the permanent changes in the optical properties subsequent to the laser irradiation, thus allowing for strategies to enhance the flexibility in femtosecond machining of transparent materials. This type of research is tightly linked to the capability of monitoring transient local states that the material undergoes from the very first moments of interaction until the final and permanent local modification. During this time window, essential transient physical objects such as the laser-generated electron-hole plasma and propagating pressure waves are observable [9-11]. Their characteristics are of great interest in understanding the permanent laser-induced modifications.

In the following, we report on time–resolved imaging of local modification subsequent to ultrashort light pulse irradiation in transparent materials. Transient electron-hole plasma pictures are shown as well as pressure waves. Essential differences between femtosecond and picosecond irradiation regimes are underlined.

2. Experimental set-up

The experimental apparatus is based on a noncollinear pump-probe technique [12] depicted in Figure 1. The radiation source is an ultrafast laser system delivering 160 fs pulses at 800 nm operated at an effective repetition rate of 2Hz. The laser beam is divided into two parts, one being used to excite the material, while the second is spatially magnified, frequency doubled, and used to image the excitation region in a perpendicular geometry with a time resolution of 0.6 ps.

A microscope objective (numerical aperture NA = 0.45) focuses the pump beam into the bulk of the samples, which are polished parallelepipeds of 3 × 20 × 10 mm. The focal plane is fixed at a depth of 200 µm from the surface, minimizing the effect of spherical aberration generated at the air-dielectric interface. The blue probe beam enters the illumination path of a modified phase contrast microscope (Olympus BX41). Two scattering elements ensure a uniform illumination of the collector. The final picture is recorded with the help of an intensifying CCD camera.

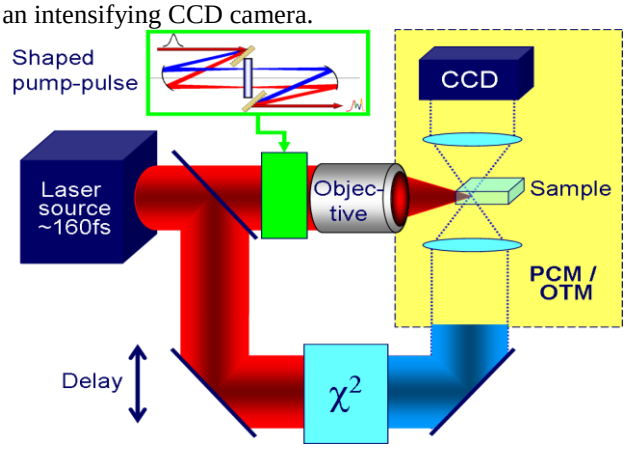

Fig. 1 Scheme of the experimental set up; PCM: Phase contrast microscopy, OTM: Optical transmission microscopy, χ^2: Frequency doubling cristal.

A bandpass filter centered around 400 nm is placed in front of the camera, rejecting parasite light emitted by the irradiated specimen (electronic plasma emission) and pump light scattered from the modified region. This setup allows for ultrafast imaging in optical transmission microscopy (OTM) mode or in phase contrast microscopy (PCM) mode by simple rotation of the phase contrast module. The onset of the free carrier absorption is used to define the temporal synchronization. Speckle noise generated by the employment of scattering plates was reduced by mounting one scattering plate on a rotating motor and by accumulating N times the image of the same event on the CCD matrix. In OTM, we chose N = 50 and in PCM, N = 100. The sample was translated after each laser shot.

A linear spatial light modulator is inserted into a 4-f zero compressor after the beam splitter enabling the generation of user defined, shaped pulses on the pump arm while preserving the higher available temporal resolution for the probe.

3. Results and discussion

Figure 2 presents phase contrast pictures of single ultra-short pulse irradiation in fused silica glass at 4.3 µJ for various pulse durations (laser comes from left).

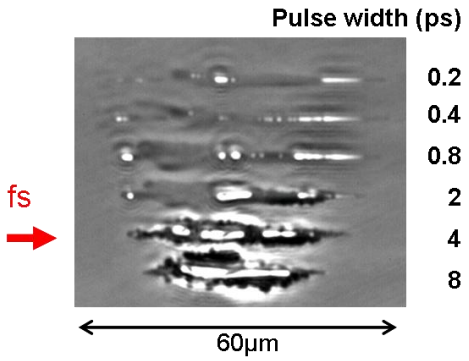

Fig. 2 Single pulse femtosecond irradiation in fused silica glass

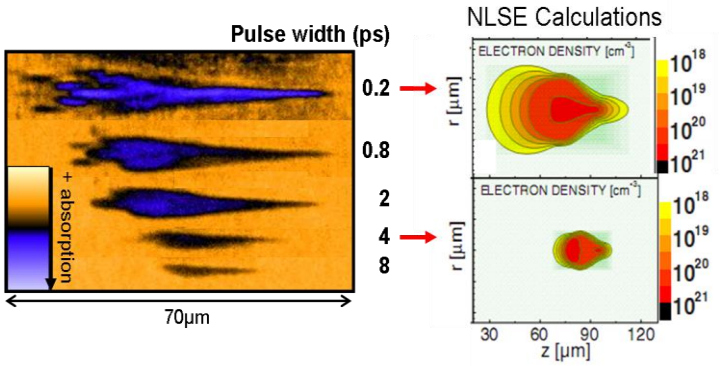

Fig. 3 Pump probe OTM pictures for 2 ps delay for various pump durations with NLSE predictions of the focal electronic densities for 0.2 ps and 4 ps pulse duration.

This picture represents the permanent optical change as it was taken a few seconds after irradiation. In the case of femtosecond single pulses sequence, the modification consists of alternating black and white regions, mainly due to positive and negative refractive index change (positive phase contrast microscopy). The length of the structures may be as large as 60 µm while remaining quite thin (<5 µm). For longer pulses, the modified area diminishes in length while increasing in width. Picosecond pulses clearly create a higher contrast and stronger damage. These observations give evidence for a more confined area of energy deposition in the case of picosecond irradiation.

The consequences of a ps envelope are twofold [14]. Firstly, the ps envelope induces a delayed, low density, spatially-modulated plasma. This reduces intensity dependent changes of the refractive index, thus creating a smaller negative shift for the incoming energy and less defocusing, helping to contain the energy in the focal region. Secondly, the nonlinearity of excitation is reduced, allowing efficient absorption only in a restricted region around the geometric focal point.

In order to experimentally verify these considerations, OTM time-resolved pictures were taken at a delay of 2 ps after irradiation, when the electron-hole plasma appears to be the strongest. This experimental observation was conducted for different pump pulse duration between 0.2 ps and 8 ps. Figure 3 shows the corresponding data (laser from left)

One can readily see that the extension of the electronic plasma is much more constricted in the picosecond regime. Calculation results of the electronic densities based on the nonlinear Schrödinger equation are presented in Fig. 3 for 0.2 ps and 4 ps pulse duration. Details of the simulation approach are given in reference [13]. It is sufficient for the present discussion to mention that key features taken into account by the calculations are photoionization, plasma defocusing, self-focusing, diffraction and multiphoton and avalanche ionization.

The simulated electronic density extends on an area approximately two to three times smaller in the picosecond regime than in the femtosecond regime, a behavior confirmed by the experimental images of the electronic cloud for different pulse durations. This experimental evidence of the effect of the pulse envelope on the transient electron-hole plasma has direct consequences on the choice of laser parameters for material bulk processing.

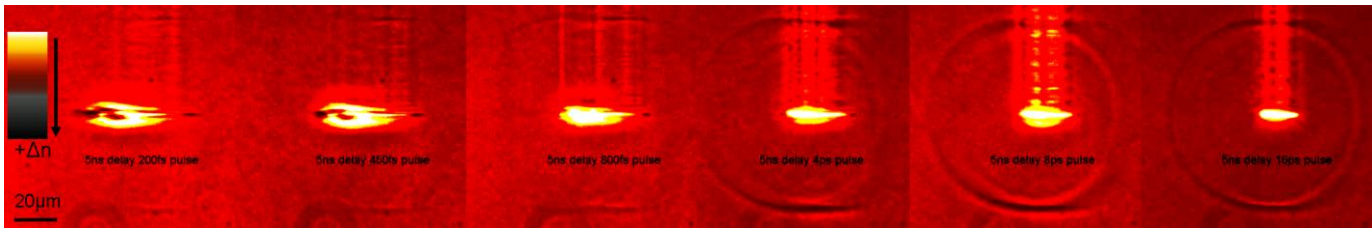

Fig. 4 PCM time resolved pictures at 5 ns delay for various pulse durations

Along with the electronic cloud, the pressure wave generated after a few nanosecond carries information of great value. Figure 4 depicts PCM time resolved pictures 5ns after irradiation for different pulse durations (laser comes from left). PCM is well adapted for this observation as it is able to transcribe minute refractive index variation into well contrasted grayscale images.

The launch of a pressure wave after 2 ns is readily observable for all pump pulse durations. We mention here that the travelling speed of this perturbation matches closely the speed of sound in fused silica [15].

What appears to be obvious is the amplitude dependence of the pressure wave with the pump pulse duration. For picosecond pulses the pressure wave is much more contrasted as for the femtosecond regime (Fig. 4). This also speaks in favor of a stronger confinement of the energy deposition leading to a stronger pressure wave. The more damaged aspect of the permanent modification for picosecond pulses appears to be linked with the amplitude of the released pressure wave.

4. Conclusion

Time resolved side imaging of transient local changes in fused silica subsequent to ultrashort pulse irradiation is reported. A pump probe set-up enabled the monitoring of significant physical objects such as the laser-induced electron-hole plasma and the pressure wave generated from the processing volume. Both give reliable indicators towards a better understanding of the physical phenomenon active during the laser-matter interaction. Experiments were conducted to determine differences between picosecond and femtosecond irradiation regimes. A more confined electronic plasma, along with a stronger pressure wave are characteristic of the picosecond domain, which is confirmed my numerical simulations based on the nonlinear Schrödinger equation. Clear experimental evidence of a higher energy confinement in the case of picosecond irradiations is given.

Acknowledgments

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References

[1] K. Itoh, W. Watanabe, S. Nolte, and C. Schaffer: MRS Bull, 31, (2006) 620–625.

[2] E. Bricchi, B. G. Klappauf, and P. G. Kazansky: Opt. Lett., 29, (2004) 119–121.

[3] S. Nolte, M. Will, J. Burghoff and A. Tuennermann: Appl. Phys. A, 77, (2003) 109-111.

[4] S. M. Eaton, H. Zhang, M. L. Ng, J. Li,W. Chen, S. Ho, and P. R. Herman : Opt. Express, 16, (2008), 9443–9458.

[5] D. J. Little, M. Ams, P. Dekker, G. D. Marshall, J. M. Dawes, and M. J. Withford: Opt. Express, 16, (2008) 20029–20037.

[6] C. Mauclair, A. Mermillod-Blondin, N. Huot, E. Audouard, and R. Stoian: Opt. Express, 16, (2008) 5481-5492.

[7] A. Mermillod-Blondin, C. Mauclair, A. Rosenfeld, J. Bonse, I. V. Hertel, E. Audouard, and R. Stoian: Appl. Phys. Lett., 93, (2008) 021921–021924.

[8] C. Mauclair, G. Cheng, N. Huot, E. Audouard, A. Rosenfeld, I. V. Hertel, and R. Stoian: Opt. Express, 17, (2009) 3531-3542.

[9] S. Juodkazis, K. Nishimura, S. Tanaka, H. Misawa, E. Gamaly, B. Luther-Davies, L. Hallo, P. Nicolai, and V. Tikhonchuk: Phys. Rev. Lett., 96, (2006) 166101.

[10] A. Salleo, S. T. Taylor, M. C. Martin, P. R. Wendy, R. Jeanloz, T. Sands, and F. Y. Genin: Nature Materials, 2, (2003) 796.

[11] J.-F. Lin, H. Fukui, D. Prendergast, T. Okuchi, Y. Q. Cai, N. Hiraoka, C.-S. Yoo, A. Trave, P. Eng, M. Y. Hu, et al: Phys. Rev. B 75, (2007) 012201.

[12] A. Mermillod-Blondin, J. Bonse, A. Rosenfeld, I. V. Hertel, Yu. P. Meshcheryakov, N. M. Bulgakova, E. Audouard, and R. Stoian: Appl. Phys. Lett., 94, (2009) 041911.

[13] A. Mermillod-Blondin, I. M. Burakov, Yu. P. Meshcheryakov, N. M. Bulgakova, E. Audouard, A. Rosenfeld, A. Husakou, I. V. Hertel, and R. Stoian: Phys. Rev. B 77, ((2008) 104205.

[14] I. M. Burakov, N. M. Bulgakova, R. Stoian, A. Mermillod-Blondin, E. Audouard, A. Rosenfeld, A. Husakou, and I. V. Hertel: J. Appl. Phys., 101, (2007) 043506.

[15] A. Mermillod-Blondin, "Analysis and optimization of ultrafast laser-induced bulk modifications in dielectric materials," Ph.D. Thesis Freie Universitat Berlin (2006).