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Influence of self-focusing effect on femtosecond collinear nondegenerate optical Kerr measurements

Lihe Yan, Sen Jia, Jinhai Si*, Feng Chen, Wenhui Yi, Xun Hou

Key Laboratory for Physical Electronics and Devices of the Ministry of Education and Shannxi Key Lab of Information Photonic Technique, School of Electronics and information Engineering, Xi'an Jiaotong University, Xianning-xilu 28, Xi'an, 710049, China

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ABSTRACT

We performed the femtosecond nondegenerate optical Kerr shutter (OKS) measurements in a 10-mm-thick optical glass. The time-resolved OKS signals showed an obvious asymmetry and a large broadening with respect to that for thin samples. The time-resolved OKS signals in samples with different thicknesses showed that, the temporal broadening of the OKS signals was attributed to the group velocity mismatch between pump and probe beams of different wavelengths. By imaging the filament induced inside the glass, we attributed the asymmetry of the time-resolved OKS signals to the focusing position change of pump beam caused by self-focusing and re-focusing effect.

1. Introduction

Using femtosecond laser pulses, nondegenerate pump-probe measurements have been widely used to investigate the ultrafast dynamic process of a great variety of materials and systems [1–4]. Among all these techniques, optical Kerr shutter (OKS) technique has been developed as a key tool for studying the nonlinear optical properties of materials, due to their precision, sensitivity, easy operating, and etc. [5–7]. Moreover, femtosecond OKS technique has been widely used in investigating the ultrafast fluorescence spectroscopy, high time-resolved imaging, ballistic light imaging, and etc. [8–10].

When femtosecond two-color pump-probe OKS experiments are performed in a Kerr medium, however, two factors might influence the experimental results. On one hand, the refractive index difference between the two incident beams will cause the group velocity mismatch, and affect the overlap of the signal and gating pulses in a Kerr medium [11]. On the other hand, self-focusing effect and re-focusing effect might take place due to optical Kerr effect caused by the high peak power of the femtosecond laser pulses [12–17]. This effect might change the focusing position of incident beams inside the sample. Both these two effects will influence the temporal behavior of the OKS signals, and deteriorate the performance of an OKS configuration.

In this paper, we performed the femtosecond collinear nondegenerate OKS experiments in a 10-mm-thick glass. The time-resolved OKS signals showed an unexpected asymmetry and a large broadening, which was estimated to be more than 2.5 ps. By measuring the temporal behavior of the OKS signals in glasses with different thickness, we found that the broadening of the signal widths was attributed to the group velocity mismatch between the two-color incident beams. The recorded images of the focal volume showed that, the focal point change of the pump beam induced by self-focusing and refocusing effect caused the asymmetry of the time-resolved OKS signals.

2. Experiments

In our experiments, we used commercially available optical glass with dimensions of 15 mm × 15 mm × 10 mm. The chemical composition of the glass sample was Na_2O (12.34 wt%), SiO_2 (79.71 wt%), Al_2O_3 (1.56 wt%), and K_2O (6.39 wt%), as determined by energy dispersive X-ray spectroscopy (EDS) analysis. A femtosecond collinear two-color pump-probe OKS arrangement was employed. The output of 1 kHz, 30 fs, and 800 nm femtosecond laser was split into two beams, one of which was used as the pump beam. The other beam was passed through a BBO crystal to generate second harmonic as the probe beam. The probe beam and temporally delayed pump beam were collinearly incident into an achromatic lens (f = 150 mm) and focused into the 10-mm-thick glass perpendicularly to the incident surface. The sample was fixed on a stage, which was movable along the optical axis of the incident light. The filaments formed inside the glass were imaged from the side by a charge coupled device (CCD) camera. To stop the pump beam and white-light continuum induced in the sample, a 20 nm band-pass filter centered at 400 nm was inserted before the detector. To

* Corresponding author.
E-mail address: jinhaisi@mail.xjtu.edu.cn (J. Si).

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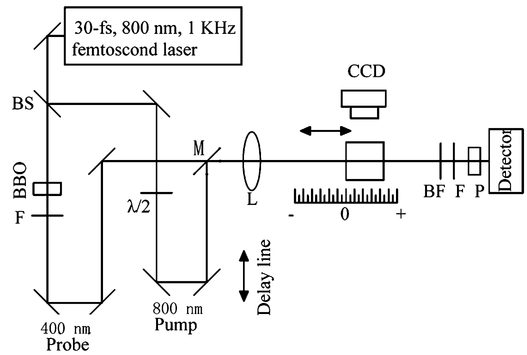

Fig. 1. Schematic diagram of the experimental setup. BS: beam splitter, F: filter, M: dielectric mirror, L: convex lens, BF: bandpass filter, P: polarizer.

optimize the OKS signals, the polarization plane of the pump beam was rotated by 45° with respect to that of the probe beam [18] (Fig. 1).

3. Results and discussion

3.1. Temporal broadening and asymmetry of time-resolved OKS signals in thick mediums

Fig. 2(a) shows the time-resolved OKS signals measured in the 10-mm-thick glass when the sample was fixed at different positions and the pump power was fixed at 3 mW. Here, we define d as the distance between the incident surface of the sample and the focal point of the lens, while the value was negative inside the focal length and positive outside. The solid squares in the figure show the time-resolved OKS signals for a 1-mm-thick glass, which showed a symmetric response with the full-width at half-maximum (FWHM) estimated to be less than 240 fs. Comparing with that for the thin sample, the time-resolved OKS signals for the thick sample showed an abnormal broadening, and the rising-edge occurred when the probe pulse was about 2.5 ps before the pump pulse. Moreover, the time-resolved signals showed unexpected asymmetry, and the peak value of the OKS signals did not take place at zero delay time again.

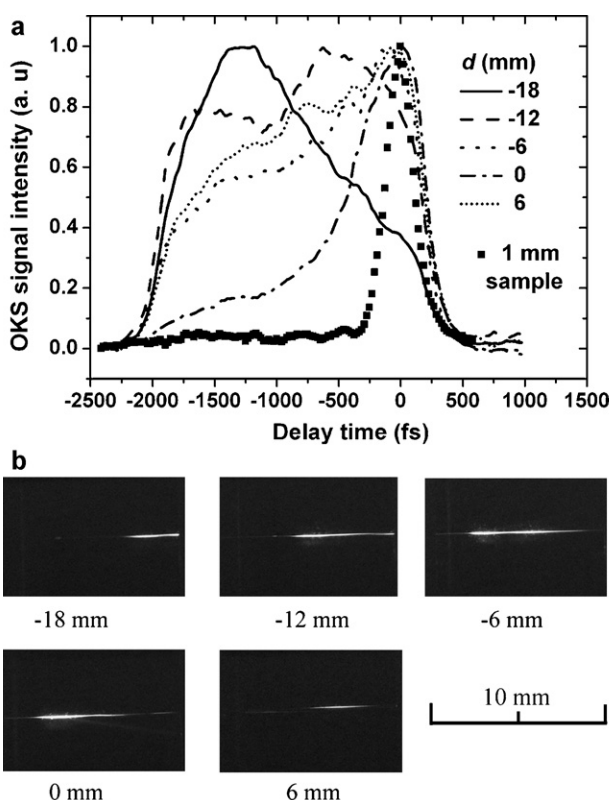

Fig. 2. (a) Time-resolved two-color OKS signals, and (b) recorded images of the formed filament in the sample, when the 10-mm-thick sample was fixed at different positions. The pump power was fixed at 3 mW.

As shown by the solid line in Fig. 2(a), in which the value of d was adjusted to -18 mm, the peak value of the time-resolved OKS signals occurred when the probe pulse arrived at the sample about 1.3 ps before the pump pulse. The dashed, dotted, and dash-dotted lines show the time-resolved OKS signals when the value of d was adjusted to -12 mm, -6 mm, and 0, respectively. Fig. 2(b) shows the recorded images of the filament formed in the sample, when the distance between the sample and the focal point was adjusted to the corresponding values. From the figures we can see that, when the value of d increased from -18 mm to 0, the focal volume moved from the back surface to the incident surface of the sample because of self-focusing effect induced inside the sample; meanwhile, the peak value of the time-resolved signals shifted from negative delay time to zero-delay time as shown by the curves in Fig. 2(a). However, when the incident surface of the sample was moved 6 mm behind the focal point, the beam re-focused after passing about 5 mm inside the sample due to the self-focusing effect. Correspondingly, the OKS signals intensity at negative delay time was enhanced again, as shown by the short dotted line.

In our experiments, the two beams were collinearly incident into the sample, so the overlapping region was throughout the whole propagating path inside the glass and made contributions to the OKS signals. Because of the group velocity mismatch inside the material, however, the probe pulse would be temporally delayed by $\Delta n_G L_s / c$ after passing through the sample with respect to the simultaneously incident 800 nm pump pulse. Here, Δn_G is the group refractive index difference between the two pulses, and L_s is the length of the sample. c denotes the light velocity in vacuum. In other words, if the probe pulse was incident into the sample before the pump pulse, they might be overlapped after passing a certain distance inside the sample and make contributions to the OKS signals. In the collinear nondegenerate OKS measurements, therefore, the width of the time-resolved signals will be broadened by $\Delta n_G L_s / c$ because of the group velocity mismatch between the pump and probe pulses. Considering that the intensity of OKS signals is proportional to the square of pump laser intensity [18], the focal volume located at a certain position inside the sample, where the light intensity is much higher, would cause the enhancement of the OKS signals at the corresponding delay time. The shift of the focal region which resulted from self-focusing and refocusing effect, therefore, will influence the temporal behavior of the OKS signals remarkably.

3.2. Influence of the group velocity mismatch between the pump and probe beams

For further understanding of the asymmetry and broadening of the OKS signals, we investigated the temporal behaviors of the OKS signals for samples of different thicknesses. The curves in Fig. 3 show the time-resolved OKS signals for different thickness glasses. Here, the incident beams were focused on the incident surface of the samples and the pump power was also kept at 3 mW. From the figure we can see that, when the sample thickness was increased the rising-edge of the time-resolved signals shifted forwards proportionally. In our experiments, the group refractive indexes of the glass for 400 nm and 800 nm pulses were measured to be 1.612 and 1.534, respectively. Substituting the group refractive index difference by 0.078, the rising-edge of OKS signals will be shifted

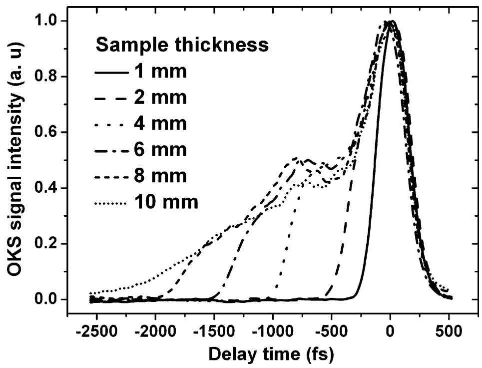

Fig. 3. Time-resolved two-color OKS signals for glasses with different thicknesses.

forwards by 260 fs for each additional millimeter of the sample thickness. The width of the time-resolved OKS signals for the 10-mm-thick glass is estimated to be about 2.6 ps, agreeing well with the experimental results shown in Figs. 2 and 3.

3.3. Influence of the self-focusing effect on the time-resolved OKS signals

Fig. 4(a) shows the time-resolved OKS signals for different pump powers, when the focal point was located at 1 mm before the back surface. The solid, dashed, dotted, and dash-dotted lines show the OKS signals when the pump power was adjusted to be 1.2 mW, 2.4 mW, 3.6 mW, and 5.0 mW, respectively. Fig. 4(b) shows the recorded images of the focal volumes formed inside the glass for the corresponding pump power. When the pump power was fixed at 1.2 mW, the focal volume was adjacent to the back surface, so the peak value of the OKS signals occurred on the rising-edge of the signal. With increasing the pump power, the focal volume moved to the incident surface due to the self-focusing effect. As mentioned above, the overlapping volume at different position inside the glass would contribute to the OKS signals at the corresponding delay time. So the main components of the envelope shifted to the

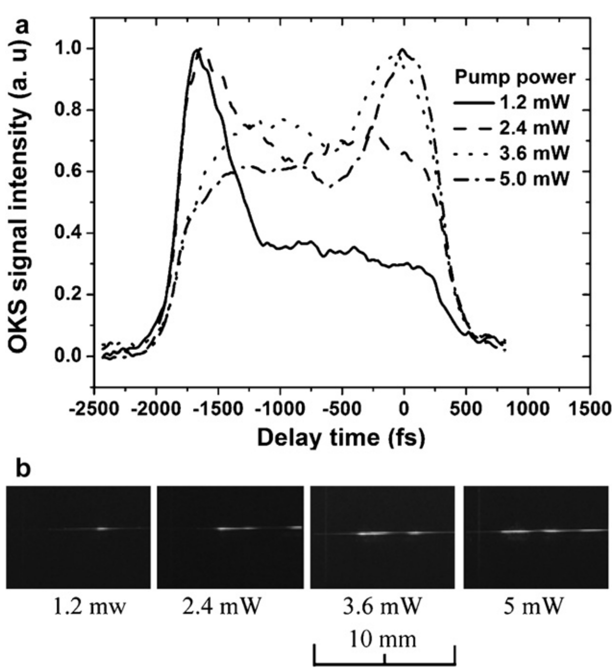

Fig. 4. (a) Time-resolved two-color OKS signals for different pump powers, and (b) recorded images of the formed filament in the 10-mm-thick glass.

direction of zero-delay time, when the focal volume moved to the incident surface of the sample.

When a femtosecond laser pulse propagates inside a nonlinear material, some other nonlinear effects, such as self-phase modulation, cross-phase modulation, pulse splitting effect etc, might happen during the self-focusing [12,13,19,20]. All these effects might modify the envelope and duration of the pulse, and change the temporal behavior of the OKS signals. To exclude the influence of the mentioned effects on the laser pulse, we performed the non-collinear OKS measurements to probe the pulse change during self-focusing. After passing through the beam splitter, the 800 nm probe beam was focused into a 10-mm-thick glass using a 150-mm lens. The probe beam was self-focused inside the glass, and the light was collimated by a confocal lens and then focused into a 1 mm thick glass sample with the pump beam at an interaction angle of 10°. To stop the white light, a 20 nm bandpass filter centered at 800 nm was inserted before the detector. The measured time-resolved OKS signal was compared with that without self-focusing configures. The results showed that no obvious distortion of the laser pulse was observed, indicating that the influence of above-mentioned effects can be excluded in our experiments.

4. Conclusion

In summary, we have investigated the femtosecond collinear nondegenerate OKS signals in a 10-mm-thick optical glass. The time-resolved OKS signals showed obvious asymmetry and a large broadening with respect to that for thin samples. By measuring the OKS signals in samples with different thicknesses, we demonstrated that the shifting forwards of the rising-edge of OKS signals was attributed to the group velocity mismatch between pump and probe beams. By imaging the filament induced in the glass, we attributed the asymmetry of OKS signals to the change of focusing position of the incident beams which caused by self-focusing and re-focusing effect.

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