Pump power dependence of Kerr signals in femtosecond cross pump-probe optical Kerr measurements

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Abstract: We investigated the influence of self-diffraction effect on femtosecond cross pump-probe optical Kerr shutter (OKS) measurements for fast response and slow response materials, respectively. In the slow response material of CS_2, a fast response resulted from self-diffraction effect was observed in the time-resolved OKS signals, and the signal intensity showed different pump power dependences at different delay time. For the fast response material of bismuth oxide (BI) glass, the pump power dependence of OKS signals varied when the polarization angle was varied. The pump power dependences of OKS signals for the both materials showed that, the OKS signals resulted mainly from laser induced birefringence effect at low pump powers, and the contribution of self-diffraction effect to OKS signals was enhanced with increasing the pump power.

OCIS codes: (190.4400) Nonlinear optics, materials; (190.3270) Kerr effect; (190.7110) Ultrafast nonlinear optics; (320.7100) Ultrafast measurements

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1. Introduction

For the last few decades, femtosecond optical Kerr shutter (OKS) technique has been proven to be one of the most convenient nonlinear optical (NLO) measurements due to its precision, easy operating, sensitivity, and etc [1]. Using the femtosecond laser and OKS technique, much effort has been done to investigate the optical nonlinearities and ultrafast responses of many materials, e.g. liquids and gases [2,3], inorganic glasses [4,5], organic or polymeric materials [6], semiconductors [7], and glasses doped with nanocrystallites or metals [8,9], etc.

In the optical Kerr effect (OKE), the electronic field of light incident on a transparent sample induces an anisotropic index, and a phase shift occurs between the probe filed components polarized parallel and perpendicular to the polarization plane of the pump beam. Such photoinduced birefringence effect will cause the depolarization of the probe beam and the transmission through the crossed analyzer behind the sample. When the pump and probe beams with equal wavelength are used in the cross pump-probe OKS experiments, however, the superposition of the two coherent pulses might yield a spatially modulated distribution of the energy density, and the interaction with the material leads to the creation of laser-induced transient gratings (LITG). If the laser pulse duration is comparable or shorter than the response time of nonlinear medium, energy transfer between two beams by self-diffraction effect induced by LITG will take place [10]. Some studies have shown that when femtosecond pulses were used in OKS experiments for some organic molecular systems and noble-metal nanoparticle systems, Kerr signals resulted mainly from transferred pump intensity into the weaker probe beam by LITG [6,8]. In the process, the interference of the pump and probe beams leads to LITG, so the temporal behavior of the self-diffraction effect due to the LITG, which is dependent only on the filed correlation time of the two incident pulses, does not reflect the NLO response time of the materials. In femtosecond OKS measurements, therefore, separating the self-diffraction effect from the photoinduced birefringence effect is necessary for measuring the NLO response of the materials.

In this paper, we investigated the influence of self-diffraction effect on femtosecond optical Kerr shutter (OKS) measurements for CS_2 and bismuth oxide (BI) glass, the nonlinear response of which showed a slow and a fast response, respectively. In CS_2, a fast response resulted from self-diffraction effect was observed in the time-resolved OKS signals, and the signal intensity showed different pump power dependences at different delay time. For BI glass, influence of self-diffraction effect was not able to be distinguished from the OKS effect, as both effects showed ultrafast responses in the glass. By measuring the pump power dependence of OKS signals in BI glass we find that, the dependence varied when the polarization angle between the pump and probe beams was different. The pump power dependences showed that, the OKS signals resulted mainly from laser induced birefringence effect at low pump powers, and the contribution of self-diffraction effect to OKS signals was enhanced with increasing the pump power.

2. Experiments

The glass sample of the composition Bi_2O_3-B_2O_3-SiO_2 prepared by melting method was employed. The raw material of it contained 60%-$BiO_{1.5}$, 20%-SiO_2, 20%-B_2O_3 and 0.15%-CeO_2 (mol. %). The melted mixtures of the raw materials were poured onto a stainless-steel plate at room temperature to remove strain in the glass. The linear absorption spectrum of the sample indicated that the edge of absorption band of the glass sample located at 450 nm [11]. A Ti: sapphire laser, emitting 30-fs and 800 nm laser pulses at a repetition rate of 1 kHz, was employed in our non-collinear pump-probe arrangement. We used a part of the output of the laser as the probe beam, and the rest as the pump beam. The power of the probe beam was kept at 0.1 mW, and the pump power was varied below 5 mW. Delay time between

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the two pulses was varied by a delay-line controlled by a stepping motor. To detect the energy transfer from the pump beam to the probe beam by self-diffraction effect, a photomultiplier tube was positioned behind the sample on the optical path of the probe beam. A half-wave plate was introduced into the optical path of the pump beam, which was used change the angle between the polarization planes of the pump and probe beams.

In our experiments, the pump and probe beams were focused into the sample at an angle of 15° by two lenses with focal lengths of 150 mm and 200 mm, respectively. To avoid the influence of self-focusing effect on the overlapping of the two beams, the sample was polished to be about 1.5 mm thick, and the two spots of the focused beams were carefully overlapped in the sample. To avoid the white light generation, the sample was placed about 5 mm behind the focal points of the beams, where the spot diameter at the sample surface of the pump beam was estimated to be about 200 μm, being about twice larger than that of the probe beam. As mentioned in our previous report, the nonlinear refractive index of the BI glass was estimated to be about 3×10^{-15} cm^2/W [12]. According to A. Brodeur's reports, the threshold of white light generation for BI glass was estimated to be about 1.2×10^6 MW/ cm^2, while that for CS_2 was even higher [13]. In our experiments, the pump power was kept below 5 mW, while the energy density was estimated to be below 0.53×10^6 MW/ cm^2. Hence, the influence of white light generation on our experimental results was excluded.

When we measured the OKS signals induced in the sample, an analyzer perpendicular to the polarization plane of the probe beam was positioned in front the detector. In the OKS effect, the observed intensity depends on the induced phase shift $\Delta\phi$ and the angle θ between the polarization planes of the pump and probe beams according to [14]

$$I = I_0 \sin^2(\Delta\phi / 2)\sin^2(2\theta) . \quad (1)$$

Using the effective length of the sample L_{eff} and the wavelength of the probe beam λ_p, the phase shift $\Delta\phi$ is given by

$$\Delta\phi = \frac{2\pi n_{2B(Kerr)} L_{eff}}{\lambda_p} I_g . \quad (2)$$

Here, $n_{2B(Kerr)}$ corresponds to the third-order nonlinear coefficient, and I_g is the intensity of the pump beam.

3. Results and discussion

3.1 Pump power dependence of OKS signals in slow response materials

Figure 1(a) shows the normalized time-resolved OKS signals in CS_2 when the pump power was fixed at different values. The solid line in the figure shows the time-resolved OKS signal when the pump power was adjusted to 1 mW. As the nonlinear response of CS_2 was attributed to the reorientation of molecules, it has a slow response lasting more than 1 ps [2]. The dashed, dotted and dash-dotted lines in the figure show the time-resolved OKS signals, when the pump power was fixed at 2 mW, 3 mW and 4 mW, respectively. From the figure, we can see that the rising edge of the OKS signals became shaper with increasing the pump power.

The distortion of the time-resolved OKS signals in CS_2 was attributed to the self-diffraction effect resulted from the LITG effect. As shown by Fig. 1(b), the dashed and solid lines show the self-diffraction and OKS signals in CS_2, respectively. Here, the pump power was kept at 5 mW. The full-width at half-maximum (FWHM) of the time response of the self-diffraction signal was estimated to be about 105 fs, which was dependent only on the filed correlation time of the two incident pulses. At the sample, the 30-fs pulses would broaden after passing through the optical mediums. The temporal behavior of the OKS signals at 5mW shows that, the OKS signals consisted of two components: a fast response and slow response, which was attributed to the self-diffraction effect and laser induced birefringence effect, respectively. In addition,

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from Fig. 1(a) and (b), we can see that the onset of the OKS signal appears around −200 fs, which is attributed to the broadened pulses at the sample. The interaction of the trailing edge of the probe beam with the leading edge of the pump beam makes the onset of the OKS signals much earlier than the zero-delay time.

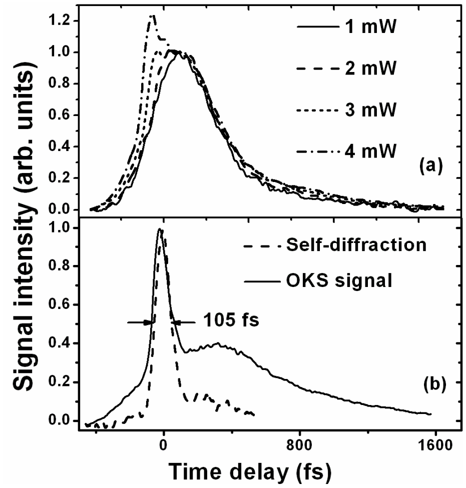

Fig. 1. Time-resolved signals of CS_2. (a), OKS signals when the pump power was kept at 1 mW, 2 mW, 3 mW, and 4 mW, respectively. (b), OKS and self-diffraction signals when the pump power was kept at 5 mW.

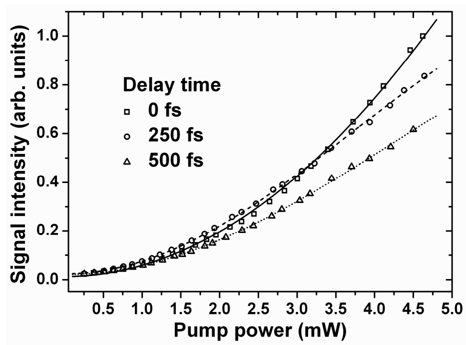

Fig. 2. Pump power dependence of OKS signals in CS_2 when the delay time between the pump and probe pulses was kept at 0 fs, 250 fs, and 500 fs, respectively.

For the further investigation of the influence of self-diffraction effect on the femtosecond OKS measurements, we measured the pump power dependence of the OKS signals in CS_2, when the delay time between the pump and probe pulses was adjusted to different values. As shown by Fig. 2, the squares and circles indicate the pump power dependence of OKS signals when the delay time was kept at 0 fs and 250 fs, respectively. From the figure, we can see that when the pump power was kept at a low power, the signal intensity at 0 fs was lower than that at 250 fs. However, the signal intensity at 0 fs increased more rapidly with increasing the pump power, because of the contribution of self-diffraction effect in the OKS signals. When the pump power was increased more than 3 mW, the signal intensity at 0 fs exceeded that at 250 fs, indicating that self-diffraction effect played a more important role in the OKS signals at 0 fs when the pump power was kept at a high value. The tangles in Fig. 2 show the OKS signals as a function of the pump power when the time delay was adjusted to 500 fs. The signal intensity was proportional to that of 250 fs despite of that the pump power was increased, indicating that

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the self-diffraction effect influenced the OKS signals only when the delay time was kept around 0 fs.

3.2 Pump power dependence of OKS signals in ultrafast response materials

For ultrafast response materials due to the electronic process, such as BI glasses, their response time is much shorter than the pulse width of the femtosecond laser, so the temporal behavior of the OKS signals is limited by the pulse duration of laser pulses [8,10]. Therefore, it was impossible to separate the contributions of the self-diffraction and birefringence effects in BI glass using the time-resolved OKS technique described above. The dashed and solid lines in Fig. 3 show the experimental results for the temporal behaviors of self-diffraction signals and OKS signals of BI glass, respectively. Both the FWHMs of the self-diffraction and OKS signals of BI glass were estimated to be less than 100 fs.

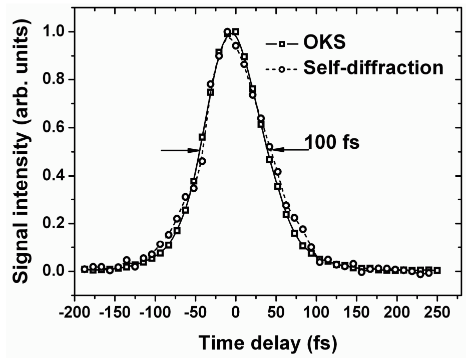

Fig. 3. Time-resolved measurements of the self-diffraction and OKS signals of BI glass.

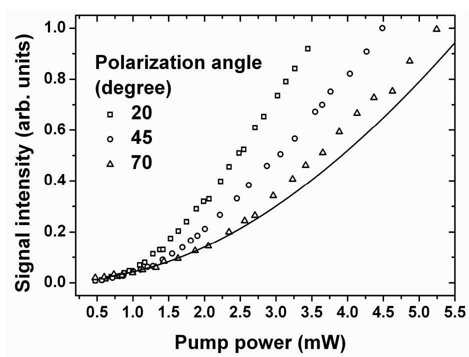

Fig. 4. Pump power dependences of OKS signals of BI glass at different polarization angles.

In order to investigate the influence of the self-diffraction on OKS signals of BI glass, we measured the pump power dependence of OKS signals at different polarization angles. The squares, circles and triangles in Fig. 4 show the measured results when the polarization angle between the pump and probe beam was kept at 20°, 45° and 70°, respectively. Here, the pump power was varied between 0.4 mW and 5.5 mW, being kept below the damage threshold of sample. The solid line in Fig. 4 indicates the pump power dependence of OKS signals, which is the fitted result for OKS signals measured at low powers using Eqs. (1) and (2). From the figure, we can see that when the pump power was kept below 2 mW, the pump power dependence of OKS signals agreed well with the calculated curve, and the OKS signals was mainly originated from the photoinduced birefringence effect. When the pump power was further increased the signals deviated from the theoretical curve, and the OKS signals showed different pump power dependences at different polarization angles.

To confirm that the pump power dependence of OKS signals measured at high powers should be mainly attributed to the self-diffraction effect, we measured the dependence of OKS signals on the polarization angle between the pump and probe beams. The solid fitted line in Fig. 5 shows the dependence of OKS signals on the polarization angle between pump and probe beams. Here, to avoid the influence of the self-diffraction effect, the pump power was kept at a low value of about 1 mW. The fitted line shows that the maximum and the minimum values occur at $\pi/4+n\pi/2(n=0,1,2,\ldots)$ and $\pi/2+n\pi/2(n=0,1,2,\ldots)$, respectively. The fitted dashed line in Fig. 5 shows the polarization dependence of self-diffraction effect in BI glass. The maximum and minimum values of the self-diffraction signals occur at $n\pi(n=0,1,2,\ldots)$ and $\pi/2+n\pi(n=0,1,2,\ldots)$, respectively. From the figure we can see that when the polarization angle decreases from 45°, the self-diffraction effect enhances and the OKS signals related to the photoinduced birefringence decrease. Therefore, when the polarization angle was adjusted to 20°, the proportion of self-diffraction to OKS signals was larger than that of 45° and the OKS signals increased more rapidly with increasing the pump power. When the polarization angle was adjusted to 70°, the contribution of self-diffraction effect became smaller and the experimental data for the signals agreed better with the fitting curve for the OKS signals.

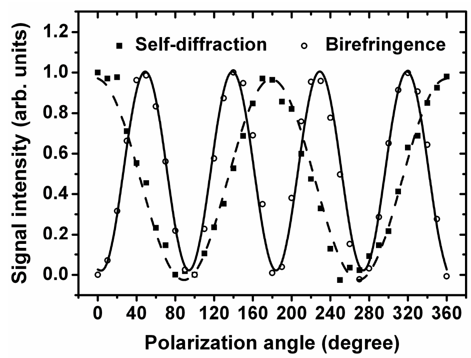

Fig. 5. The dependence of self-diffraction and OKS signals on the polarization angle between the pump and probe beams.

4. Conclusion

In summary, we demonstrated the self-diffraction effect on femtosecond in pump-probe optical Kerr shutter (OKS) signals for fast response and slow response materials, respectively. In the slow response material of CS_2, a fast response resulted from self-diffraction effect was observed in the time-resolved OKS signals, and the signal intensity showed different pump power dependences at different delay time. For the fast response material of bismuth oxide (BI) glass, the pump power dependence of OKS signals varied when the polarization angle between the pump and probe beams was varied. The pump power dependences of OKS signals for the both materials showed that, the OKS signals resulted mainly from laser induced birefringence effect at low pump powers, and the contribution of self-diffraction effect to OKS signals increased with increasing the pump power.

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