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Induced birefringence in glass: depletion and enhancement by orthogonal-polarized femtosecond pulses

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Abstract
We show spectacular and different effect of the sequential and simultaneous writing of two perpendicularly polarized ultrashort laser pulse trains on profile and magnitude of induced optical retardation inside fused silica glass. Clear birefringence was observed in the region exposed to linearly polarized pulse train radiation. It found out that the induced birefringence is erasable. It means that, when the sample is irradiated again with pulse train having perpendicular polarization, the induced birefringence is vanished and can be totally erased by optimizing the pulse energy. However, in a simultaneous writing approach, a contradictory result was observed. When the glass substrates were simultaneously (i.e. with an accuracy better than the pulse duration) exposed to two beams with perpendicular polarization the induced birefringence not only remained but also enhanced. Discussion and study on the results of interaction of polarized single ultrashort laser pulse and sequential laser beams (which spatially overlapped) having different polarizations and also change of energy ratio of simultaneously writing pulse trains helps us to analyze different results of the simultaneous interaction of two orthogonally polarized ultrashort laser pulse trains with transparent material. Our results provide pieces of evidence for further understanding the physical mechanism of creation of the birefringence using ultrashort laser pulses. Additionally, they provide the ability to manipulate the transient electron dynamics to control the profile and tailor of the induced birefringence.

Keywords Ultrashort laser pulses · Induced birefringence · Optical retardation · Nanograting · Fused silica glass

1 Introduction

Generation of volume subwavelength structures using ultrafast laser pulses is a field which has attracted considerable attention. In this field, generation of three-dimensional birefringence in the bulk of isotropic transparent materials such as glass has found various applications such as 5-D optical memories [1], optical wave plates [2, 3], polarization converters [4], polarization-sensitive holograms [5], polarization-sensitive waveguides [6], Fresnel zone plates [7], and nanofluidics channels for lab-on-chip devices [8]. To fabricate polarization devices, the distribution and the magnitude of the induced birefringence inside the materials should be carefully adjusted and controlled. Therefore, it is essential to investigate the dependency of the induced modifications on the pulse parameters of the writing laser beam.

Ultrashort laser pulses with intensities above the multiphoton ionization threshold can induce refractive index changes inside glass [9]. When the sample is irradiated by several hundreds of pulses the birefringence is induced inside the material in the focal region. Such birefringence was first observed in the bulk of fused silica by Sudrie et al. [10]. In 2003, Kazansaky et al. showed that the induced birefringence is due to the formation of self-organized periodic nanostructures in the material volume. The produced periodic nanostructures are aligned perpendicular to the direction of the polarization of the writing laser beam with periodicity determined by the laser wavelength [11]. Because of the impact of the polarization state of the laser beam on the orientation of the nanostructures, one can consider an electronic origin for this phenomenon.

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Despite the importance of the self-organized nanogratings, the mechanism of their formation has not been fully elucidated. Although several hypotheses have been proposed to describe the formation of such periodic structures, none of them fully justify the actual mechanism of the formation. Recently, Buschlinger et al. have suggested that nano-size inhomogeneities are required as the seeds to initiate the formation of volume nanogratings [12]. In 2016, Rudenko et al. developed this theory and considered the effect of increasing the number of irradiating pulses on the induced nanostructures [13]. They reported that the formation of plasma nanoplanes perpendicular to the polarization is due to the interference between the incident wave and the inhomogeneity-scattered waves and the interference in multiple scattering from nearly touching inhomogeneities [13].

In addition to the theoretical studies, different experiments have been performed to clarify the mechanism of nanograting formation and the effects of effective parameters on that. The results show that the amount of birefringence induced by the laser pulses depends on the polarization direction of the writing beams [14]. Also, it is observed that irradiating by a tightly focused, linearly polarized, ultra-short laser pulse can create nanostructures with strong birefringence which are erased when the resulting pattern is illuminated by another writing beam with orthogonal polarization and is simultaneously created new nanogratings with orientation which is solely determined by polarization of the later writing beam [15, 16]. In order to investigate nanograting erasure dynamics, Shimotsuma et.al. wrote a series of dots with different time delays between two perpendicularly polarized pulses. They observed the azimuth angle of the slow axis was determined solely by the last pulse polarization for longer delays [17]. Furthermore, in our pervious paper, we demonstrated that the optical retardation of the spatially overlapped regions written by pulses with the same polarization is enhanced while for the pulses with orthogonal polarizations is reduced [18].

In this paper, we report on experimental study of the effects of the temporal overlap of two writing ultra-short pulse trains with perpendicular polarizations for two different pulse energy ratios. We find that if the two perpendicularly polarized pulse trains are simultaneously focused in the sample, the amount and distribution of the induced birefringence inside the silica glass will be very different with that resulted from the spatial overlap of two perpendicularly polarized sequential writing beams. To analyze such results, the dependency of induced optical retardation on the polarization of ultrashort laser beams and also, the effect of the induced inhomogeneities in fused silica on induced optical retardation value and size of modified regions are utilized.

To obtain optical retardation profiles and directions of the slow and the fast axes of the induced birefringence and, hence, the nanograting orientation a colorimetry-based retardation measurement (CBRM) based on the Michel–Levy interference color chart using a polarization microscope was used [18]. Our results illustrate that the birefringence schemes inside the glass can be controlled via varying, both, of the spatial overlap of the modified regions and the temporal overlap of the two writing perpendicularly polarized pulse trains.

2 Experimental setup

The experiments were performed using a mode-locked amplified Ti:Sapphire laser system operating at central wavelength of 800 nm with minimum pulse duration of 25 fs and repetition rate of 1 kHz. To control the time delay between pulses in double-beam configuration a Mach–Zehnder interferometer (Fig. 1a) was used. It worth mentioning that to have a wider window for time delay between pulses the pulse duration was stretched to 100 fs using the prisms in the pulse compressor. The laser beam was focused at depth of about 100 μm below the glass surface using a microscope objective (50X, 0.65NA). The focused laser beam was scanned inside the sample using a three-axis translation stage so that the lines were written in x-direction inside material. Next, the distribution of the induced refractive index modifications inside the glass was imaged using a positive optical phase-contrast microscope (PCM). To measure the induced

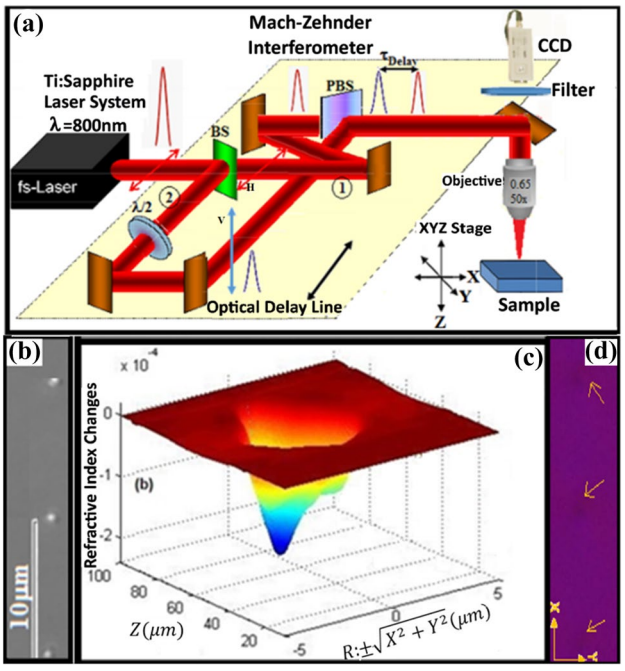

Fig. 1 Layout of experimental setup (**a**), PCM image of the modified regions (**b**), the profile of the induced refractive index changes (**c**) and PM image of the modified regions (**d**) by single shots with energy of 1 μJ and a duration of 100 fs

optical retardation a polarization microscope (PM) was used by employing two methods of applying a 'tint' plate in CBRM based on the Michel-Levy chart and a 'Senarmont' plate in measurement of polarization rotation angle. Detailed discussion on the used measurement methods can be found in our previous work [18].

3 Results and discussions

First, to verify the type of refractive index modifications, the laser beam in one arm of the Mach–Zehnder interferometer was blocked while the polarized beam of the second arm with the pulse energy of 1 μJ and duration of 100 fs was focused at depth of 100 microns under the sample surface. The scanning speed was adjusted to 1 cm/s so that it results in a 10 microns separation between two adjacent modified regions. The modified region size is estimated about 4 μm, hence, no spatial overlap of the laser spots occurs when the scanning speed is adjusted to 1 cm/s. This leads to single pulse exposure. Level of pulse energy is selected so that type II modification occurs. That's because the most studies illustrated that the nanograting structures usually occur in type II modification with negative refractive index change [19, 20]. The PCM image of the modified regions by single shot indicates that the refractive index change at the center of the irradiated regions is negative (Fig. 1b). Our comprehensive numerical calculations for the interaction of a focused ultrashort laser pulse with fused silica glass, based on the thermo-elasto plastic model, also clearly confirm that for this energy level, the refractive index changes in focal region is negative (Fig. 1c) [9]. However, the PM image of the radiated sample by single pulse does not show any induced birefringence which proves that the nanostructures are not formed in this condition (Fig. 1d) [21]. That is in agreement with the experimental results of studies that have shown that no nanostructures are observed for the pulse number of less than 100 pulses [17].

Next, we reduce the scanning speed to 1 μm/s to increase the number of irradiating pulses to more than 2000 in each region. Figure 2a shows the profile of the lateral variation of the optical retardation perpendicular to two lines separated by 8 microns and their PM image in the inset. The left/ right line is written by using H/V-polarized pulse train with blocking the second /first arm. As can be seen, a large amount of optical retardation and so birefringence has been induced at modified regions. That is due to breaking of the symmetry in the *X*–*Y* plane because of the created nanostructures. Having the birefringence axes of the tint plate, it is found that the fast-axis of the induced birefringence in the blue line is along x-axis that is parallel to the polarization direction of the writing pulse train (H-polarized) and to the written line, while it is perpendicular to the x-axis for the orange line written by V-polarized pulse train.

The maximum amount of the optical retardation of the line written by pulse energy of 1(0.5) μJ is measured equal to + 190 nm (+ 80 nm) and − 120 nm (− 60 nm) for H- and V-polarizations, respectively. It is observed that the amounts of induced retardation for two pulse trains with perpendicular polarizations of V and H are different. Such polarization dependence is also observed for metals that explained by the boundary conditions and different Fresnel coefficients [22]. But, Gecevičius et.al. proved that the cause of the polarization dependence of induced retardation inside transparent

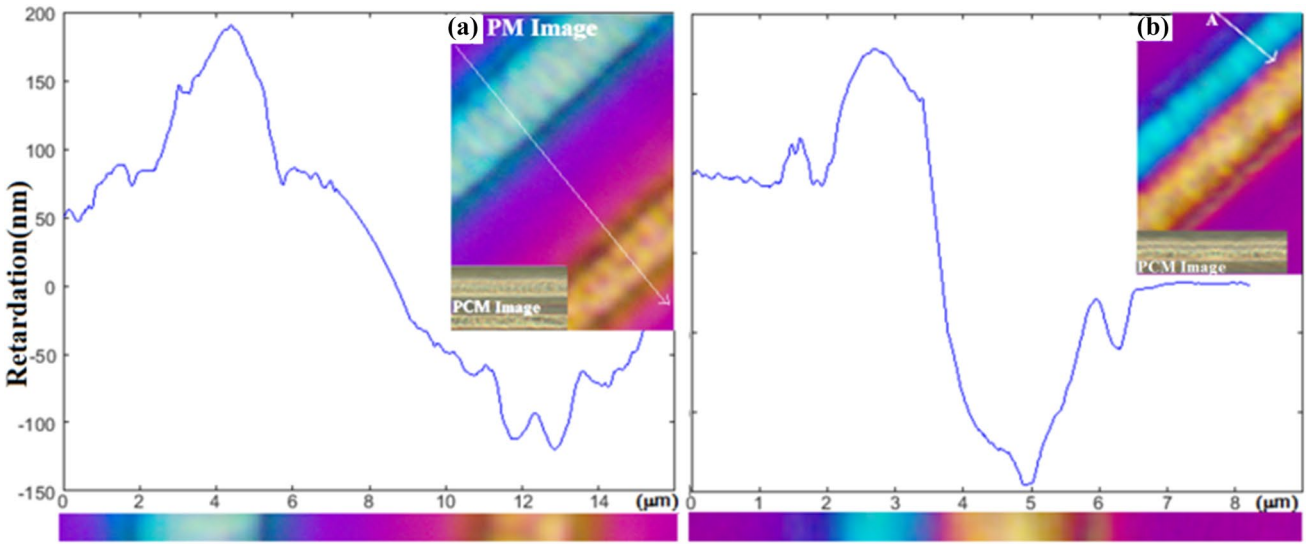

Fig. 2 Transverse profile of the optical retardation and PM images (in inset) of two lines inscribed by H- and V-polarized pulse trains with energy of 1 μJ per pulse with transverse separation of **a** 8 μm and **b** 2 μm

material could not be related to difference of the reflection coefficients [23]. They experimentally showed that if the both orthogonal polarizations are at angle of 45 degrees relative to the pulse front tilt (PFT) direction, the minimum difference in the induced optical retardation is observed. Therefore, it was properly proven that the cause of such polarization dependence is the PFT which occurs in femtosecond laser systems and intensifies in the vicinity of the objective focal plane.

The electron plasma generated by multiphoton ionization experiences different ponderomotive forces and inverse Bremsstrahlung absorption when interacting with V- or H-polarized pulse trains with tilted intensity front. It means that the energy acquired by the excited electrons in the conduction band through interaction with those pulse trains and, consequently, the electron density resulted from avalanche ionization rate is different. Therefore, in our experiment, tilted pulse fronts for H- and V-polarization are such that the former pulse is more absorbed and the thickness of induced modified region and induced birefringence are more pronounced as compared to those of the V-polarized pulses (Fig. 2a).

As shown in our previous experiments [18], the spatial overlap of two perpendicularly polarized sequential writing beams results in depletion of the induced optical retardation in the overlapped region (Fig. 2b), so that, the nanogratings created by H-polarized pulses, which are perpendicular to the scanning direction, are erased by those created by V-polarized pulses, and the optical retardation of the overlapped region is depleted (purple color line marked as 'A' in the inset of Fig. 2b).

Further examination of these images illustrates that the thickness of the second written line (orange line) is increased from 2.3 microns in Fig. 2a to 2.6 microns in Fig. 2b and the maximum amplitude of induced optical retardation is increased from − 120 to − 145 nm. Such increase can be described using the theory that the nano-size inhomogeneities are required as seed for volume nanograting formation. It seems that by writing the first line, the density of inhomogeneities increases in the location and vicinity of the line leading to reduction the threshold energy for the formation of the nanogratings for the neighboring lines. Hence, by decreasing the separation between two lines the thickness of the modified region in the line written by V-polarized pulses increases (Fig. 2b). This behavior can be also seen in PCM images in Fig. 2. Furthermore, comparison of Fig. 2a, b shows that the thickness of written blue line is reduced from 3.1 to 1.8 µm.

Similar behavior can be found in the PCM images of Fig. 3 when two line are sequently written by the similar H-polarized pulse trains where the upper line is written at first. When the focus position of the writing pulses are far from each other (Fig. 3a), both lines have equal thickness. However the thickness of the second line becomes larger when it overlaps the first line (Fig. 3b). That is due to the lower threshold for nanogratings formation for the second line as explained earlier.

These descriptions help to analyze the spectacular modifications induced by simultanously H and V-polarized pulse trains that will be described in the following:

It is observed that if two spatially overlapped H and V-polarized pulse trains have also temporal overlap the obtained results are completely different (see Fig. 4a). For the lines written unidirectionally by two temporally overlapped H- and V-polarized beams with pulse energy of 0.5 µJ, focused at the depth of 100 µm the resulted optical retardation is about − 100 nm. That is even larger as compared with the retardation of − 60 nm induced by V-polarized single beam with pulse energy of 0.5 µJ. As described for Fig. 2a, the electron plasma generation by V-polarized pulse train and the resulting induced retardation are less pronounced than those by H-polarized pulse train because of

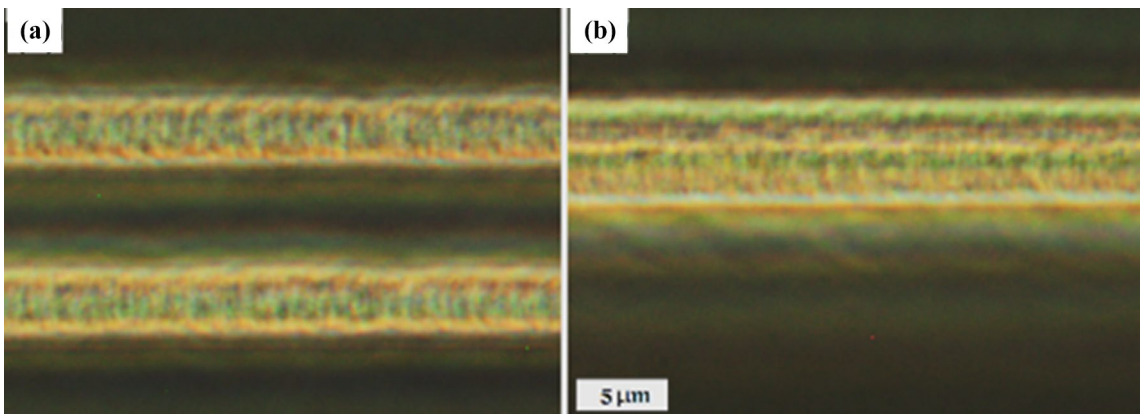

Fig. 3 PCM images of the two lines inscribed by H-polarized pulse trains with energy of 1 µJ per pulse with transverse separation of **a** 6 µm, **b** 2 µm

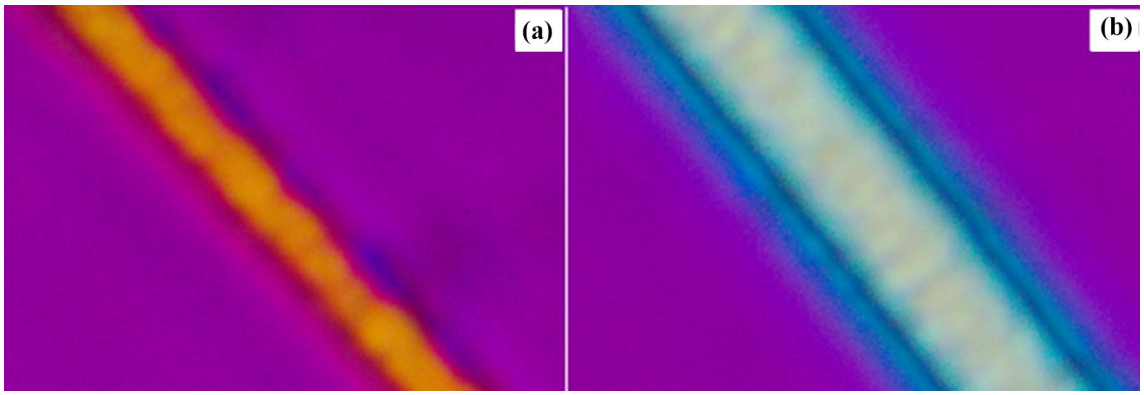

Fig. 4 PM image of written line by perpendicularly polarized simultaneous two pulse trains **a** energy of 0.5 μJ per both polarized pulses and **b** 1 μJ per H-polarized and 0.5 μJ per V-polarized pulse

tilted pulse front. Therefore, when the sample is irradiated with temporally overlapped pulses from two orthogonally polarized unidirectional beams the electron plasma is dominantly generated by H-polarized pulses. But, the orientation of generated plasma is determined by the interference between the incident wave and the inhomogeneity-scattered waves of V-polarized pulse train which are more intense due to its lower absorption inside the material (Fig. 4a).

To validate such analysis more, another experiment is performed. It is observed that with increasing the energy of the H-polarized pulse train to 1 μJ (2 times higher than that of the V-polarized pulse train) the measured retardation value increases to + 270 nm that is greater than that generated by a single H-polarized pulse train with the same energy (Fig. 2a). It seems that the density of the inhomogeneities created by the leading edge of the H-polarized pulse is greater than that of previous situation (0.5 μJ per pulse) and it leads to reduce the threshold energy for electron plasma generation by V-polarized pulses, and hence, to increase the excited electron density. Under this condition, the more intense field at the trailing edge of the H-polarized pulse orients the electron plasma planes and thus, the orientation of the nanogratings is dominantly determined by H-polarized pulses (Fig. 4b).

4 Conclusion

In summary, our results illustrate that birefringence induced by a linear polarized pulse train radiation can be erased by the irradiation of another pulse train having perpendicular polarization. However, if the sample is simultaneously irradiated by two perpendicularly polarized pulse trains, the distribution of the induced birefringence will be very different. Although the former setup results in depletion of induced birefringence, the later results in enhancement. Our results illustrate that the arrangement of writing, energy ratio of two perpendicularly polarized writing pulse trains and the spatial separation between written regions offer degrees of freedom to produce desired induced birefringence distribution inside the glass. We will examine more about the effect of these different parameters on the transition process of the orientation of nanogratings in our future research.

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