Photosensitivity control of an isotropic medium through polarization of light pulses with tilted intensity front

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Abstract: We present the first experimental evidence of anisotropic photosensitivity of an isotropic homogeneous medium under uniform illumination. Our experiments reveal fundamentally new type of light induced anisotropy originated from the hidden asymmetry of pulsed light beam with a finite tilt of intensity front. We anticipate that the observed phenomenon, which enables employing mutual orientation of a light polarization plane and pulse front tilt to control interaction of matter with ultrashort light pulses, will open new opportunities in material processing.

OCIS codes: (260.0260) Physical optics; (320.0320) Ultrafast optics; (140.3390) Laser materials processing.

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1. Introduction

Material processing with ultrafast lasers [1] has recently attracted considerable interest due to a wide range of applications including laser surgery [2, 3], optical data storage [4], integrated optics [5, 6], 3D micro- [7–9] and nano-structuring [10]. However, despite significant efforts, the physics of interaction of intense ultrashort light pulses with transparent materials still is not fully understood [11]. Recently, two remarkable phenomena have been reported: nonreciprocal photosensitivity manifesting itself as dependence of material modification on light propagation direction in non-centrosymmetric crystal [12] and a quill writing effect revealing strong dependence of the material modification in glass on the orientation of writing direction relative to the direction of the pulse-front tilt [13–15]. In these effects, dependence of the imprinted structures on polarization of the laser beam can be attributed to the intrinsic anisotropy of the experiment associated with the beam movement. However, in stationary conditions, the optical anisotropy produced by an intense polarized pump beam can be revealed only using a second light beam of different polarization, i.e. the light induced anisotropy cannot be accessed by a single polarized beam or a pair of equally polarized pump and probe beams. In particular, photosensitivity of an isotropic medium is polarization independent, i.e. the change in the absorption coefficient and/or refractive index produced by a light beam does not depend on the beam polarization. Here we observed that it is not necessarily true when isotropic medium interacts with intense femtosecond optical pulses possessing a tilted intensity front. We demonstrate that the modification of glass depends on the mutual orientation of the light polarization azimuth and the pulse front tilt (PFT). We present the first experimental evidence of *anisotropic* photosensitivity of an *isotropic* homogeneous medium under uniform illumination and interpret this new phenomenon in terms of intrinsic anisotropy of light-matter interaction involving ultrashort light pulses with a tilted intensity front. The PFT can be controlled using prism or grating pairs, which are commonly used as pulse compressors/stretchers in ultrashort laser amplifier systems. We anticipate that the observed phenomenon, which enables employing mutual orientation of a light polarization plane and intensity front to control interaction of matter with ultrashort light pulses, will open new opportunities in optical material processing and manipulation.

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2. Experiments

In the experiment, a series of dots was written in alumosilicate glass (sample composition SiO_2 64, Al_2O_3 17, B_2O_3 5, CaO 15 wt%) using a focused linearly polarized femtosecond laser beam at normal incidence. We employed a mode-locked, regeneratively amplified Ti: Sapphire laser system (Coherent; RegA 9000), operating at 800 nm wavelength with 70 fs pulse duration and 250 kHz repetition rate. The beam was focused via a 50× (0.80 N.A.) microscope objective (Nikon; LU Plan Fluor) at about 60 μm below the surface of the glass slab. The polarization plane azimuth of the beam was different for each dot. The beam spot size in the focus was estimated to be ~1.5 μm and the pulse energy was 2.4 μJ. The spatial and polarization properties of the beam were characterized on the laser output, before and after the microscope objective. The beam had a circular cross-section with linear polarization purity of as high as 98% maintained by dielectric mirrors. The beam power measured after the microscope objective and after the sample was the same for all polarizations. We did not observe any indications of the asymmetry in the focal plane intensity distribution. However, unexpectedly we observed pronounced dependence of the white light emission from the irradiated region on the polarization plane azimuth (Fig. 1a). Moreover, inspection of the irradiated regions revealed that at a certain orientation of polarization azimuth, strong coloration was accompanied by asymmetrically distributed bubbles (Fig. 1b). The diameter of the central coloured region (20 μm) was significantly larger than the beam spot size indicating that the high fluence and heat accumulation [3] produced colour centres away from the focal volume. The Raman spectra of the encrypted structures were measured using laser a confocal Raman spectrometer (Tokyo Instruments; Nanofinder 30). Microscans along the irradiated regions showed strong Raman bands at 150-190 cm^{-1} and 500 cm^{-1} (see Figs. 1d, e), which can be assigned to the O-Si-O bond deformation of the coupled tetrahedral SiO_4 groups and the bending vibration modes of Si(Al)-O-Si(Al) bridges in glass, respectively. One can observe from Fig. 1(b, c) that intensity of Raman signals at 155 cm^{-1} and 500 cm^{-1} correlate with the coloration intensity of the light affected zones, however the signal is weaker for the beam with vertical polarization. At 155 cm^{-1}, the Raman intensity shows a minimum in the centre of the light effected zone with the maxima at about 5 μm on both sides from the center for both polarizations.

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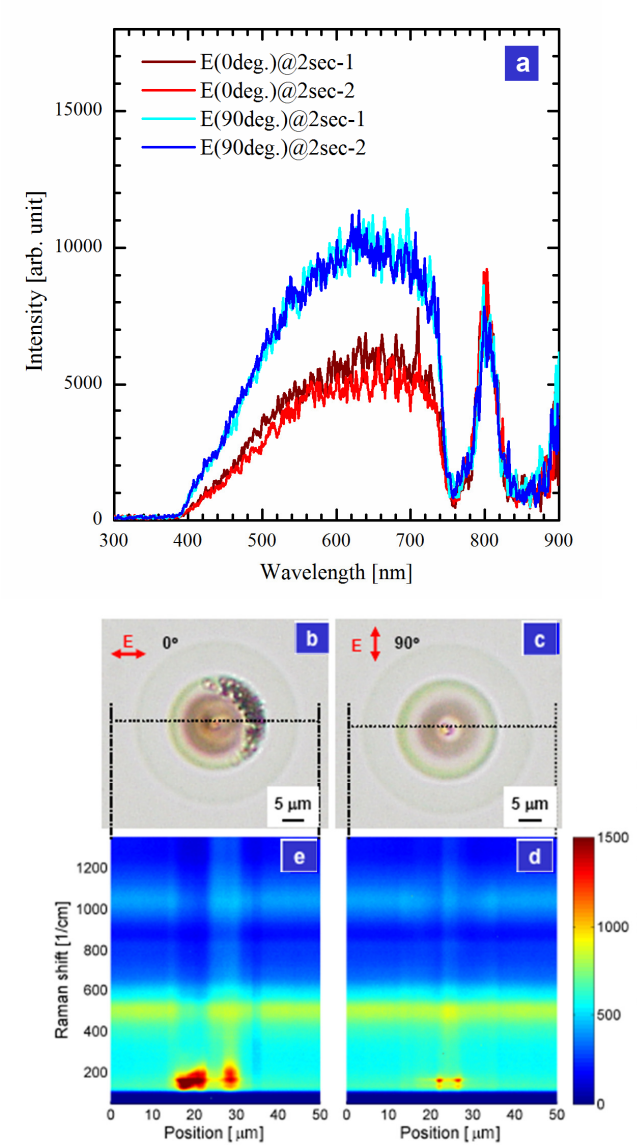

Fig. 1. (a) Spectra of transmitted light during laser exposure with two orthogonal polarizations. The pump power at 800 nm was reduced by a factor of 105 using notch filter. The results of two independent measurements for each polarization display good reproducibility. Transmitted light optical microscope images of modified regions irradiated for 1 s with two orthogonal polarizations (b-c), corresponding Raman spectra along line scans through the irradiated regions (d-e).

The absorption spectra of the irradiated regions were measured using a microscope spectrometer (Craic; QDI 302) with the spectral resolution of 1 nm and spectral range of 380 - 850 nm. The induced phase change in the irradiated regions was measured using quantitative phase microscopy (QPM) via the optical microscope equipped with a microscope automation piezostage (Physik Instrumente; PIFOC P-737). A quantitative phase map of the structures was produced using the QPM software (Iatia Vision Sciences). The refractive index change of the structure is estimated from $\Delta n = \Delta\varphi\lambda/(2\pi d)$, where $\Delta\varphi$ is the phase shift

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of the light at wavelength λ, and d is the thickness of the structure. The stress region is analysed using a quantitative birefringence imaging system (CRI Abrio). We observed that in the spectral range of 380 - 850 nm, the absorption increases towards a shorter wavelength, while absorbance shows cosine-like dependence on the polarization azimuth angle α (Fig. 2 a). The measured phase shift $\Delta\varphi$ allowed us to map the refractive index change Δn at λ = 550 nm. One can observe from Fig. 2 (c, insert) that Δn is at its maximum in the central part of the modified area. Assuming the thickness of the structure along the light propagation direction $d \approx 20$ μm, we arrive at $\Delta n \approx -2\times10^{-2}$ when the polarization azimuth is $\alpha = 90°$ and $\alpha = 270°$.

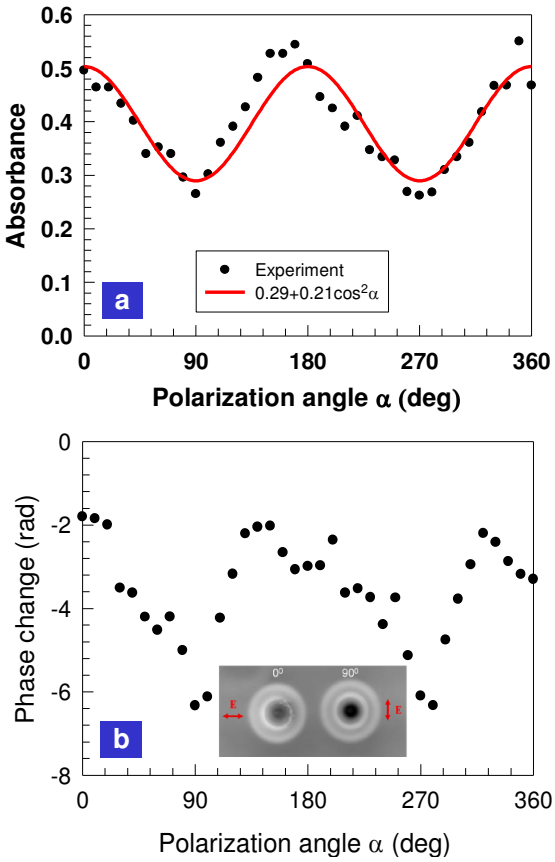

Fig. 2. (a) The absorbance of the irradiated region at 400 nm as a function of the polarization angle α of writing laser. Solid line corresponds to $\cos^2\alpha$ fit of the experimental data. (b) Experimental dependence of the induced phase change in the center of irradiated regions on the polarization plane azimuth of writing laser. Inset shows induced phase change maps for two orthogonal polarizations of femtosecond light pulses. Darker color corresponds to more negative index change.

We also analyzed the dependence of induced modifications on the exposure time (from a single shot to 10 seconds) for two polarizations corresponding to the maximum and the minimum of the measured absorbance and phase shift. The difference in absorbance and phase shift was found to reach steady-state value after a few seconds when the average heat production in the focus area was balanced by the thermal diffusion processes. The reduction of the refractive index in the irradiated region heated above glass transition temperature (720 °C) is likely to be produced by density reduction accompanying glass expansion.

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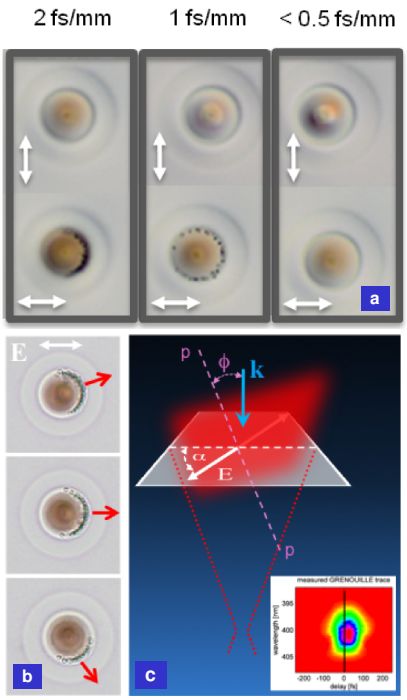

Fig. 3. (a) Dots written with orthogonal polarizations and three different PFT values. (b) Transmitted light optical microscope images of modified regions produced by three different orientations of pulse front tilt. White and red arrows indicate directions of the writing laser polarization and the pulse front tilt. (c) Pulse with tilted intensity front is represented by the red tetragon. An electromagnetic wave with wave vector $\boldsymbol{k}$ is incident on a planar density gradient produced by tilted intensity front at a nonzero angle of incidence ϕ and has the electric field vector $\boldsymbol{E}$ lying at an angle α to the plane determined by $\boldsymbol{k}$. (Insert) Typical measured GRENOUILLE trace. The black line indicates zero delay. The shift of the trace center in delay axis indicates the pulse-front tilt.

Characterization of the spatio-temporal properties of the ultrashort pulse laser beam using the GRENOUILLE device (Swamp Optics; 8-50, see Fig. 3c, insert) and PulseCheck autocorrelator (APE) revealed that the femtosecond pulse has the Gaussian intensity profile and a small pulse front tilt of about 1.8 fs/mm, which may originate from the angular dispersion and simultaneous spatial and temporal chirp [16, 17]. However the absence of astigmatic focal spots in the light modified glass indicates that the contribution of angular dispersion to the PFT is negligible. The PFT is proportional to the beam diameter and can be significantly increased in the vicinity of beam waist (we calculate that PFT reaches ~180 fs/mm 10 μm before the geometrical focus in air).

In order to visualize the effect of the PFT on the observed anisotropic photosensitivity we changed the spatial chirp (hence the intensity front) in both transverse directions independently by tuning the grating pulse compressor and the acousto-optic Bragg cell (employed as pulse picker) in the laser system. We observed that the internal structure of the light-modified region depends on both the PFT value (Fig. 3a) and the beam polarization azimuth angle α. Measurements revealed that α = 0° and α = 90° correspond to the direction along and perpendicular to intensity front normal, respectively. By adding PFT in the other transverse axis we changed the direction of the intensity front normal. In the experiment, we observed the correlation between the orientation of the pulse front with respect to the polarization azimuth of the writing laser beam and the direction associated with the strongest induced absorption (Fig. 3b). The position of the area where bubbles were formed with

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respect to the centre of the beam spot is also correlated with the direction of the PFT. This experiment unambiguously demonstrates that the tilt in the intensity front of the pulse is responsible for the observed anisotropic photosensitivity of the isotropic glass.

Cross-sections along the beam propagation direction (Fig. 4) show that the strongest modifications occur in the pre-focus region and that modification morphology differs dramatically for beam polarized parallel and perpendicular to the PFT. Specifically, when the beam is polarized along the PFT, the region of strong coloration in the tail and the coloured frozen jet in the head of the imprinted structure. The stunning coloured regions are accompanied with the bubble belt in the middle and few axial bubbles respectively for 8 s (Fig. 4, top) and 2 s (Fig. 4, bottom) exposures. An extraordinary, coloured flower-shaped structure was also observed for the beam polarized perpendicular to the PFT.

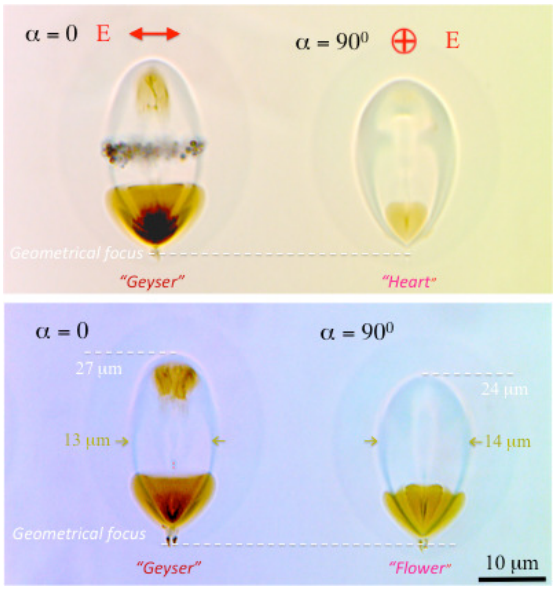

Fig. 4. Optical microscope images of modified regions along beam propagation direction for writing beam polarised along (α = 0) and perpendicular (α = 90°) to the pulse intensity front normal for exposure time 2 s (bottom) and 8 s (top).

Our experiments do not reveal any evidence of filamentation, which often takes place when an intense ultrashort light pulse propagates in a transparent medium. We believe that this is because of the high numerical aperture (NA=0.8) of the employed objective. It has been already reported [18–20] at such tight focusing conditions the filamentation effects are not dominant. In the intense focused femtosecond pulse, the plasma reaches the maximum density at a certain distance before the geometrical focus [19]. This distance in our experiment is estimated to be about 15 μm corresponding to a beam diameter of about 5 μm.

In the experiment, we observed a weak dependence of the absorbed light power and strong dependence of the hot plasma emission on the light polarization. This can be seen as another manifestation of the hidden anisotropy of the light beam when the same amount of absorbed energy for different polarization plane orientations can result in different material modifications. It is worth noting that the PFT-induced anisotropy of the light-matter interaction manifests itself in the modification of the glass produced even by a single shot indicating that it originates from electronic rather than thermal mechanism.

4. Discussion

One may recall that exciton generation via multi-photon absorption/ionization is considered as one of the main mechanisms of defect formation, which are responsible for material

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modifications under ultrafast laser irradiation in wide band gap dielectrics (e.g. silica glass [21, 22]). In our experimental conditions, this implies that pondermotive forces associated with anisotropic intensity gradients at finite PFT in the prefocus region effectively remove defects from the beam path for the light polarized along the pulse front tilt reducing photon losses. That is the intensity of such a beam at the focus will be higher than that of one polarized perpendicular to the PFT one and hence will produce stronger modification of the glass as we observed in the experiment (see Fig. 4). The inner cores of the elongated regions of “Geyser”, “Flower” and “Heart” (Fig. 4) are molten due to the heat produced by light absorption. Small vacuum or gas-filled bubbles are formed in the molten core in about 1 ns. The strongly coloured region of “Geyser” is composed of colour centres, which are produced by UV harmonics generated near the beam focus where high intensity of light is reached. The detailed quantitative description of this absorption mechanism requires significant theoretical efforts and will be considered elsewhere.

In conclusion, we demonstrate for the first time that modification of the transparent isotropic glass by intense ultrashort laser pulse can depend on the polarization azimuth of the laser beam. We attribute this new phenomenon to the anisotropy of the light-matter interaction caused by space-time couplings in ultrashort light pulses, which opens an interesting opportunity in the control of photon flux interacting with a target submerged into condensed isotropic medium. We refer the phenomenon as *ultrafast light blade*, drawing an analogy between strong material modifications produced by ultrashort light pulses polarized along a tilted front and material cutting with sharp blade.

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