**Anisotropic nanostructure generated by a spatial-temporal manipulated picosecond pulse for multi-dimensional optical data storage**

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Anisotropic nanostructures can be generated in fused silica glass by manipulating the spatiotemporal properties of a picosecond pulse. This phenomenon is attributed to the laser-induced interband self-trapped excitons. The anisotropic structures exhibit birefringent properties, and thus can be employed for multi-dimensional optical data storage applications. The generation of data voxels by such short laser irradiation enables on-the-fly high-speed data recording. © 2021 Optical Society of America

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Optical data storage is considered an ideal candidate for storing massive digital data owing to its high capacity, longevity and green aspects [1]. However, further increasing its lifetime and capacity is a prohibitive task in conventional diffraction-limited polymer-based optical disk systems. Therefore, novel techniques such as holographic [2], multi-dimensional [3] and super-resolution data storage [4] have been developed to increase data density. Additionally, bulk glass materials have been employed to exert their high Young’s modulus and high resistance properties for long-term data storage [5–7].

However, from a practical standpoint, many state-of-the-art techniques require multiple laser pulses or long laser exposure to form a single data voxel structure, thereby substantially hindering the data writing speed. Five-dimensional optical data storage based on volume nanostructures was successfully implemented in glass with an almost unlimited lifetime [5], but such a structure requires multiple pulses to be generated due to an incubation effect [8]. In nanoplasmonic hybrid glass, the data writing of each multiplexed data voxel requires several tens of milliseconds of laser irradiation [7]. Furthermore, a four-step millisecond-scale writing procedure was required to record a diffraction-unlimited data spot based on photoswitchable fluorescent proteins [4].

In recent years, tremendous efforts have been devoted to reducing the overall laser exposure time using various optical data storage techniques. As a precursor substrate, nanoporous glass has been exploited to reduce the minimum number of pulses; however, a pulse sequence on a microsecond timescale was still required [9,10]. A three-pulse burst scheme successfully enabled anisotropic structure generation in fused silica in sub-nanoseconds, but the uniformity was unsatisfactory [11]. Other approaches based on laser-induced micro-void [12], photoreduction [13] and carbon fluorophores [14] only require single-shot femtosecond pulses, but require further mask correction or confocal fluorescence microscopy. A practical, easily implemented and inexpensive approach with a fast data writing speed and dense capacity is yet to be realized. To achieve a breakthrough, the instantaneous electrodynamic properties of materials can be harnessed by manipulating either the spatial or temporal, or both properties of light, thus introducing a new degree of freedom in the ultrafast laser system [15–20].

Herein, we demonstrate anisotropic nanostructures generated in fused silica by manipulating the spatial and temporal distribution of a picosecond beam. The spatiotemporal manipulation of the beam was accomplished using a delay-line and a spatial light modulator, which were then replaced by a simple birefringence crystal. In contrast to the conventional simultaneous spatial and temporal focusing (SSTF) technique, in which a spatial chirp and angular dispersion are created in the focal region [16], our scheme allows for the dynamic manipulation of a significant beam displacement of up to 3 μm (with current objective lens) and a longer temporal delay of up to 660 ps. The generation of polarization-independent anisotropic nanostructures is attributed to the temporal interband excitons and the refractive index increase. Such nanostructures exhibit birefringence properties and can be used for optical data storage.



Fig. 1. Schematic illustration of the experiment configuration. (a) Schematic illustration of the setup: beam splitter (BS), delay line (DL), half-wave plate (HWP), polarizer (POL), Spatial Light Modulator (SLM) and objective lens (OL). (b) Schematic illustration of the setup with a birefringent crystal (BC). (c) Schematic drawing of a beam temporally divided into two beams with orthogonal polarizations. (d) Schematic drawing of two beams with dynamically manipulated spatial locations.

A frequency-doubled femtosecond laser system (YSL, FemtoYL) with a wavelength of 515 nm and pulse width of 300 fs was employed in the optical system [Fig. 1]. In our experiments, the system irradiated only a single femtosecond pulse. The pulse was split into two beams using a beam splitter. The pulse energy of each beam could be manipulated individually in each arm. In one arm, a motorized delay line (Thorlabs, ODL100) was used to control the temporal delay between the two pulses. The pulse energy was tuned using a programmed rotating half-wave plate and a linear polarizer in each arm. A liquid crystal spatial light modulator (UPOLABS) was employed to dynamically manipulate the extraordinary light [dashed green light in Fig. 1] by placing corresponding blazed grating phase patterns (0.0–0.2 lines/mm). A fixed flat phase shift was applied to the ordinary light [solid green light in Fig. 1]. Thus, the extraordinary light can be placed at variable distance (*d*) and azimuthal directions (*θ*) to the ordinary light on the focal plane [Fig. 1(d)]. By using this setup, the laser parameters, including the pulse energies and polarizations of the first and second pulses, pulse delay, and relative location of the second pulse can be tuned. On the focal plane, the center-to-center distance between the two beams ranged from 0±20 to 550 nm in our experiments. To achieve such near-perfect overlapping of the two beams, the second beam was scanned in a circular trajectory by switching hologram patterns. Then the relative positions of the two beams were verified by the corresponding birefringent profiles in multi-pulse nanogratings regime. The angle difference and longitudinal displacement between the two beams were negligible at the focus based on the Debye theory calculations [21]. A dry objective lens with a numerical aperture of 0.55 (New Focus 5720) was employed to focus the beam 30 μm below the surface. A programmed three-axis translation stage (AUS-PRECISION) was used to position and scan the sample.

We used a parameter sweep strategy to determine the parameter regimes where the generated structures exhibited uniform birefringent profiles. The parameters included delay time, total pulse energy, energy ratio between the first and second pulse, and distance (*d*). *θ* was either 90° or 270°. Uniform birefringent structures were observed only when the energy ratio was approximately 2:1 and *d* ranged from 300 to 400 nm. The corresponding temporal and pulse energy window ranged from 1 to 300 ps, and 180 to 250 nJ respectively. For example, from the experiment results of varying *d* (fixed 1 ps delay, 230 nJ total pulse energy, 2:1 energy ratio), uniform birefringent structures were observed when *d* ranged from 300 to 400 nm [Fig. 2(a)]. The experiment results of two sets of total pulse energies with varying delay (fixed 400nm distance, 2:1 energy ratio and *θ* = 90°) are shown in Fig. 2(b). At a low pulse energy (190 nJ), birefringent structures were observed when the delay ranged from 1 ps to 300 ps [Fig. 2(b)]. The temporal processing window narrowed with an increase in the pulse energy. At a high pulse energy (230 nJ), similar birefringent structures were only observed when the delay ranged from 1 to 20 ps [Fig. 2(b)]. Above 50 ps, structures with randomly distributed structural and stress-induced birefringence were observed. Notably, structures exhibiting negligible birefringent signals were observed when the time delay was ± 0 fs. Figure 2(c) shows the structuring regimes as a function of the first and second pulse energies of the double single-pulse experiments for a fixed delay time of 1 ps (fixed 400nm distance, *θ* = 90°). The observation of uniform birefringent spots when the energy ratio was approximately 2:1 provides evidence for energy-dependent phenomena occurring on a picosecond timescale [Fig. 2(d)].



Fig. 2. Structuring regimes of anisotropic structures. (a) Slow-axis orientation images of the spots generated with varying distance between on-axis and off-axis beams. (b) Slow-axis orientation images of the spots generated with varying delay time. Pulse energies: (H) 230 nJ and (L) 190 nJ. (c) Structuring regimes with varying pulse energies of the first and second pulses. Dots in the retardance map indicate the conditions at which uniform birefringent structures were generated. A reference line (P1, P2 pulse energy ratio 2:1) is drawn as a visual aid. (d) Birefringent images of the corresponding structures in (c). Pseudo colors in (a), (b) and (d) indicate the orientation of the slow axis. The scale bars are 2 μm.

It is noteworthy that based on the microscopic images, isotropic void-like structures were generated by either two on-axis or two off-axis (i.e., displaced) single-pulses, which were not displaced relative to each other, with varying delay from ±0 fs to 660 ps [Fig. 3(a)]. The slow axis orientations of the birefringent structures are parallel to and determined by *θ* [Fig. 3(b)], irrespective of the polarization states of the two pulses. The glass was polished and etched to view the structures from the top by SEM [Fig. 3(c)]. Additionally, the structure was directly imaged from the side using an optical microscope [Fig. 3(e), inset]. The morphologies revealed that a tilted nanostructure with a cross-sectional width of 600 nm was responsible for the birefringent signal.



Fig. 3. (a) Contrast-enhanced microscopic images of the spots generated by the on-axis (up) and off-axis (down) double-pulses. Insets are the corresponding birefringent images. The scale bar is 3 μm. (b) Top-view birefringent and (c) SEM images of spots when changing the *θ* to 0°, 45°, 90° and 135°. Pseudo colors indicate the orientation of the slow axis. The scale bars are 2 μm and 500 nm in (b) and (c) respectively. (a-c) Pulse energy: 230 nJ; pulse energy ratio between the first and second pulse: 2:1; *d*: 400 nm; delay: 1 ps. (d) Schematic illustration of the proposed formation mechanism of the tilted nanostructure. (e) The FDTD (Lumerical FDTD Solutions) simulated intensity distribution when the second pulse is focused into a gradient positive refractive index change (+0.1 maximum) region. The geometrical centers of the first and second beam are labelled using the white and black solid triangles, respectively. Inset is the corresponding side-view contrast-enhanced optical image of the structure in ((b), 90°). The scale bars are 500 nm (e) and 3 μm ((e), inset), respectively.

Based on the observations above, we speculate that two phenomena occur depending on the pulse energy: one is introduced at a relatively low threshold with a lifetime of ~ 300 ps and the second is induced after 20 ps by a pulse with higher energy. The 300 ps time matches the lifetime of self-trapped excitons (STEs) generated after the femtosecond laser pulse [22,23]. The induced STEs possess a 2.1 eV energy above the valence band [24], and result in an increase in absorption [22]. These properties of STEs can be harnessed to break the diffraction limitation [18], but cannot explain the structural tilt in our experiments. A positive refractive index change was found during the lifetime of the STEs [25–30], and was attributed to the trapping of electrons [25,28]. The thermo-optical effect can also introduce a positive refractive index increase (up to 0.1) on a timescale of approximately 10 ns immediately after plasma relaxation [27,30]. The structures induced by a 126-nJ (two-thirds of 190 nJ) energy single-pulse exhibit no refractive index change under a defocusing microscopy [31]. Therefore, we speculate that the STEs generated at such energy levels relax into non-bridging oxygen hole centers (NBOHCs) [26], without inducing a permanent refractive index change [8]. At higher pulse energies, for example, 153 nJ (two-thirds of 230 nJ), we predict that the STEs evolve into oxygen deficiency centers (ODC) [8,32] and lead to the generation of interstitial oxygen [33,34]. This relaxation of STEs started immediately after self-trapping and may result in void formation on a picosecond timescale [22,35]. Thus, the non-uniform structures that were generated at high pulse energy when the delay was over 50 ps are likely the result of structural-inhomogeneity-induced field enhancement [36].

Therefore, the formation mechanism of the observed tilted nanostructure can be explained as follows: The first pulse led to the generation of STEs. The STE region possessed a high absorption with a lifetime of 300 ps and yielded a positive refractive index on a nanosecond-timescale. Consequently, the second pulse was mainly absorbed in the pre-modified overlapped region. Meanwhile, the material in this region served as a convex lens and focused on the second pulse with a 400 nm displacement [Fig. 3(d)]. Thus, a tilted intensity distribution was introduced [Fig. 3(e)]. Although the intensity tilt has a relatively small angle based on our simulations, we speculate that the subsequent thermodynamic process dominates the structural evolution and causes the structure to grow into its final morphology [Fig. 3(e), inset].



Fig. 4. (a) Slow axis orientation image of the recorded 4D voxel array. The *θ* was changed by switching hologram patterns on the SLM. The spot separation is 1.5 μm. The scale bar is 20 μm. (b) Slow axis orientation image of the 3D recorded data. The spot separation is 1.5 μm. The scale bar is 10 μm. Inset is enlarged data arrays highlighted by the white square frame in (b) and the corresponding decoded letters. Letters were encrypted based on the 7-bit ASCII codes. Pulse energy: 230 nJ. Pulse energy ratio between the first and second pulse: 2:1. *θ* was fixed at 90° in (b).

Voxel recording experiments were performed to demonstrate high-speed data writing. The birefringent spots with dynamic manipulated slow axis orientation as an additional degree of freedom (4th dimension) were generated by changing *θ* through the SLM [Fig. 4(a)]. The logos of Huazhong University of Science and Technology (up) and Wuhan National Laboratory for Optoelectronics (down) were printed. To increase the speed, a digital micromirror device (DMD) or pockels cells can be employed to replace the SLM. Another scheme was a three-dimensional optical data storage, whose recorded voxels possessed a fixed slow axis orientation. The laser system shown in Fig. 1(a) was simplified to a direct laser writing setup with a birefringence crystal [Fig. 1(b)]. The employed birefringence crystal was a commercially available TiO2 crystal (10 mm × 10 mm × 1 mm), which introduced a one-picosecond temporal delay between two orthogonally polarized beams. The pulse energy ratio between the two beams was tuned by manipulating the polarization state of the beam before the crystal. The outer surface of the crystal was slightly polished to an inclined plane to generate a certain spatial displacement *d*. A text file was recorded in glass 30 μm underneath the surface. The readout process is illustrated in Fig. 4 (b). The accuracy reached up to 99.09% (134 bits errors out of 14,641 bits). Benefiting from the birefringent property of the data voxels, only a crossed-polarized imaging system is required for data readout. Both two schemes allow for a laser-repetition-rate scale data writing speed with an on-the-fly rotational disk system similar to Blu-ray.

In conclusion, we have presented a novel method of introducing anisotropic structures using a spatial-temporally manipulated pulse with a duration down to one picosecond. The appearance of such birefringent structures has been attributed to the properties of the temporal interband excitons generated after femtosecond laser irradiation. Such a method can streamline the beam manipulation procedure and enable state-of-the-art high-speed data recording. As the orientations of these nanostructures are independent of the polarization states of the pulses, polarization manipulation becomes non-essential. Additionally, by manipulating the spatiotemporal properties of the beam, a tilted nanostructure with arbitrary 3D orientation can be generated [37]. Thus, a six-dimensional optical data storage technique could be implemented in the future. The dimensions include three-dimensional space, slow axis orientation, retardance, and the polar angle of the tilted structures.

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**Data Availability**. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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