Three-dimensional laser structuring of materials at tight focusing

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Pulse shaping and phase control of femtosecond pulses are shown to have effect on the dielectric breakdown of sapphire. The influence of a pulse tilt on a dielectric breakdown is demonstrated. Three-dimensional structuring of transparent dielectric and semiconducting materials at tight focusing is discussed.

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Three-dimensional (3D) laser microfabrication using femtosecond laser pulses has found an increasing number of applications in different fields: bit-like[1], holographic[2] and photo-refractive[3] optical memories, nano-/microfluidics[4], photonic crystals[5,6], waveguide recording, glass welding, nano-structuring of surfaces, and cutting and drilling[7]. Spatial resolution of 3D structuring with feature size of tens-of-molecules has been demonstrated by direct laser writing[8] and holographic recording[9]. The high-precision control over temporal and spatial light-intensity distribution is the key to fabrication with feature size smaller than 100 nm.

In this paper, we demonstrate that the pulse tilt and spherical aberration strongly affects the axial light localization and plays an important role in 3D structuring of materials by dielectric breakdown. Also, it is shown that control of relative phase of two co-axially introduced pulses provides an additional parameter which changes conditions of an in-bulk dielectric breakdown of dielectrics as demonstrated in the case of sapphire.

Femtosecond laser pulses from Hurricane and Spitfire (both from Spectra Physics) lasers with 150- and 70-fs duration, respectively, at central wavelength of 800 nm were used in experiments. Temporal pulse shaping was made by Dazzler (Fastlite). Two pulses have been formed by Dazzler out of a single 70-fs pulse (Tsunami) and amplified by Spitfire. The amplitudes of two pulses were equal with controllable temporal separation. The phase of second pulse was set separately. The light-induced damage threshold (LIDT) of dielectric samples was determined by a method introduced earlier[10]. Samples used in this study were viosil silica, sapphire, and rutile-TiO_2. Pulse energy is given at the entrance of the microscope if it is not specified otherwise. Transmission of the objective lens of numerical aperture (NA) of 1.35 was 58% at 800-nm wavelength and additionally 60% losses occurred from the entrance to the objective lens (mainly due to clipping of the laser beam by an entrance pupil of the objective lens). Beam diameter was measured by a knife-edge method using a fit of the measured laser power, P, by a function $P(x)/P_0 = 0.5\mathrm{erfc}(\sqrt{2}x/\omega_0)$, where x is the position of the knife edge ($x = 0$ corresponds to the center of beam), ω_0 is the e^{-2}-spot-radius, and erfc is the error function. Pulse duration was measured by a grating-eliminated no-nonsense observation of ultrafast incident laser light E-fields (GRENOUILLE, Swamp Optics) method and numerically retrieved via frequency-resolved optical gating (FROG) algorithm[11]. The tilt of femtosecond pulses (Hurricane, Spectra Physics) was measured by a specially designed auto-correlator (Light Conversion, Ltd.)[12].

In order to achieve a well controlled 3D structuring of materials, it is necessary to control light-intensity distribution at the focus (the point spread function (PSF)) with high fidelity. The axial extent of the PSF is larger than the lateral one even for a tight aberration-free focusing according to Ref. [13]:

$$l = d\frac{\sqrt{3 - 2\cos\alpha - \cos 2\alpha}}{1 - \cos\alpha}, \qquad (1)$$

where the lateral diameter of the focal spot size is defined by the Airy disk for a plane-wave focusing, $d = 1.22\lambda/\mathrm{NA}$; here, the numerical aperture $\mathrm{NA} = n\sin\alpha$ with n being the refractive index at the focus, λ the wavelength in vacuum, and α the half-angle of a focusing cone. Equation (1) is valid for a large numerical aperture $\mathrm{NA} > 0.7$ focusing typically used in 3D laser microstructuring. The spherical aberration additionally elongates the focal spot size as observed at similar irradiation conditions in different materials[14]. The other effect which also can cause an axial smearing of light intensity is the pulse tilt[12]. Its influence to the focal-intensity distribution was investigated.

Whenever a femtosecond laser pulse passes throughout a prism or is reflected by a grating, the pulse front tilt occurs. Femtosecond laser pulses obtained via chirped pulse amplification (CPA) method can have a significant tilt if compression, usually made by a pair of gratings, is not optimized[12]. We explore here the effect of a pulse tilt on the dielectric breakdown of viosil silica glass.

Figure 1 shows experimental data of the light-induced damage threshold dependence on the pulse tilt. Prisms of BK7 glass were inserted into the laser beam to introduce tilt. Two prisms, an equal-sided triangular prism with 60° corners and a right-angle prism with 45° corners, were inserted into the beam in such a way that the minimum amount of glass occurred in the beam, i.e., just the corner of prism was inserted to accommodate the entire beam diameter. In this way the temporal spreading of the pulse due to group-velocity dispersion (GVD) in glass was minimized and the smallest LIDT could be determined.

The tilt of an output of Hurricane was measured in two perpendicular directions as shown in Fig. 1(a). The tilt

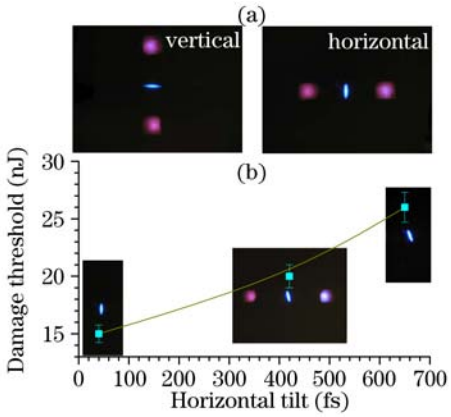

Fig. 1. (a) Tilt images of Hurricane (Spectra Physics) laser pulses (the central strip is a vectorial SHG at 400 nm of two time-synchronized 150-fs pulses at 800-nm fundamental wavelength). Vertical trace was taken with a $\lambda/2$-plate inserted into laser beam for a polarization rotation by a $\pi/2$ angle. (b) Light-induced damage threshold of viosil silica at a 20-μm depth at different horizontal tilt values. Tilt was produced by inserting a BK7 glass prism into the beam. Experimental data are superimposed with images.

values were comparatively small, 35 ± 10 fs, for an 8-mm diameter beam and were digitized using an image processing with Matlab. For calibration, a 180-μm-thick cover glass ($n = 1.5$) was inserted into one arm of the auto-correlator and had introduced a shift of a second harmonic generation (SHG) signal, a middle feature in images shown in Fig. 1. Then, the tilt was measured with the inserted BK7 prism at the same conditions as the LIDTs were determined. Results are summarized in Fig. 1(b). When focused, the tilted pulses tend to spread axially at focus causing a correspondingly elongated PSF. As a result, a larger pulse energy is necessary to reach the LIDT irradiance. This explains the experimentally measured dependence shown in Fig. 1.

In order to minimize the effect of pulse tilt on 3D laser microstructuring, the diameter of laser pulse should be enlarged using a telescope. Then only a central part of the pulse is used for sample irradiation (the area clipped by the aperture of the objective lens) and proportionally smaller amount of the tilt is in the pulse. This helps to achieve PSF closer to the ideal one of a plane wave and to have the axial extent of PSF given by Eq. (1).

The spherical aberration is proportional to the focusing depth and refractive-indices mismatch between, in our case, an immersion oil ($n = 1.515$ at 632 nm) and medium where focus is located[14,15]. Figure 2 shows single and multi-pulse damage sites in rutile-TiO_2 made with tightly focused, NA= 1.35, 180-fs pulses of 800-nm wavelength (Hurricane, Spectra Physics). The spherical aberration is very strong in rutile since the refractive index $n = 2.5$ is among the highest of wide-bandgap semiconductors (the bangap energy $E_g = 3.1$ eV). In order to create a void-like damage in rutile (Fig. 2(a)) pulse energy was more than 100 times larger than that in silica or sapphire[14,16]. It is noteworthy that two-photon absorption of rutile could facilitate formation of a filament-like damage via Gauss-to-Bessel beam/pulse transformation[17−19]. This mechanism needs further detailed investigations. Control of a laser beam divergence can provide means to establish an almost spherical aberration-free PSF at the required depth in materials with different refractive indices[15].

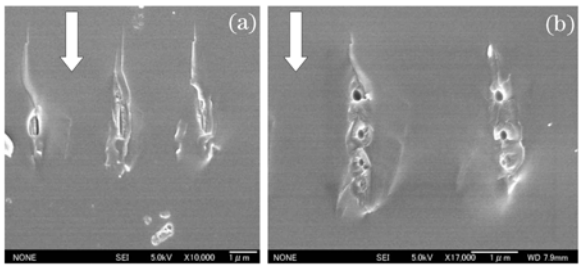

Fig. 2. Side-view SEM images of damage sites in rutile-TiO_2 made by (a) a single 1-μJ pulse and (b) 10 pulses of 0.3 μJ at a 10-μm depth; pulse duration 180 fs (estimated at the focus), wavelength 800 nm (Hurricane, Spectra Physics). Arrows mark the direction of the pulse propagation; the separation between irradiation sites was 3 μm.

As we reported earlier, a two-pulse damage of PMMA[20] and silica glass[21] showed complex dependence of the LIDT on the pulse separation when Mach-Zehnder interferometer was used to produce two cross-polarized pulses. Here, we report on the dependence of LIDT on pulse separation when two pulses of the same polarization were produced by pulse shaper Dazzler.

Figure 3 shows dependence of LIDT (in terms of pulse energy, E_p) versus separation between the two pulses in case of dielectric breakdown of sapphire with focal position just beneath the surface at 5−10 μm depth where PSF distortions due to the spherical aberration were minimal. The threshold of optically recognizable damage was measured for different pulse separations and phases. Despite of a considerable statistical noise there is a distinct difference in the LIDT values for two pulses with equal phases, $\phi = 0$, and when the phase of second pulse was shifted by $\pi/2$, at time separation smaller than 100 fs which is close to the pulse duration t_p. The physical reason for such threshold increase when $\phi = \pi/2$ as follows: the cumulative energy dose of two pulses calculated as an integral of intensity $D \propto \int (E_1(t,0) + E_2(t,\phi))^2 dt$ has maximum when $\phi = 0$ and is smaller for other ϕ values[22]. Hence, the cumulative dose D of two pulse is always smaller when $\phi \neq 0$, which means that larger

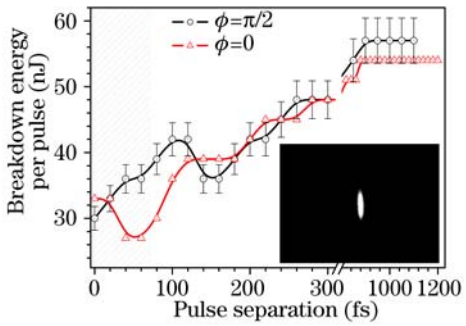

Fig. 3. Double-pulse energy, E_p, required for a dielectric breakdown of sapphire versus separation between the pulses (each having the same pulse energy, $E_p/2$) for two different phases ϕ. Pulse shaping has been performed by Dazzler. The pulse duration is marked by dashed region. Lines are drawn as eye guides. The inset shows a FROG trace (frequency versus time) of amplified pulses; the retrieved pulse duration was $t_p = 50 \pm 10$ fs (Spitfire, Spectra Physics).

pulse energy is necessary for the dielectric breakdown as observed experimentally (Fig. 3). It is noteworthy, that there should be no breakdown when two pulses are added up with the opposite, $\phi = \pi$, phases even if the energy of the pulses would be large. The cumulative intensity would be zero due to destructive interference. However, it is impossible to generate such a waveform which would be able to produce two π-phase-shifted pulses with temporal separation comparable with $t_{\rm p}$ in the Dazzler crystal.

It is noteworthy, that the pulse duration of approximately 90 fs corresponds well to typical phonon energies of 40−50 meV in sapphire[23]. Hence, there is a theoretical possibility to excite coherent phonons in sapphire. The experimentally observed oscillations on LIDT plot in Fig. 2 had a distinguishable period of approximately 100 fs, however, the uncertainties were too high to make a positive assignment that the oscillations are due to coherent phonons. Pulse shaping of femtosecond pulses using Dazzler or spatial light modulators provides a possibility to launch and probe propagation and decay of coherent phonons and it is expected to be focus of future experiments.

In conclusion, formation of a linear-like damage in dielectrics and semiconductors due to pulse the front tilt and spherical aberration have been demonstrated experimentally. The 3D recording of the axially elongated damage in rutile has been demonstrated by single and multi-pulse irradiation. The phase control of two fs-pulses allows to modify the cumulative pulse energy at which dielectric breakdown ensue. The phase control of two interfering pulses might be used to control the transmitted power of strongly focused ultra-short laser pulses. Possibility to excite coherent phonons by femtosecond pulses has been discussed.

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