Influence of self-diffraction effect on femtosecond pump-probe optical Kerr measurements

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Abstract: We investigated influence of the self-diffraction effect on optical Kerr signals of Bi_2O_3-B_2O_3-SiO_2 glass (BI glass) and CS_2 in femtosecond non-collinear pump-probe optical Kerr experiments. By measuring the dependence of the Kerr signals on the pump power and on the polarization angle between pump and probe beams, we found that the optical Kerr signals of CS_2 consisted of two components: a fast response and a slow response, which were attributed to the self-diffraction effect and the photoinduced birefringence effect, respectively. The contribution of the self-diffraction effect to the Kerr signals increased with increase of the pump power. For the BI glass, no evident influence of self-diffraction on the Kerr signals was observed.

OCIS codes: (190.4400) Nonlinear optics, materials; (190.3270) Kerr effect; (190.7110) Ultrafast nonlinear optics; (320.2250) Femtosecond phenomena; (320.7100) Ultrafast measurements.

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1. Introduction

For the last few decades, research of ultrafast all-optical switch has been stimulated by its high-bit-rate handling of optical signals for the need of future integrated optical system [1-3]. As the two main parameters characterizing the optical switching properties of materials, a large third-order nonresonant optical nonlinearity and a fast response are the most important properties for the realization of such all-optical switches. Using the optical Kerr shutter (OKS) technique and a femtosecond laser, many efforts have been invested in achieving these goals in many materials, e.g. inorganic glasses [4-6], organic or polymeric materials [7-9], semiconductors [10,11], and liquid materials [12].

Optical Kerr shutter based on femtosecond non-collinear pump-probe technique can be used to measure nonlinear optical (NLO) response of materials. When the pump and probe beams with equal wavelength are used in the non-collinear pump-probe OKS experiments, optical Kerr signals usually originate from two kinds of effects: the photoinduced birefringence effect [7] and self-diffraction that results from laser-induced transient gratings (LITG) [8, 9]. Some studies have shown that when femtosecond pulses are used in OKS experiments for some organic molecular systems [8, 9], Kerr signals result mainly from self-diffraction of the pump beam by the LITG. In the process, self-diffraction, which is a four-wave-mixing process, is only limited by the duration of the pump laser pulses, even when nonlinear materials showing slow response are used, so the temporal behavior of the Kerr signals measured using the OKS setup is the field correlation time of the two pulses between the pump and probe beams, and not the NLO response time of the materials. Therefore, it is important to investigate the influence of the self-diffraction effect on femtosecond optical OKS measurements.

In this paper, we investigated the self-diffraction and optical Kerr effects of Bi_2O_3-B_2O_3-SiO_2 glass (BI glass) and CS_2 using the femtosecond non-collinear pump-probe OKS technique. By measuring the dependence of the Kerr signals on the pump power and on the polarization angle between pump and probe beams, we found that the optical Kerr signals of CS_2 consisted of two components: a fast response and a slow response, which were attributed to the self-diffraction effect and the photoinduced birefringence effect, respectively. By varying the pump power, we were able to control the influence of the self-diffraction effect on the Kerr signals. For the BI glass, no evident influence of self-diffraction on the Kerr signals was observed. This is probably because the optical nonlinearity of BI glasses arose from the electronic polarization process whose response time is shorter than the pulse duration.

2. Experiment

The glass sample of the composition Bi_2O_3-B_2O_3-SiO_2 prepared by melting $BiO_{1.5}$, SiO_2, B_2O_3 and GeO_2 according to a certain proportion was employed. The details about the preparation of the sample were given in the reference [5]. The thickness of the sample was about 1.5 mm. The linear absorption spectrum of the sample indicates that there is no evidence absorption above

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the wavelength of 450 nm, so there is no resonance absorption at the wavelength of 800 nm.

In our experiments, a femtosecond non-collinear pump-probe arrangement was employed. The output of a Ti: sapphire laser, which emitted 30-fs and 800 nm laser pulses with a repetition rate of 1 kHz, was split into two beams. The power of the pump beam varied between 1 mW and 8 mW, being kept below the threshold of white light generation [13]. For the sake of a large intensity ratio of the diffracted pump beam to the probe beam as well as a large signal-to-noise ratio (SNR), the probe beam was much weaker, 5% of the pump power for instance. The two beams were focused into the sample at an angle of 15° and the spots of the focused beams overlapped carefully. Time delay between the two pulses was varied by a delay-line controlled by a stepping motor. A photomultiplier tube was positioned behind the sample on the optical path of the probe beam to detect the self-diffraction signals. When we measured the Kerr signals of the sample, an analyzer was positioned in front the detector. To investigate the dependence of the self-diffraction signals as well as the Kerr signals on the polarization angle between the two beams, a $\lambda/2$ wave plate was introduced into the optical path of the pump beam. To optimize the intensity of the Kerr signals, the polarization plane of the pump beam was rotated by $\pi/4$ from that of the probe beam.

When a laser beam with an ultrashort laser pulse such as a femtosecond pulse is focused, the light intensity I might cause a refractive index change of the sample, as given by the following equation [14]:

$$n = n_0 + n_2 I. \tag{1}$$

Here, n_0 is the linear refractive index and n_2 denotes the nonlinear refractive index, which corresponds to the third-order nonlinear coefficient. When the probe pulse overlaps with the pump beam temporally, it will take the anisotropy of the refractive index, causing a partial depolarization of the beam and a leakage of the intensity through the analyzer. On the other hand, the spatial superposition of the two coherent lasers might yield a spatially modulated distribution of the energy density, and the interaction with the material will lead to the creation of LITG and the self-diffraction of the pump beam [15-20].

3. Results and discussion

3.1 Influence of self-diffraction effect on time-resolved Kerr signals

Figure 1 shows the results measured for the time response of the self-diffraction signals and the Kerr signals of CS_2. The pump power for self-diffraction effect and Kerr effect was fixed at 5 mW and 1 mW, respectively. 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 much faster than the decay process (more than 1 ps) of the Kerr signals. Since the response of the self-diffraction effect depended only on the field correlation time of the two pulses, the response of the process was limited only by the duration of the femtosecond pulses at the sample, which was broadened to about 100 fs due to the group velocity dispersion of the 30-fs laser pulses in the optical path. The slow decay process of the Kerr signals of CS_2 originated from the orientation of the molecules [21].

Figure 2 shows the time-resolved measurements of Kerr signals of CS_2 for different pump powers. The polarization angle between the pump and probe was kept at 45°. At low pump power which was smaller than 1 mW, there was no evident distortion of the time-resolved Kerr signals, as shown by the solid line. With increase of the pump power, the rising edge of the Kerr signals became sharper, as shown by the dotted and short dashed curves in Fig. 2, in which the pump power was fixed at 3 mW and 4 mW, respectively. The pump dependence of the time response of the Kerr signals was attributed to the self-diffraction of the pump beam. The self-diffraction and optical Kerr effects had different time and pump dependence, so when the pump power became stronger, the fast component of the Kerr signals that was attributed to the contribution of the self-diffraction to the Kerr signals became more evident. In order to confirm it, we measured the time-resolved Kerr signals for different polarization angles, in which the

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pump power was adjusted to 5 mW. The results are shown in the inset of Fig. 2, from which we can see that when the polarization angle between the pump and probe beams was adjusted to 0°, the Kerr signal showed a fast time-response that arose mainly from the self-diffraction of the pump beam. When the polarization angle was adjusted to 30° or 60°, the Kerr signals showed two components of a fast response and a slow response, which were attributed to the self-diffraction and the photoinduced birefringence, respectively.

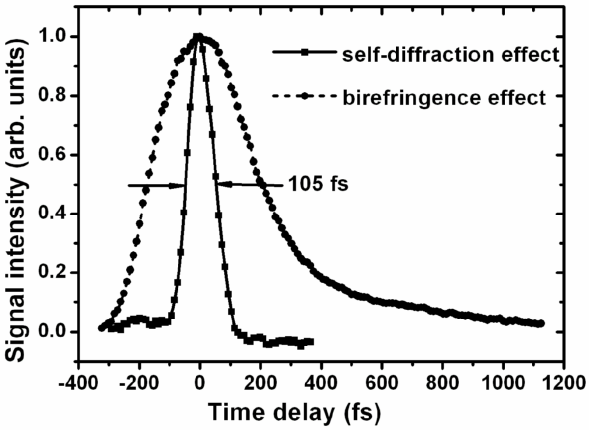

Fig. 1. Time dependence of the self-diffraction effect and Kerr effect measured in CS_2, as shown by the solid and short dashed line respectively. The FWHM of the self-diffraction signal is estimated to be less than 105 fs, depending only on the pulse width.

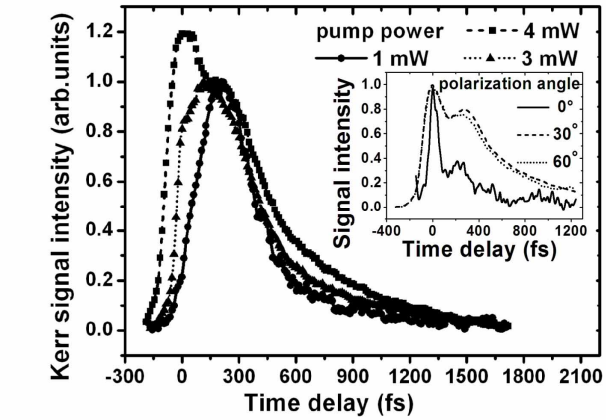

Fig. 2. Time-resolved measurements of Kerr signals of CS_2 for different pump powers. The polarization angle between the pump and probe beams was kept at 45°. The inset shows the time dependence of Kerr signals for different polarization angles. The pump power was 5 mW.

To compare the Kerr signals of CS_2 with that of NLO materials showing a fast response, we measured time-resolved Kerr signals of BI glass for different pump power, and the results are shown in Fig. 3. The inset shows the time-resolved measurements of the self-diffraction for BI glass, where the FWHM of the time-resolved signals was estimated to be equal to the width of the laser pulses. As the nonlinearity of the BI glass originated mainly from the electronic process [5, 6], its response is much faster than CS_2 and there was no slow component in the NLO process. From Fig. 3, we can see that no evident difference for the time-resolved Kerr signals was observed when the pump power increased from 2 mW to 8 mW. The response time of the photoinduced birefringence that results from the electronic polarization process is shorter

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than the pulse duration, therefore influence of the self-diffraction on the Kerr signals couldn't be distinguished, even though there might be a self-diffraction component in the Kerr signals.

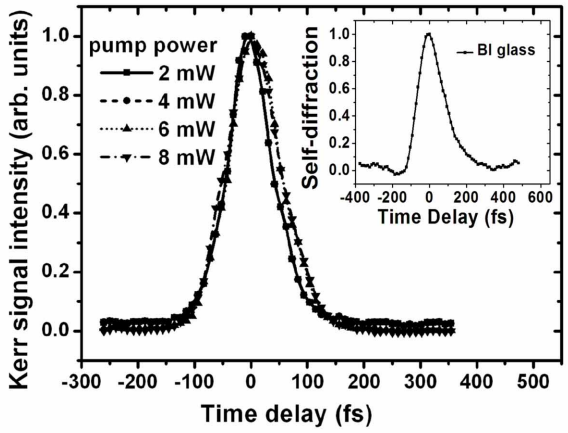

Fig. 3. Time-resolved Kerr signals of BI glass for different pump powers. The inset shows the time-resolved self-diffraction signals.

3.2 Polarization dependence of photoinduced birefringence and self-diffraction effects

In the previous reports, people have investigated the formation of the grating induced by a femtosecond laser [15-17]. It has been clarified that, the LITG is induced by the interference of the two coherent beams, and the mechanism is attributed to the transient refractive-index modulation. For the further investigation of the mechanisms of the Kerr signals, we measured the dependence of the self-diffraction signals on the polarization angle between the two incident beams. The polarization dependences of the signal intensities for both CS_2 and BI glass are depicted in Fig. 4. We can see from the figure that, the period of dependence is π, with the maximum values of the signals occurring at $k\pi$ ($k = 0,1,2,\cdots$), and the minimum values occurring at $k\pi + \pi/2$ ($k = 0,1,2,\cdots$). So, we can conclude that LITG is responsible to the self-diffraction of the pump beam.

To discriminate the mechanism of Kerr effect from that of self-diffraction effect, we also measured the polarization dependence of Kerr signals. Here, to avoid the contribution of self-diffraction effect to the Kerr signals, the pump power was adjusted to be below 1 mW. Figure 5 shows the dependences of the Kerr signals of BI glass and CS_2 on the polarization angle between the pump beam and probe beam. The experimental results show a sinusoid of the period of $\pi/2$, with the maximum and minimum values occurring at $n\pi/2 + \pi/4$ and $n\pi/2$ ($n = 0,\ 1,\ 2\cdots$), respectively.

In Kerr configurations related to photoinduced birefringence effect, an anisotropic refractive-index induced by the electric field of pump beam incident on a transparent sample causes a partial depolarization of probe beam so that Kerr signals are detected after the analyzer. In that case, the Kerr signal intensity is given by:

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

Here, θ is the intersecting angle between polarization of the probe and pump beam, $\Delta\phi$ is the phase shift. $\Delta\phi$ holds constant while the pump power is invariable, and the Kerr signal intensity depends only on the polarization angle θ. Comparing the results shown in Fig. 5, we can conclude that, the photoinduced birefringence effect was responsible for the Kerr signals in our experiments, which is different from the mechanism of self-diffraction effect.

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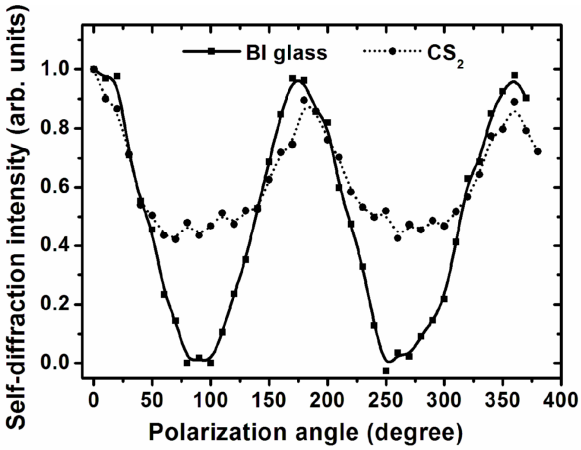

Fig. 4. Polarization dependence of the self-diffraction signals in BI glass and CS_2 induced by 30-fs pulses.

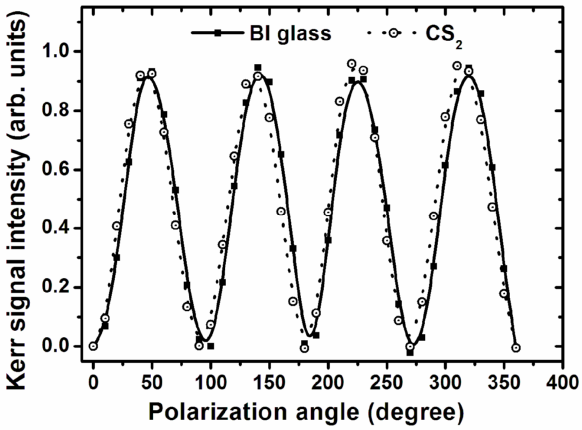

Fig. 5. Polarization dependence of the optical Kerr signals of BI glass and CS_2 induced by 30-fs pulses.

4. Conclusion

In summary, we have investigated the self-diffraction effect in both CS_2 and BI glass in femtosecond non-collinear pump-probe optical Kerr experiments. We have found that, there were two components in the Kerr signals of CS_2: a fast response that is attributed to the self-diffraction effect, and a slow response that is attributed to the photoinduced birefringence effect. By varying the pump power, we were able to control the contribution of the self-diffraction effect to the Kerr signals.

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