High speed ultrafast laser anisotropic nanostructuring by energy deposition control via near-field enhancement

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1. INTRODUCTION

The current storage technology is not growing fast enough to keep up with the vast amount of digital data generated worldwide. To meet the ever-increasing data storage demand, cloud providers rely on many storage technologies including hard disk drives (HDDs), magnetic tape, and even optical disks. However, the drawbacks of the HDDs are their high energy consumption and their short lifespan of several years [1], and the long average-case response time of magnetic tapes prevents their application. More futuristic DNA-based data storage is capable of holding hundreds of terabytes (TB) of data per gram, but its durability is limited [2]. Optical data storage has been heralded as an energy-efficient solution with a longer lifetime, but traditional optical data storage technology of CDs and DVDs only has a capacity of hundreds of gigabits per disk and a lifetime of a decade.

Advances in the femtosecond (fs) laser writing in wide bandgap materials have opened a door to high-density data storage with a long lifespan by rapid energy deposition with high precision [3,4]. The idea of optical recording based on fs laser writing was first proposed and demonstrated in photopolymers [5,6]. Later, three-dimensional (3D) optical data storage was realized in silica glass by tight focusing of femtosecond laser pulses [7,8]. The high-capacity optical recording was demonstrated by multiplexing new degrees of freedom including intensity, polarization, and wavelength by harnessing silver clusters embedded in glass [9] and plasmonic properties of metallic nanoparticles [10]. Polarization multiplexed data recording with high capacity and virtually unlimited lifetime was implemented using self-assembled nanogratings [11–13] generated by ultrafast laser writing in silica glass by means of the slow axis orientation (fourth) and retardance (fifth) in addition to three spatial coordinates [14,15]. More recently, a new type of ultralow-loss birefringent modifications constituted by oblate nanopores with a size of <40 nm has been demonstrated in silica glass [16], which can be utilized in multilayer data storage (≥100 layers) with ultrahigh readout accuracy [17]. The generation of these structures requires irradiation with dozens of fs laser pulses, which limits the speed of writing.

The throughput of optical data storage based on fs laser writing can be improved by utilizing high repetition rate lasers [18] if unwanted thermal accumulation could be eliminated. Thermal accumulation could be reduced by temporal modulation of picosecond laser pulses in welding [19] or by gigahertz bursts of fs laser pulses in ablation-cooled material removal [20]. Alternatively, the detrimental thermal effects could be reduced by improving the efficiency of energy deposition during laser processing. One possible approach is using near-field effect to locally enhance the electric field of laser pulse for material modification. Although optical near-field enhancement has been tailored by sharp tips
distinguishable birefringent dots were observed with the slow axis pulse number from 20 to 100. At the repetition rate of 1 MHz, with different repetition rates from 1 to 10 MHz [Fig. 1(b)] and a matrix of voxels was imprinted in silica glass by pulses (E = 30 nJ) with a rate of 10^6 voxels/s and a data capacity of terabytes/disk. Furthermore, the proposed mechanisms of microexplosion and near-field enhancement presented here [25] are generic, and the methodology can inspire other studies of fs laser processing of various materials.

2. METHOD

We used two different laser systems to cover a broad range of laser parameters. The experiments were carried out with a mode-locked Yb:KGW regenerative fs laser system (PHAROS, Light Conversion Ltd.), operating at a wavelength of 1030 nm and the second harmonic (515 nm) with a repetition rate set to 500 kHz and a pulse duration tuned in the range of 190–700 fs. The second system was a Yb-doped fiber fs laser (Satsuma, Amplitude), operating at 515 nm with a repetition rate of 10 MHz and a pulse duration of 250 fs. The pulse energy modulation (PEM) was achieved by the control of the electro-optic modulator (EOM) in the laser (Fig. S1 in Supplement 1). The laser beam was focused with a 0.60 NA objective lens (Olympus) with an aberration correction collar 170 µm below the surface of a silica glass substrate (Corning 7980), which was mounted on an XYZ linear air-bearing translation stage (Aerotech Ltd.). The diameter of the laser spot at the focus was ∼1 µm, which was measured with a knife-edge technique (Fig. S2 in Supplement 1). The energy of the laser pulses was controlled by an attenuator and measured after the objective lens. The maximum laser peak power of 0.25 MW stays well below the self-focusing critical power 0.9 MW at 515 nm in silica glass. A combination of a polarizer, a Pockels cell, and a quarter-wave plate [Fig. 1(a)] was used to control the polarization azimuth of the laser beam.

The laser-induced birefringence, retardance and slow axis orientation, were quantitatively analyzed with an Olympus BX51 optical microscope equipped with a birefringence measurement system (CRi Abrio imaging system) operating at 546 nm wavelength. The optical transmission spectra and phase change were measured with a VIS/NIR microspectrometer (CRAIC) and a wavefront sensor (SID4-HR, Phasics), respectively. After lapping/polishing and etching with a KOH solution (1 mol/L) in 24 h, the laser-modified region was imaged by a scanning electron microscope (SEM, Zeiss Evo50).

3. RESULTS AND DISCUSSION

A. Writing of Anisotropic Nanostructures

As mentioned before, a high repetition rate fs laser could improve the throughput of laser nanostructuring. To examine the feasibility, a matrix of voxels was imprinted in silica glass by pulses (E = 30 nJ) with different repetition rates from 1 to 10 MHz [Fig. 1(b)] and pulse number from 20 to 100. At the repetition rate of 1 MHz, distinguishable birefringent dots were observed with the slow axis orientation perpendicular to the laser beam polarization. However, modification accompanied by strong stress appears in the writing region due to the heat accumulation for a repetition rate of 5 and 10 MHz.

One solution to reduce the thermal accumulation is using a smaller laser spot. Alternatively, the thermal effects could be reduced if a modification is produced with an efficient energy deposition, meaning more energy is used to modify rather than heat the material.

More specific, birefringent modifications can be written by fs laser pulses in silica glass based on anisotropic nanostructures, such as nanogratings and elongated nanopores. The generation of these structures requires irradiation with dozens of fs laser pulses, resulting in increased energy consumption per voxel and low writing speed. Single pulse with energy density above the Young’s modulus is able to produce a circular nanovoid [7,26]. The subsequent pulses with less energy can change the shape of the nanovoid to an anisotropic one via near-field enhancement, where the localized field still can modify the material [27–29].

The optical near field forms at the edge of a nanovoid during the irradiation of laser pulses. According to the boundary conditions, the light field is enhanced at the edge along the circumference of the nanovoid in the plane perpendicular to the light polarization azimuth [Fig. 2(a)]. The light intensity enhancement is decreased from 1.3 for a 40 nm nanovoid to 1.15 with a larger nanovoid diameter of 200 nm [Fig. 2(b)]. The enhanced light field could induce localized ionization, resulting in the formation of anisotropic nanostructure with less energy compared to what is required for isotropic nanovoid formation. Therefore, the detrimental thermal accumulation produced by high repetition rate pulses could be reduced.

The nonlinear transmission of silica glass for a single pulse was measured to justify laser parameters such as pulse duration and energy (Fig. S3 in Supplement 1). A shorter pulse duration was chosen as less pulse energy is required to produce modifications. As previously stated, a nanovoid can be generated if the absorbed energy density is larger than the Young’s modulus of silica glass, meaning pulse energy should be higher than 32 nJ. Considering
the near-field enhancement, about 30% less pulse energy, 24 nJ, is sufficient to convert the isotropic nanovoid to an anisotropic one.

In the experimental demonstration, two seeding pulses ($E_s = 32$ nJ, 18.9 TW/cm$^2$, 3.6 J/cm$^2$) and eight writing pulses ($E_w = 14.4$ nJ, 8.5 TW/cm$^2$, 1.6 J/cm$^2$) were focused in the silica glass sample [Fig. 3(a), left]. The writing pulse energy of 14.4 nJ was used, since the damage was detected with a writing energy of 24 nJ. Well-defined birefringent voxels were observed with an average retardance of 2.6 nm and the azimuth of the slow axis perpendicular to the laser beam polarization. On the other hand, modification with evidence of collateral damage occurred when writing with 10 pulses of 32 nJ energy [Fig. 3(a), middle-left]. No visible birefringent modification was detected when writing with two seeding pulses of 32 nJ or 10 writing pulses of 14.4 nJ [Fig. 3(a), middle-right and right].

The temperature evolution as a function of time was simulated at the center of the laser spot by the thermal diffusion equation (Section 4 in Supplement 1), and the heat accumulation was evident for 10 MHz pulses of 32 nJ [Fig. 3(b), red curve]. If two seeding pulses ($E_s = 32$ nJ) and eight writing pulses ($E_w = 14.4$ nJ) were focused, the estimated material temperature is reduced from 21,000 K to below 1200 K by less energy deposition [Fig. 3(b), blue curve]. In the simulation, we used the measured nonlinear absorbance of 30% for 32 nJ pulse, 1% for 14.4 nJ pulse, and the absorbed volume of 0.12 µm$^3$.

Birefringent voxels with 3.5 ± 0.3 nm retardance were fabricated by two seeding pulses ($E_s = 36$ nJ) and eight writing pulses ($E_w = 16.8$ nJ) with a repetition rate of 500 kHz and pulse duration of 190 fs at 515 nm wavelength [Fig. 4(a)]. The corresponding SEM imaging reveals the localized nanolamella-like structures orientated perpendicular to the polarization direction of the incident beam [Fig. 4(b)], which is responsible for the birefringence. We assume that all nanovoid structures were etched to a whole depth of a few micrometers within an imaging area of 10 µm x 15 µm. A circular nanovoid was created by seeding pulses at the center of the photoexcited region, and the nanolamella-like structure was subsequently produced by writing pulses via near-field enhancement [Figs. 4(c) and 4(d)]. The length and width of the nanolamella are of about 460 and 50 nm, respectively [Fig. 4(c)]. The round shape nanovoid of 130 nm was surrounded by a compacted region,
which can be attributed to laser-induced microexplosion when the energy density (>105 GPa) in the absorption volume exceeds the Young’s modulus (73 GPa) of silica glass [26,30]. Based on the mass conservation law, the estimated density of the densified region is $1.10 \pm 0.04$ times higher than in pristine silica glass (Fig. S4c in Supplement 1) [26]. The nanovoid diameter of 130 nm was only one-seventh of the laser spot size of 1 μm, suggesting that the microexplosion happens only in the central part of the photoexcited region owing to multiphoton ionization (four photons of 2.4 eV for silica glass) and threshold for reaching critical plasma density. We assume that the irradiation of the nanovoid with the first writing pulse produces an enhanced near field, yielding a short nanotip-like structure along the electric field direction. During the irradiation with subsequent writing pulses, the nanotip eventually evolves into a nanolamella several hundred nanometers long [Fig. 4(e)]. In addition, the nanolamella-like structure is asymmetric when laser beam polarization is perpendicular to the sample translation direction (Fig. S5 in Supplement 1). The reason is that an asymmetric light field distribution is generated due to a 100 nm offset between the center of the nanovoid and the last pulse of the eight writing laser pulses caused by stage movement.

B. Nanolamella-Like Structure Properties and Dependence on Laser Parameters

There are several crucial factors for the formation of high-quality birefringent voxels via near-field enhancement with the PEM method. The first factor is the number of writing pulses. Using PEM, the retardance of the birefringent voxels initially shows an approximately linear increase followed by a plateau-like behavior with increasing of writing pulse number [Fig. 5(a)]. In this experiment, the voxels were imprinted by two seeding pulses ($E_s = 36 \text{ nJ}$) and different numbers of writing pulses ($E_w = 16.8 \text{ nJ}$) with a repetition rate of 500 kHz and pulse duration of 190 fs at 515 nm wavelength. Although a single pulse is sufficient to produce a nanovoid, here we used two seeding pulses due to the limitation of the fast energy control unit, which has been solved in an upgraded version. Without PEM, where the energy of each pulse is 36 nJ, the retardance increase from 0.9 to 5.0 nm was observed when the writing pulse number increased from 10 to 100. To reach a retardance of 3.5 nm, only two seeding and eight writing pulses were enough to produce voxels with a smaller deviation of retardance due to the reduction of thermal accumulation, while 60 writing pulses were required without PEM.

We also investigated the dependence of anisotropic nanostructure writing on pulse energy. Three types of modifications (damage, birefringent voxels, and unreproducible voxels) were observed at different parameter windows of writing and seeding pulse energy [Fig. 5(b)]. When the writing pulse energy was smaller than 18 nJ, inconsistent birefringent voxels were observed at higher (>30 nJ) seeding pulse energy, meaning some of dots were birefringent but others are not under the same laser writing conditions [Fig. 5(b), right-bottom]. If the writing pulse energy was larger than 18 nJ, well-defined birefringent voxels were produced [Fig. 5(b), right-middle]. Nevertheless, when the energy of writing pulses was larger than 22 nJ, all the voxels were accompanied by collateral damage [Fig. 5(b), right-top].

The retardance and the azimuth error of birefringent voxels with different pulse durations were compared [Fig. 5(c)]. The energy of two seeding pulses and eight writing pulses were 33 and 18 nJ, respectively. The increase of retardance is only 0.4 nm, but the azimuth error increased from 2.0 deg to 2.8 deg with a longer pulse duration. Birefringent voxels imprinted by 190 fs pulses are more accurate in terms of the retardance and azimuth of the slow axis compared with those written by 700 fs pulses [inset in Fig. 5(c)]. Furthermore, we observed that writing birefringent voxels at 1030 nm wavelength requires seeding pulse energy higher than 120 nJ. Compared with 515 nm wavelength, the retardance was 2 times higher, and no saturation was observed for the writing

![Fig. 5](image-url)
pulse number up to 50 [Fig. 5(d)]. Nevertheless, larger modification size and higher transmission loss were observed for IR laser writing.

The transmission of 10-layer birefringent nanolamella-like structures with voxel separation of 1 µm was compared with the same layer number structures imprinted in the regime of nanopores (Type X, voxel separation 3 µm) or nanogratings (Type 2, voxel separation 1 µm) [Fig. 5(e)]. The transmittance of Type X in the visible range is higher than 99%. The transmission in the visible range of nanolamella-like and nanograting structures written by 515 nm were higher than 90% and about 60%, respectively, while the transmission of nanolamella-like structures imprinted by 1030 nm was less than 60%. The transmission loss can be attributed to Rayleigh scattering in Type X and Mie scattering in other cases.

The optical image of the structures along the beam propagation direction (side view) shows that the modified region consists of a dark head and a bright tail [Fig. 6(a)]. Though the retardance of the birefringent structures imprinted by PEM increases with a higher writing pulse number, the length of the structure remains almost the same—3.5 µm, which is 1.5 times smaller than that without PEM [Fig. 6(b)]. The maximum birefringence (Δn) of the nanolamella-like structure was 0.8 × 10⁻³ [Fig. 6(c)], which is close to Type X modification (0.6 × 10⁻³), but about 5 times smaller than the typical birefringence (3–5 × 10⁻³) of nanogratings (Type II). The phase measurement of the structure reveals that the nanovoid or nanolamella-like structure was created in the dark head with negative index change, and densification occurred in the bright tail with positive index change [Fig. 6(d)]. Using the same total pulse number of nine, the structure created without energy modulation shows a maximum negative phase difference of 13 nm in the head part, which is about 2 times larger than its counterpart generated by PEM. The positive phase change reveals that the densifications are at the same level in both cases. For voxels written by PEM, the phase change of the dark head and bright tail show small absolute value growth with the increase of the writing pulse number [Fig. 6(e)].

C. 5D Data Storage by Nanolamella-Like Structures

As a demonstration, nanolamella-like structures were imprinted in silica glass for 5D optical data storage, and the information was encoded into two retardance levels and eight azimuths of slow axis, implying 4 bits of information per voxel. A laser with a wavelength of 515 nm and a repetition rate of 10 MHz was used to write data with a rate of 10⁶ voxels/s, as one voxel was produced by five or eight pulses. However, such high throughput by raster scanning with the dot separation of 1 µm requires a translation velocity as high as 1000 mm/s [Fig. 7(a)], which is a significant challenge for a high-precision translation stage. As a more pragmatic alternative, we used an acousto-optic deflector (AOD) in the optical setup to scan the focused laser spot in the direction perpendicular to the stage scanning. The AOD channel number could be controlled, and here we chose four channels in the schematic diagram [Fig. 7(b)]. The laser pulses were first focused to write a data voxel and then were deflected by the AOD perpendicular to the stage moving direction to imprint other voxels, so the stage translation speed could be reduced to 100 mm/s for 10⁶ voxels/s writing.

The matrix of data voxels was imprinted with a rate of 10⁶ voxels/s [Fig. 7(c)] as the number of AOD channels was 10. One seeding pulse (30 nJ) and seven writing pulses (13.5 nJ) were used for the high retardance level of ~3 nm, while one seeding pulse (24 nJ) and four writing pulses (13.5 nJ) were focused for the low retardance level of ~1 nm. The data readout accuracy without any error correction algorithm was 100% [Fig. 7(d)], suggesting this method can be utilized for practical 5D data storage.

![Fig. 6.](image)

**Fig. 6.** Optical side-view image of voxels written by PEM and without PEM. The voxels were imprinted by one seeding pulse (E₀ = 33 nJ) and different numbers of writing pulses (Eₚ = 18 nJ) with a repetition rate of 500 kHz and pulse duration of 190 fs at 515 nm wavelength. Nine pulses with energy of 33 nJ were used for the case without PEM. (a) Optical images of the modified structures created with a different number of writing pulses. The green arrow shows the laser propagation direction. (b) Measured retardance and the longitudinal length of the voxels presented in (a); (c) calculated birefringence (Δn₀) of structures in (a); (d) phase image of structures under unpolarized light illumination and the extracted spatial optical phase difference (OPD) distribution; (e) OPD in different parts of the structure versus writing pulse number. The open square and circle indicate the phase of structure without PEM.

![Fig. 7.](image)

**Fig. 7.** Optical data storage of 5 GB data. (a) Schematic diagram of raster scanning by stage translation and (b) combination of raster and AOD scanning. Each green circle indicates one data voxel, and the numbers show the temporal sequence of data recording. (c) Birefringent voxels written by the combination of the PEM and AOD with 10 channels. The pulse trains include one seeding pulse and seven (or four) writing pulses (515 nm, 250 fs, 10 MHz, 96 mm/s, 9.6 × 10⁶ voxels/s). Pseudo-colors (inset) indicate the local orientation of the slow axis. (d) Distribution of the readout data points from (c) with eight azimuths of slow axis orientation and two levels of retardance. (e) Illustration of data encoding and decoding.
The amount of 5 GB digital data was written in 120 mm × 2.4 mm across 50 layers in the bottom-to-top layer order, and the data writing speed was 225 kB/s with the imprinting rate of 6 × 10⁶ voxels/s. The parameters of dot separation 1.2 μm, layer separation 10 μm, stage scanning speed 90 mm/s, and AOD channel number 8 were used. The text data were encoded by the American Standard Code for Information Interchange (ASCII) code, where every two voxels correspond to a character and the hexadecimal number of each spot varies from 0 to 15 [Fig. 7(e)]. The data readout accuracy for the top layer and the bottom layer data are 99.5% and 96.3%, respectively, which could be improved to 100% by an error correction algorithm. Applying the same parameters, 1.64 TB digital data can be stored in a silica glass plate of 127 mm size and 3 mm thickness.

4. CONCLUSION

In summary, we demonstrated the rapid writing of highly localized nanolamella-like structures in silica glass using energy deposition control via near-field enhancement. The nanolamella-like structure (~460 nm) was produced by the near-field enhancement from a circular nanovoid (~130 nm) generated by microexplosion. Anisotropic nanostructures with imprinting rates of 10⁶ voxels/s and data density of TB/disk have been exploited to realize multilayer optical data storage with nearly 100% readout accuracy. The current data readout speed of several bytes/s, limited by the manually controlled imaging system, could be improved to tens of megabytes/second by automatic polarization imaging and more powerful decoding algorithms.

Using this method, MB/s data writing speed could be achieved with a 40 MHz repetition rate femtosecond laser. By encoding 8-bit information in a voxel and reducing lateral voxel and layer separation to 0.2 and 3 μm, the potential data capacity of a 4 mm thick 127 mm glass plate could be ~500 TB. Nanochannels with a width of ~50 nm can be effectively produced in silica glass by continuously scanning of the laser spot to extend the nanolamella-like structure, which can be harnessed in integrated optics and microfluidics for chemical and biology applications [31]. Since nanovoids can be produced in various materials and near-field enhancement is a universal phenomenon, we anticipate the controllable energy deposition method could be used for high-rate nanostructuring of transparent materials.

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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.

Supplemental document. See Supplement 1 for supporting content.

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