"Quill" writing with ultrashort light pulses in transparent materials

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A remarkable phenomenon in ultrafast laser processing of transparent materials, in particular, silica glass, manifested as a change in material modification by reversing the writing direction is observed. The effect resembles writing with a quill pen and is interpreted in terms of anisotropic trapping of electron plasma by a tilted front of the ultrashort laser pulse along the writing direction. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2722240]

Writing is the main method for information storage, which has been known from the dawn of civilization. These days direct write technologies, including plasma spray, micropen, ink jet, e-beam, focused ion beam, and laser beam are of increasing importance in material processing.[1] More recently, direct writing in transparent materials by intense ultrashort light pulses has attracted considerable interest due to new applications and phenomena ranging from three-dimensional (3D) optical waveguides[2] and microexplosions[3] to 3D self-organized subwavelength structures.[4] However, it is well recognized that reversing the writing direction should not affect material processing and associated modifications. Here we report the observation of a remarkable phenomenon in direct writing and ultrafast laser processing in transparent materials, in particular, silica glass, manifested as a change in material modification by reversing the writing direction. The effect resembles writing with a quill pen and is interpreted in terms of anisotropic trapping of the electron plasma by a tilted front of the ultrashort laser pulse. We anticipate that the observed type of light-matter interaction, sensitive to the direction of movement of a laser beam, will open new opportunities in direct writing, material processing, optical trapping, and manipulation.

A key advantage of using femtosecond pulses, as opposed to longer pulses, for direct writing is that such pulses can rapidly and precisely deposit energy in solids.[5] It has been shown that 3D self-assembled subwavelength planar structures, aligned perpendicular to the polarization direction of the writing laser, are responsible for anisotropic scattering, reflection, and negative birefringence in the regions of glass irradiated by femtosecond laser pulses in a certain intensity interval.[6] Recently, interesting applications of self-assembled nanostructures for nanofluidics and rewritable 3D optical memory have been demonstrated.[7]

The laser radiation, in a Gaussian mode, produced by a regeneratively amplified mode-locked Ti:sapphire laser (150 fs pulse duration, 250 kHz), operating at 800 nm, was focused via a 50× (numerical aperture of 0.55) objective into the sample. A series of lines was directly written by scanning in alternating directions at a depth of 0.5 mm below the front surface. The writing speed was 200 μm/s and each line was written with only one pass, in one direction, of the laser, with the polarization directed perpendicular to the line and pulse energy of 0.9 μJ. After writing, the structures were side polished and imaged with a scanning electron microscope (SEM). The SEM images expose tracks elongated in the direction of light propagation due to the beam's confocal parameter, and enhanced by self-focusing effects, with a periodic structure in the direction of light polarization (Fig. 1). On closer inspection we were surprised to observe a difference in the structures written in opposite directions. This difference is revealed in small variations of the length of the tracks and of a tilt of the periodic structures written in the forward and reverse directions. The periodic planar nanostructures are aligned along the direction of the writing laser polarization and are responsible for form birefringence of the irradiated regions.

In another experiment we wrote a series of lines using an IMRA-FCPA μJewel D-400 amplified ytterbium fiber laser system, operating at 1045 nm, with pulse duration <500 fs and repetition rates ranging from 100 kHz to 1 MHz. The

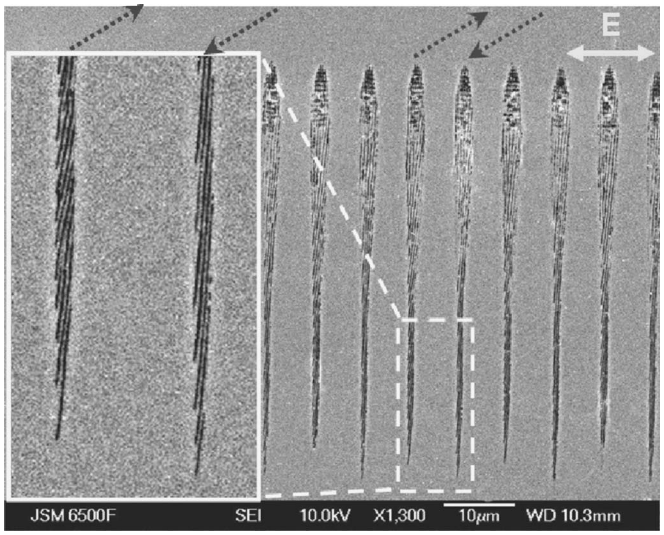

FIG. 1. SEM images of cross sections of the structures in glass along light propagation. The distance between lines is 7 μm.

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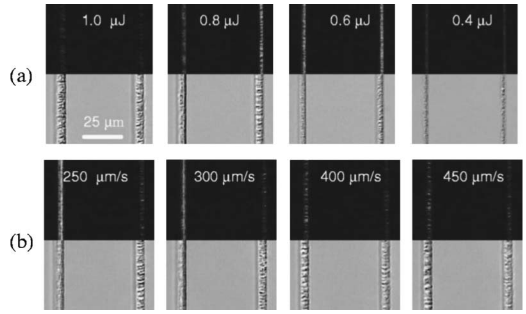

FIG. 2. Images in crossed polarizers (dark part) and Nomarski-DIC (light part) of the lines written in glass in opposite directions at repetition rate of 500 kHz with (a) writing speed of 500 μm/s and different energies and (b) pulse energy of 0.9 μJ and different writing speeds.

high stability of the FCPA laser system is crucial for systematic studies. The polarization of the laser was aligned perpendicular to the writing direction. The lines were written in alternating directions from forward to reverse and using different pulse energies ranging from 0.2 to 1.8 μJ. After writing, microscope images were captured using both crossed-polarized (CP) and Nomarski-differential interference contrast (DIC) illumination (both back illumination). Composite images were created, which show the same portion of each feature using the two illumination techniques. With these composite images, the amount of birefringence visible with the CP illumination can be compared with the texture of the feature using the DIC imaging technique. In one of the experiments we wrote ten groups of four lines with alternating writing direction and each series with different repetition rate and speed. The product of repetition rate and writing speed was kept constant in order to maintain the same total fluence. Lines written in both directions at low energies were the same (Fig. 2). However, with an increase in energy we observed the appearance of directional dependence in the written lines, which was strongest at about 0.8–0.9 μJ. The directional dependence is more clearly seen in the birefringence of the lines. This dependence can also be observed in the morphology (texture) of the lines written in opposite directions, with a line written in one direction being rougher than a line written in the reversed direction [Fig. 2(a)]. However, with further increase of energy above a certain threshold value, both lines become uneven with indications of collateral damage, and the birefringence of the lines disappears as well as the directional dependence. We believe that the latter phenomenon can be explained by a cumulative thermal effect. This is supported by the presence of modifications with rough features, much bigger than the spot of the beam, at high repetition rates (500 kHz–1 MHz) and the absence of such features with collateral damage at low repetition rates (below 300 kHz). This agrees with the heat diffusion time of about 1 μs. We also tested the dependence of the observed effect on writing speed near the energy threshold of the disappearance of directional phenomenon and observed that the directional dependence strengthens at lower writing speeds [Fig. 2(b)].

When the beam was turned by 90°, using a two-mirror periscope, and other writing parameters maintained the same including the polarization of the laser beam perpendicular to the stage movement, we observed only weak difference in structures written in opposite directions. This indicates that an asymmetry in the structure of the beam can be responsible for the observed phenomenon.

An intriguing result is the observation of *different* textures in the processed material for laser polarizations perpendicular and parallel to the movement of the sample *in one direction* and the *same* textures for two polarizations when writing in the *opposite* direction [Fig. 3(a)]. The SEM images of the cross sections of the lines, along the light propagation, revealed a different texture in the lines written in opposite directions [Fig. 3(b)]. Remarkably, the nanograting of about 300 nm period, which is responsible for the form birefringence of irradiated regions, can be seen only in the initial part of cross sections of lines written in one of two directions. This small area is followed by one with a collateral damage due to thermal effect, which correlates with a weak birefringence of these lines. It is also observed that in almost entire cross sections of the lines, written in opposite direction, there is the nanograting along the direction of light polarization with the period of about 250 nm together with the additional periodicity, along the direction of light propagation, of about 720 nm, which is of the wavelength of light (λ/n, λ=1045 nm, n=1.45) [Fig. 3(b)]. These lines demonstrate no evidence of the collateral thermal damage and much stronger birefringence [Fig. 3(a)].

Lines written at a repetition rate of 100 kHz also clearly show different textures in opposite directions, without any evidence of collateral damage due to thermal effect. The

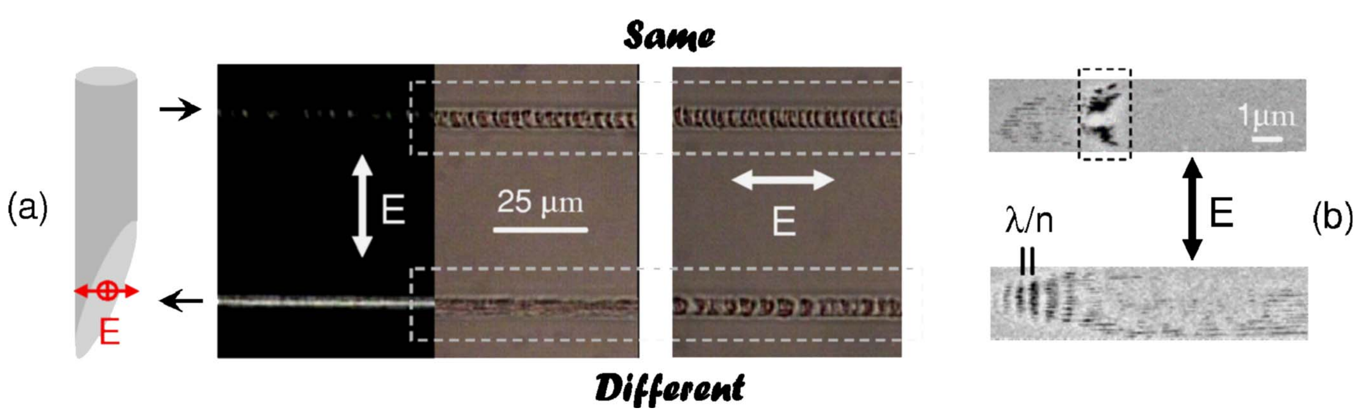

FIG. 3. (a) CP and DIC images of the lines written with orthogonal polarizations with 500 kHz repetition rate, writing speed of 250 μm/s, and pulse energy of 0.9 μJ. The difference in texture for two polarizations is observed only for one writing direction. The tilted front of the pulse along writing direction is shown. (b) SEM images of cross sections of lines written with polarization perpendicular to writing direction are also shown. The region of collateral damage is marked with a black dashed line.

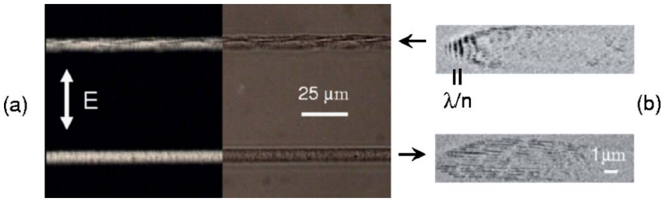

FIG. 4. (a) Optical microscope CP and DIC images and (b) corresponding SEM images of cross sections of the lines written in glass in opposite directions with repetition rate of 100 kHz, writing speed of 100 μm/s, and pulse energy of 2 μJ.

SEM images reveal the presence of the nanograting in the direction of light polarization almost in the entire cross section for one writing direction and the nanograting, again with additional periodicity along the propagation direction of about the wavelength of light, for opposite writing direction [Fig. 4(b)].

The writing anisotropy is observed only at particular pulse energies, which excludes the stage movement as the cause. Inspection of the intensity distribution of the laser beam did not reveal any peculiarities in the shape of the beam, which was close to being circular (Gaussian shape). The only possibility left to explain the puzzle of the writing direction anisotropy is related to the anisotropy of the frequency distribution (frequency chirp) in the beam. A spatial frequency chirp and related pulse front tilt are quite common in femtosecond laser systems.[8] Even a small delay across the beam that corresponds to ~10% of the pulse duration results in a pulse tilt as strong as tens of degrees in the vicinity of the focal plane. The pulse front tilt is enhanced in a dispersive media, as in the case of electron plasma close to plasma frequency, which is formed in the focus of the beam due to multiphoton ionization of glass. The pulse front tilt is a tilt in the intensity distribution in the front of the pulse. It is known that in the presence of intensity gradients, the charges (e.g., electrons) experience the ponderomotive force (light pressure), which expels the electrons from the region of high intensity.[9] Indeed, free electrons are affected by a variation of the laser intensity as they quiver in the electric field of the laser pulse. In the nonrelativistic case this can be expressed with the fluid equation of motion in an electromagnetic field by

$$\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v}\cdot\nabla)\mathbf{v} = -\frac{e}{m_e}(\mathbf{v}\times\mathbf{B}+\mathbf{E}),$$

where $\mathbf{v}$ is the electron velocity vector, m_e is the electron mass, and e is the electron charge. The ponderomotive force $\mathbf{F_p}$ and potential $\mathbf{U_p}$ follow from this equation by time averaging the electric field as

$$\mathbf{F_p} = -\frac{e^2}{2\varepsilon_0 c m_e \omega^2}\nabla I \text{ and } \mathbf{U_p} = \frac{e^2 I}{2c\varepsilon_0 m_e \omega^2},$$

where c is light speed in vacuum, ε_0 is permittivity of free space, ω is the frequency of light, and I is the light intensity. For very short and relativistic laser pulses, the ponderomotive force can become very important and the resulting acceleration will tend to push electrons in front of the laser pulse, as a kind of "snow-plough" effect. Estimated intensity in the focus of a laser beam in our experiments is of about 3×10^{14} W/cm^2, which will produce ponderomotive potential of 60 meV. This potential is higher than the energy at room temperature, which is of about 40 meV. Electron plasma in our experiments will still experience this kind of force in front of the pulse. Due to the tilt of the intensity distribution, the force will act on the electron plasma along the direction of the intensity gradient. By moving the beam, the ponderomotive force in the front of the pulse will trap and displace the electrons along the direction of movement of the beam and only in one direction corresponding to the tilt in the intensity distribution (we refer to this phenomenon as the "quill effect"). The electron plasma waves, excited in electron plasma, are responsible for the formation of the nanograting[6] and self-assembled form birefringence.[7] The trapping and displacement of the electrons with the movement of the beam affect the interference of plasma waves, and related form birefringence. The periodic structure, with the period of the wavelength of light, along the direction of light propagation is created as a result of the interference between plasma waves and plasma oscillation. Trapping of the electron plasma *damps plasma oscillation* and related interference, producing longitudinal periodic structure with the wavelength of light. The observed difference in the onset of the collateral thermal damage for two writing directions is also the consequence of the anisotropic tapping effect [Fig. 3(b)]. Further support of the proposed mechanism is the evidence of different textures of modified material for writing with light polarizations parallel and perpendicular to the movement in one of the writing directions [Fig. 3(a)]. This observation is explained by the difference in boundary conditions for two orthogonal polarizations at the interface of the *tilted pulse front along the writing direction*.

In conclusion, it is remarkable that a laser beam, one of the most modern writing tools, could be used for calligraphic inscription similar to writing with a quill pen, which is based on the anisotropy of a quill's tip shape. Moreover, modifications of materials by light span from photosynthesis and photography to material processing and laser writing, and there are only few parameters of the light beam which control material transformations, in particular, wavelength, intensity, polarization, exposure time, and pulse duration. Our results add one more parameter to this list—direction of beam movement or pulse front tilt.

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[1]D. B. Chrisey, Science **289**, 879 (2000).
[2]K. M. Davis, K. Miura, N. Sugimoto, and K. Hirao, Opt. Lett. **21**, 1729 (1996).
[3]E. N. Glezer and E. Mazur, Appl. Phys. Lett. **71**, 882 (1997).
[4]Y. Shimotsuma, P. G. Kazansky, J. Qiu, and K. Hirao, Phys. Rev. Lett. **91**, 247405 (2003).
[5]B. C. Stuart, M. D. Feit, A. M. Rubenchik, B. W. Shore, and M. D. Perry, Phys. Rev. Lett. **74**, 2248 (1995).
[6]E. Bricchi, B. G. Klappauf, and P. G. Kazansky, Opt. Lett. **29**, 119 (2004).
[7]C. Hnatovsky, R. S. Taylor, P. P. Rajeev, E. Simova, V. R. Bhardwaj, D. M. Rayner, and B. P. Corkum, Appl. Phys. Lett. **87**, 014104 (2005).
[8]S. Akturk, M. Kimmel, P. O'Shea, and R. Trebino, Opt. Express **11**, 491 (2003).
[9]W. L. Kruer, *The Physics of Laser Plasma Interactions* (Addison-Wesley, Redwood City, 1988), Vol. 73 p. 60.