Three-Dimensional Laser Microfabrication

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ABSTRACT
The mechanism of three-dimensional (3D) laser micro-structuring of dielectrics (resists, polymers, glasses, and crystals) by the direct laser writing is discussed. Overview of a light-matter interaction of a tightly-focused femtosecond pulse with dielectrics is presented. The void formation is demonstrated inside silica glass by a single picosecond pulse. Peculiarities of tight focusing and an axial superposition of two co-propagating pulses at the focus are revealed by numerical simulations. Potential of the fabricated 3D patterns in micro-photonics, micro-fluidics, and sensor applications is discussed.

Keywords: photo-structuring of glass, laser microfabrication, dielectric breakdown, shock wave formation, ionization, filamentation

1. INTRODUCTION
Three-dimensional (3D) laser micro-structuring of dielectric materials by short, sub-1 ps, pulses can be realized since a very high pulse irradiance (\(\sim 1 \text{ PW/cm}^2\)) can be obtained without self-focusing inside dielectrics. Hence, a direct laser write method can be realized, i.e., a photo-modification can be delivered at the aimed sub-1 \(\mu\text{m}^3\) volume.\(^1,2\) The aberration caused distortions of a focal light intensity distribution (the intensity point spread function), mostly by the spherical aberration, can be minimized by afocal focusing. The physical reason of photo-structural/physical/chemical modifications at the focal region are all related to the ionization. The ionization via nonlinear multi-photon and avalanche absorption is the key for description of different photo-modifications. Even a multiple ionization (up to 4 – 5 electrons) is achievable and plasma with electron densities \(\sim 10^{23} \text{ cm}^3\) can be achieved. The absorbed volumetric energy density within the ionized volume (\(\text{J/m}^3\)) is equivalent to the pressure (\(\text{N/m}^2\)), which can reach tens-of-TPa at the end of a femtosecond pulse. This pressure is many times larger than the Young modulus, hence, condition for a strong explosion are fulfilled. As a result, a shock wave is launched from the focus and affects surrounding regions where defects, new phases, and cracks can be formed. At the same time, the rarefaction wave forms the less dense center or even void. This multitude of possible photo-modifications is expected to be useful for number of applications in micro-optics, waveguiding, and micro-fluidics.

Here, we report on void formation by single femtosecond and picosecond laser pulses. Also, peculiarities of two-pulse irradiation and tight focusing are discussed and simulated.

2. EXPERIMENTAL DETAILS
We discuss here experimental results of structuring of dielectrics by tightly focused laser pulses using an objective lens of numerical aperture \(NA \geq 0.8\). The femtosecond, 200 fs (800 nm wavelength), and picosecond, 30 ps (532 nm wavelength), pulses were used for irradiation at the conditions of dielectric breakdown and void formation at the focus. More details on setup can be found in ref. \(^3\). Voids were formed in dielectrics (silica glasses, polymers, and crystalline sapphire).\(^1,2\)

Structural modifications were inspected by optical imaging and by the scanning and transmission electron microscopies (SEM and TEM). The correspondence between void formation and an optical contrast has been established. Samples of silica and sapphire were cleaved open along the pulse propagation path and were inspected by SEM. The bright center of the damage site in optical image signified the presence of the void.

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Figure 1. The normalized space-time map of intensity $|E(z, r, t')|^2$ at the focus: the intensity along z-axis of propagation at the focus for the 50 fs pulse of 800 nm central wavelength at different time moments (vertical axis). Contour lines are drawn in a 10% steps over the envelope. The confocal parameter $2z_R = 1431$ nm corresponds to the experimental conditions of focusing.

3. RESULTS AND DISCUSSION

We, first, discuss peculiarities of tight focusing when the slowly varying envelope approximation (SVEA) becomes invalid. Then, the intensity at the focus is modeled for a two pulse exposure with different delays and relative phase between the pulses. Also, the experimental results of void formation by picosecond laser pulses in low OH content silica are summarized. Finally, the recently proposed mechanisms of dielectric breakdown are discussed.

3.1. Light intensity at the focus

When two tightly focused laser pulses are aimed co-axially at the same focus there was an interesting phenomenon observed: the dielectric breakdown threshold was not the smallest when two pulses were temporally (as well as spatially) overlapped. The smallest threshold was observed when intra-pulse separation was approximately half of the pulse duration in silica$^4$ and PMMA$^5$ for two cross-polarized pulses. The pulses were overlapped using Mach-Zender interferometer. The peculiarities of tight focusing and numerical modeling of the spatio-temporal overlap of two-pulses are presented below.

3.1.1. Tight focusing: the space-time map

Here, the tight focusing of a Gaussian pulse is simulated. The slowly varying envelope approximation has not been used, since it is invalid at the tight focusing conditions by definition, i.e., when the electrical field strength is changing within the length comparable with the wavelength. The pulse Gaussian in space and time in a cylindrical coordinate system is given at the focus as:

$$E_G(z, r, t) = \frac{|E_0|}{1 - iz\lambda/(w_0^2n\pi)} \exp\left(-\frac{r^2}{w_0^2}\right) \exp\left(-2\ln2\frac{t^2}{\tau_p^2}\right),$$

where $i = \sqrt{-1}$, $\tau_p$ is pulse duration at FWHM, $w_0$ is the waist (radius), $\lambda$ is the vacuum wavelength, $E_0$ is the electric field amplitude, and $n$ is the refractive index. The oscillating E-field is then $E(z, r, t) = E_G(z, r, t) \times \text{Re}(\exp(i(\omega t - kz)))$, where $k = 2\pi/\lambda$ is the wavevector. By substitution $t' = t - z/c$, the expression of $E(z, r, t')$ can be used to reveal the space-time history of excitation. Figure 1 shows the axial cross-section of intensity at
focus at different time moments, the space-time map. Here, the waist of the beam and the Rayleigh length are calculated as diffraction limit for a plane wave focusing according to

$$w_0 = \frac{0.61 \lambda}{NA}$$

and

$$z_R = \frac{n \pi w_0^2}{\lambda}.$$  

The central part of the focal region, approximately $\lambda/2$ in length is enclosed inside the 0.9-contour line. When pulse duration is 20 fs, just a few optical oscillations reach the intensity $> 0.9$ and the subsequent intensity peak enters the central focal part after the previous one has just passed (two intensity maxima occur per optical oscillation since $I \propto |E|^2$). Obviously, this discussion does not take into account the ionization and effects on nonlinear light propagation. However, the space-time map (Fig. 1) illustrates that a location and time dependent ionization is expected when the intensity reaches the threshold of dielectric breakdown (a full ionization of the focus). This phenomenon is expected to be important in filament formation under less tight focusing.\(^6\)

### 3.1.2. Two-pulse irradiation

Two pulses of equal energy are overlapped spatially and the resultant electrical field at the focus is calculated as function of the time separation between the pulses, $\Delta t$, and relative phase, $\varphi$. Focusing is not considered at this stage. The normalized electrical field of the pulse is given by:

$$E_{1,2}(t, \Delta t, \varphi) = \exp\left(-2 \ln 2 \left(\frac{t - \Delta t}{\tau_p^2}\right)^2\right) \cos(\omega(t - \Delta t) + \varphi),$$  \(2\)

where the same notations as in eqn. 1 are used; $\varphi$ is the phase difference between the phases of two pulses and subscripts 1, 2 denotes the pulse number. The intensity of the two pulses is then calculated as $I_{12}(\Delta t, \varphi) = (E_1 + E_2)^2$ and presents a function of the delay time and phase difference. We simulate the threshold of photomodification, e.g., dielectric breakdown, as the value $Th = 0.9I_{12}^{\text{max}}$.

The experiments on determination of a light induced dielectric breakdown (LIDT) threshold\(^4,5\) can be modeled in the following way. It is necessary to calculate the cumulative energy of the pulse once the 90% of the maximum two-pulse intensity $I_{12}$ is reached (with the typical 5% pulse-to-pulse stability of the pulse energy in actual experiments). This particular value of 0.9 was not critical for the qualitative explanation of the phenomenon of dielectric breakdown by two pulses. The cumulative energy from the time moment when the $Th$ intensity is reached till the moment when it becomes lower than the same value of $Th$ represents the effectively absorbed energy and should be proportional to the electron number density (here, we do not consider electron recombination and diffusion out of the focal region). The calculations should be carried out for optically oscillating field, hence, the intensity is calculated as $(E_1 + E_2)^2$.

Figure 2 shows the numerical results of the two pulse space-time overlap with different pulse separations. The cumulative energy (an integral proportional to the ionization yield in real experiments) has a considerable

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**Figure 2.** (a) The cumulative energy calculated as $E_{12} = \int_{0.9I_{12}^{\text{max}}}^{I_{12}^{\text{max}}} I_{12}(t)dt$ vs. the delay time $\Delta t$ at two different phase differences $\varphi = 0$ and $\pi/2$. (b) The delay-time averaged presentation of the same data as in the panel (a). Simulations were carried out using eqn. 2 for the pulse duration 50 fs (FWHM) and 800 nm wavelength.
increase at the delay times just longer that the pulse duration. At such delays, two pulses effectively add up to each other forming an equivalent longer flat-top-like pulse. As a result the duration during which the intensity is larger than the $T_h$ is longer yielding in higher value of cumulative (absorbed) energy. Also, the effect of the relative phase between two pulses is obvious, as is should be expected in the case of addition of two oscillating electrical fields due to interference.

The simple analysis of spatial-temporal overlap has demonstrated that the threshold of dielectric breakdown is expected to depend strongly on the pulse separation and relative phase. In real experiments, there are number of additional factors relevant for space-time overlap: axial alignment, the aberrations (the most important ones are those for a right angle incidence: spherical and chromatic), tilt of a pulse front, and dispersion compensation. Namely, the tilt of pulse front should be minimized, since it effectively elongate the pulse axially at the focus rendering a lower maximum intensity.\(^7\) In our experiments, the pulse tilt was minimized. Also, care was taken to achieve good axial and lateral alignment of both pulses, minimization of the spherical aberration, and dispersion compensation.\(^4,5\) Realization of experiments with co-linear time-separated pulses of the same polarization can be done using Dazzler (Fastlite) setup (the principle of operation has been published in ref.\(^8\)). One can expect the functional dependence of the breakdown similar to the one depicted in Fig. 2.

The qualitative analysis of the cumulative energy should be calculated as $I_{12}(\Delta t, \varphi) = (E_1^2 + E_2^2)$ when two cross-polarized pulses are coaxially superimposed as shown in Fig. 3. For example, the cross-polarized pulses were coaxially introduced into the microscope\(^4,5\) using Mach-Zender interferometer. Here, the effect of depolarization at the focus due to tight focusing has not been considered. Though the tight focusing has not been explicitly considered in the presented simulations, the strong dependence of the two-pulse intensity on the pulse separation and relative phase has been corroborated numerically as well as observed in actual experiments.\(^4,5\) Obviously, one should expect the breakdown to occur when the threshold intensity, $T_h$, is reached and a certain energy has been absorbed (accounted for as summation of number of pulses in the presented numerical simulations). In the case of two pulses’ axial superposition, this quantity has a complex dependence on the relative phase and pulse separation as shown in Figs. 2 and 3. Future studies should focus on a quantitative modeling of tight focusing conditions used in actual experiments.\(^4,5\) Then the pulse duration at the focus can be estimated from the experimental data (proportional to the time separation of the maxima in Figs. 2 and 3).

### 3.2. Void in silica: picosecond pulses

Voids can be made by single picosecond pulses without crack formation in high purity silica ED-C (Nippon Silica Glass; OH content $< 100$ ppm).\(^3,9\) The voids were recorded inside the slab of rectangular shape allowing the top and side observation. The voids were imaged by confocal microscope using a Hg-lamp condensor illumination at
around 360 nm, which had entitled to reach an approximately 300 nm lateral resolution in the image. Figure 4 show optical side view of the voids recorded at different pulse energies: at the threshold of void formation (a) at the 1.7 of the threshold (b), and at 7 – 8 thresholds (c). The threshold value of void formation was 40 ± 10 nJ/pulse at the used focusing conditions (NA = 1.3) (the threshold value of the optically recognizable damage was approximately 15 nJ). The presence of threshold was judged by the optical contrast. The correspondence between the void formation, observed by SEM after sample cleaving, and an optical contrast has been established in the case of void formation in silica (viosil)\textsuperscript{10, 11} and sapphire.\textsuperscript{1}

Figure 4 shows that the void was close to spherical when focusing was carried out by a high numerical aperture lens (see, (a,d)). It is noteworthy, that some of the void-structures, recorded at high pulse energy (several times the threshold of void formation) were without observable cracks. The crack-resistance of this particular ED-C brand of synthetic silica could be explained by low OH content (< 100 ppm). Usually, crack propagation is exponentially enhanced by presence of water molecules. The optical contrast change around the void was concentric with the radius approximately one-to-two times larger than the diameter of central void. It is, most probably associated with the stress pattern caused by a displaced glass.

The axially elongated voids were formed by irradiation with higher pulse energy and at lower numerical aperture (Fig. 4). The waist of the pulse and the Rayleigh length are calculated as diffraction limit for a plane wave focusing according to \( w_0 = 0.61\lambda/NA \) (the numerical aperture \( NA = 1.3 \)) and \( z_R = n\pi w_0^2/\lambda \). Hence, the confocal parameter, the axial extent of the focus, was \( 2z_R \approx 1.1 \mu m \) (for the refractive index of silica \( n = 1.47 \) and \( w_0 \approx 0.25 \mu m \)). This is an approximate length of the voids of the maximum axial length. At higher pulse energy, the cracks were formed obscuring observation of filamentation (Fig. 4(c)). In contrast, the complex pattern of voids and filaments was observed in ED-C and ED-B silica under femtosecond single pulse irradiation and was considerably longer than the confocal parameter.\textsuperscript{6, 12} Interestingly, the smallest voids appeared spherical and were not following the expected aspect ratio \( z_R/w_0 = 2.17 \) as would be expected for the employed focusing conditions (diffraction limited focusing is considered).

The structural damage induced by one femtosecond (\( \sim 200 \) fs) pulse at high intensity (up to 10 thresholds) appeared very similar to image shown in Fig. 4(c). The central part had a void (the brightest central region), which had diameter comparable with that formed by pulses of 2 – 3 thresholds. This suggests that the micro-explosion had been triggered from the small central region where the plasma was, first, formed, and, then, heated.
up with subsequent micro-explosion according to the model proposed recently.1, 2 Void formation by picosecond
pulses can prove to be advantageous as compared with femtosecond irradiation since the silica glass was found
almost free of the defects (color centers) usually caused by femtosecond pulses.4 The absorption spectra of
silica irradiated by picosecond pulses was equivalent to the spectra of silica irradiated by femtosecond pulses and
annealed for reduction of color centers. Such annealing in the case of picosecond pulses can also account for the
more spherical axial cross-section of the voids formed by high numerical aperture focusing (Fig. 1(a)).

3.3. The mechanism of dielectric breakdown

The mechanism of void formation in sapphire has been recently analytically modeled and numerically simu-
lated.1, 13 The results complied well with the experimental observation. The breakdown has been described by
the multi-photon and avalanche multiplication of electrons, plasma formation, and its expansion. The model
accounted for the void formation in silica (viosil) and polystyrene as well.2 This model predicts that the energy
gain per electron occurs approximately at a ∼ 10^{12} eV/s rate due to avalanche. The multi-photon absorption
is most important in providing seeding electrons, while the avalanche makes the most effective multiplication.
It is noteworthy, that at the discussed experimental conditions, the absorption takes place at the skin depth of
ionized volume. The skin depth is < 100 nm and the light-field is effectively shielded (exponential decay) inside
the region of dielectric breakdown, which makes the multi-photon process of electron multiplication ineffective.
However, the avalanche still provides an effective mechanism, since it is linearly dependent on intensity and is
effective within the skin depth.

The tight focusing creates conditions when approximately one optical cycle is present at the waist region
(see, Fig. 1). Once the intensity of electric field is strong enough to effectively generate free electrons (starting
from multi-photon and then avalanche) the breakdown of entire focal volume ensue since the incoming optical
cycles finds a partly ionized material at the focal region. At the irradiance ∼ 10^{13} W/cm² the breakdown, a
full ionization of the focal volume, effectively occurs in few optical cycles. For the experimental detection of
the breakdown by optical plasma emission, it appears as an instant event.14 The transfer of dielectric material
at focus into a metallic (plasma) state appears step-like since the avalanche multiplication is very efficient and
ionizes the focus within one or few optical cycles. This apparent instantaneous dielectric-to-metal transition was
explained as analogous to the Mott transition.14 The intensity of the light field at the focus is the key parameter
describing the dielectric breakdown.

The tunneling mechanism has also been proposed recently15 based on the experimental results of surface
ablation16 by femtosecond pulses at 400 nm wavelength. The values of threshold irradiance were overestimated
since the used power meter was not calibrated for the low repetition rate radiation. However, the relative ablation
thresholds determined in the same measurement showed an experimental dependence $Th \sim E_g^\gamma$ with $\gamma \approx 3.1$,
very close to the expected value $\gamma = 3$ for the tunneling breakdown ($E_g$ is the effective bandgap energy). Also, a
band-gap collapse model has been put forward,17 which explains the breakdown as a collective response of the
pulse-affected volume of crystals. The physical mechanism is the Bragg-type reflections of oscillating electrons at
boundaries of the first Brillouin zone. More direct measurements of ionization with high temporal resolution are
necessary for exact determination of a detailed evolution of the dielectric breakdown in crystalline and amorphous
materials.

4. CONCLUSIONS

The tight focusing (with the vocal volume comparable with $\lambda^3$) is favorable for creation of small photo-modifications
inside the dielectrics. When short, sub-1 ps pulses are used for irradiation, the direct laser writing can be realized
since the dielectric breakdown is reached with a pulse power smaller than the threshold of self-focusing. Such
conditions are favorable for void recording inside dielectrics. As it was demonstrated, the voids can be also
formed by picosecond pulses. Short-pulses when tightly focused creates a moving ionization at the focal region
when ionization threshold is reached. The very same phenomenon leads towards filament formation at less tight
focusing via a self-action mechanism.18

It is shown that experiments with two-pulse irradiation can show complex temporal dependence of the dielec-
tric breakdown. This dependence should be considered in actual experiments and decoupled from the instanta-
neous (electronic) modulations of the local refractive index changes at the focus which affects superposition of
two pulses. It has been demonstrated that the ionization of the focal volume is qualitatively similar when the pulses of same or perpendicular polarizations are used (the depolarization effect has not been considered).

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REFERENCES


