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Photoinduced self-assembly of nanostructure in glass

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Abstract: Ultrashort-pulsed laser direct writing can be useful for a 3D material processing. Especially the localized form-birefringence originated from self-assembled nanostructure in isotropic material (i.e. SiO_2 and GeO_2 glass) was demonstrated.

1. Introduction

Material processing with ultrafast lasers has recently attracted considerable interest [1] due to a wide range of applications including laser surgery [2], 3D micro- and nano-structuring [3]. An intriguing phenomenon which currently attracts a lot of interest is self-assembly of periodic nanostructures in the direction perpendicular to the light polarization [3-5]. Uniaxial birefringence observed after femtosecond laser irradiation of SiO_2 glass [6] has been explained by induced nanogratings and referred as self-assembled form birefringence [7,8]. Self-organization process has been interpreted in terms of the interference of electron plasma waves resulting in electron concentration modulation, followed by freezing of the interference pattern by structural change in glass. In the case of SiO_2 glass, self-assembled nanostructure, which is composed of the periodic modulation of oxygen defects, has initially evolved from residual birefringence originated from internal stress distribution due to thermal distribution with steep gradients [9]. Evolution of optical anisotropy in glass can be controlled as a function of interpulse time due to strong polarization-dependent thermal accumulation [10]. Such phenomenon can also be observed in the plasma emission during laser irradiation indicating the polarization-dependent energy transfer in laser produced plasma. On the other hand, the stripe-like voids with a width of ~ 30 nm were self-aligned perpendicular to the polarization direction inside TeO_2 crystal [5]. More interestingly, in the case of GeO_2 glass, not only the form birefringence within the focus area but also the distribution of constituent elements of glass, which includes the generation of molecular oxygen [11], can be observed according to increase in laser energy.

2. Experiments

The experiments were performed using a mode-locked, regeneratively amplified Ti: Sapphire laser system (Coherent; RegA 9000), operating at 800 nm with 70 fs pulse duration and 250 kHz repetition rate. The femtosecond laser beam was focused via a microscope objective (Nikon; LU Plan Fluor, 50× 0.80 N.A.) at a depth of about 100 μm below the surface of SiO_2 or GeO_2 glass sample. Typical pulse energy was about 1.0 μJ and the beam power measured after microscope objective was independent on the orientation of light polarization. At first we have confirmed that the direction of slow axis of form birefringence (a_{slow}) by using SiO_2 glass, which is always perpendicular to the writing light polarization (E), can be controlled by rotating half-wave plate. We have also observed the modified region inside GeO_2 glass (Fig. 1) after irradiation of femtosecond laser with various pulse energy. In the case of higher pulse energy (> 0.4 μJ), the generation of molecular oxygen can be observed by Raman microspectroscopy (not shown here). Previously we have confirmed the nanograting is rewritable by tuning the time delay between orthogonally polarized double pulses. To discuss the effect of temporal pulse shape on the phase retardance induced by the form birefringence, amplified pulses were shaped with an acousto-optic programmable dispersive filter (AOPDF: Dazzler, Fastlite). The double-pulse train with different time delay was focused inside SiO_2 glass. In the experiments of the asymmetric pulse energy of double-pulse train was fixed to 3 ps.

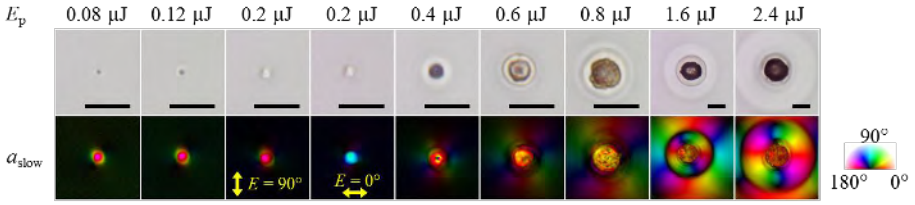

Fig. 1. Characteristic micrographs of the induced structure inside GeO_2 glass by focused femtosecond laser pulses with various pulse energy (E_p), taken with optical (upper row) and polarization (lower row) microscope(pseudo color indicates direction of the slow axis, a_{slow}, see polar legend). At the pulse energy of 0.2 μJ, two different experiments by using the orthogonal polarized femtosecond laser pulses (E = 0° or 90°) have been performed. Scale bars indicate 5 μm.

3. Results and Discussion

In the case of GeO_2 glass, the form birefringence can be observed within the focus area for the lower pulse energy than 0.2 μJ. As is the case with SiO_2 glass, the slow axis orientation is also perpendicular to the laser polarization, suggesting the nanograting structure could be self-organized inside GeO_2 glass for lower pulse energy (< 0.2 μJ). To compare between the conventional single-pulse train and the shaped double-pulse train, the total pulse energy was set the same value of 0.68 μJ (Fig. 2(a)). The phase retardance induced by the double-pulse train was larger than that of the single-pulse train, regardless of polarization direction. This phenomenon could be interpreted in terms of the enhancement of generation of free electron via the laser-plasma interaction of inverse bremsstrahlung during relaxation time of plasma excited by the first arriving pulse [12]. There was little change in the induced retardance for different double pulse delay in the experiments. Interestingly, the stress accumulation depends on the polarization direction in the case of the single-pulse train, which could be explained by the anisotropy of electron plasma absorption for *p* and *s* polarizations at the oblique, moving with the speed of light interface produced by the pulse with tilted intensity front. Such anisotropy affects the reciprocal evolution of the material modification in isotropic medium, finally exhibits a non-reciprocal quill writing effect [13]. In order to demonstrate the effect of interaction between double-pulse laser and plasma, the double-pulse experiments of asymmetric pulse energy ratio between the first arriving pulse (E^{1st}) and the total energy of double pulses ($E^{tot} = E^{1st} + E^{2nd}$) were also performed (Fig. 2(b)). Insets in Fig. 2(b) show pulse diagrams of double-pulse experiments. The maximum retardance can be induced by the one-to-one double-pulse configuration. Symmetric variation of the induced phase retardance to the energy ratio (E^{1st}/E^{tot}) may suggest individual electron excitation processes, for example, multiphoton ionization by the first-arriving pulse and avalanche ionization by the second arriving-pulse.

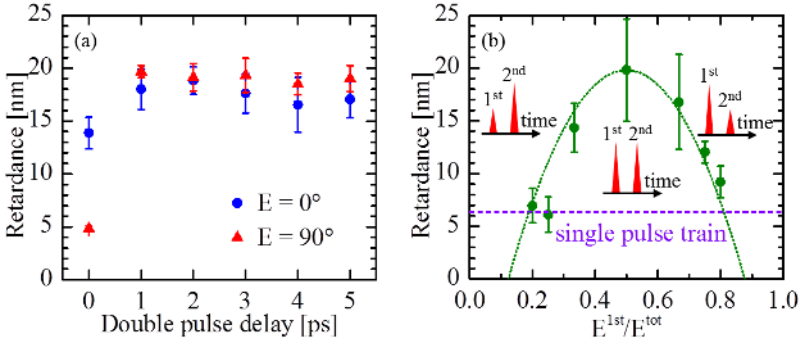

Fig. 2. (a) Phase retardance change for two orthogonal polarized double pulse (E = 0° or 90°) as a function of double pulse delay. The zero time delay indicates the results from conventional single-pulse train. (b) Variation of retardance induced by the shaped double pulse with different energy ratio of the first-arriving pulse of E^{1st} to the total energy of $E^{tot} = E^{1st} + E^{2nd}$). Dashed and dotted lines indicate the retardance for the single-pulse train and a quadratic fitting, respectively.

4. References

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