Polarization Dependence of Femtosecond Optical Kerr Signals in Bismuth Glasses

Lihe Yan, Juanjuan Yue, Jinhai Si, Sen Jia, Feng Chen, and Xun Hou

***Abstract*—Using the optical Kerr shutter (OKS) technique with an 800-nm femtosecond laser, we demonstrated that the response time of the glass was less than 85 fs. The nonlinear refractive-index of Bi_2O_3–B_2O_3–SiO_2(BI) oxide glass was estimated to be about 2.93×10^{-15} cm^2/W. By measuring the dependence of the OKS signals on the pump power and on the polarization angle between pump and probe beams, we found that the OKS signals arose mainly from the photoinduced birefringence effect, and the polarization dependence of the signals was dependent on the intensity ratio of the pump beam to the probe beam.**

***Index Terms*—Bismuth compounds, nonlinear optics, optical Kerr effect, ultrafast optics.**

I. Introduction

FOR THE last few decades, the research of ultrafast all-optical switches has been stimulated by its high-bit-rate handling of optical signals for the need of future integrated optical system [1], [2]. Using the optical Kerr shutter (OKS) technique and a femtosecond laser, much effort has been invested in investigating the optical switching properties of many materials, e.g., inorganic glasses [3], [4], organic or polymeric materials [5], [6], semiconductors [7], and glasses doped with nanocrystallites or metals [8], [9], etc. Among all these various promising materials, nonresonant-type glasses have stimulated great interest for their advantages of good thermal stability and processability, high damage threshold, and low optical loss over large spectral ranges. Several studies of glass composition have yielded large third-order susceptibility $\chi^{(3)}$ and fast nonlinear optical (NLO) response time [3], [4].

When a laser beam with an ultrashort laser pulse such as a femtosecond laser is focused, the light intensity I might cause a refractive index change of the sample. When the probe pulse overlaps with the pump beam temporally, a phase shift occurs between the probe field components polarized parallel and perpendicular to the polarization of the pump pulse. Such photoinduced birefringence effect will cause the depolarization of the probe beam and the transmission through the crossed analyzer behind the sample. 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 formation of laser induced transient grating (LITG). In the previous reports, researchers have confirmed that the OKS signal induced by ultrafast laser pulse might originate from the photoinduced birefringence effect in some inorganic or organic glasses [4], [5]. However, when femtosecond pulses are used in OKS experiments for some organic–inorganic hybrid materials or nanoparticle systems, the OKS signals mainly resulted from the self-diffraction effect caused by LITG [6], [8], [9].

In this letter, we investigated the third-order NLO properties of Bi_2O_3–B_2O_3–SiO_2(BI) oxide glass using an OKS setup with a femtosecond laser. The nonlinear refractive-index of BI glass was estimated to be about 2.93×10^{-15} cm^2/W and the response time of the ultrafast nonlinear process was measured to be less than 85 fs. To confirm the origin of the third-order NLO process, we investigated the dependence of OKS signal on the polarization angle between the pump and probe beams, revealing that photoinduced birefringence is mainly responsible to the ultrafast nonlinear response. In addition, we found that the polarization dependence of the OKS signals could be controlled by adjusting the intensity ratio of the pump beam to the probe beam.

II. 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. %). CeO_2 was used to suppress the precipitation of Bi metal during melting. 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 the wavelength of 450 nm [10]. In the experiments, a femtosecond OKS arrangement was employed. The output of a Ti : sapphire laser, which emitted 30-fs and 800 nm laser pulses at a repetition rate of 1 kHz, was split into two beams. The stronger beam was used for the pump beam, and the weaker for the probe beam. The sample was placed between a polarizer and an analyzer in a cross Nicole polarizer configuration in the optical path of probe beam. The two beams were focused into the sample at an angle of 10° and the spots of the focused beams were overlapped carefully. To control the time delay and the polarization angle between the pump and probe beams, a delay-line controlled by a stepping motor and a $\lambda/2$ wave plate were introduced into the optical path of pump beam.

III. Results and Discussion

Fig. 1 shows the time-resolved OKS signals of the reference sample CS_2 and the BI glass using an OKS setup with the pump and probe beams of equal wavelength. The dotted line and the

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The authors are with the Key Laboratory for Physical Electronics and Devices of the Ministry of Education and Shaanxi Key Laboratory of Information Photonic Technique, School of Electronics and information Engineering, Xi'an Jiaotong University, Xi'an 710049, China (e-mail: jinhaisi@mail.xjtu.edu.cn).

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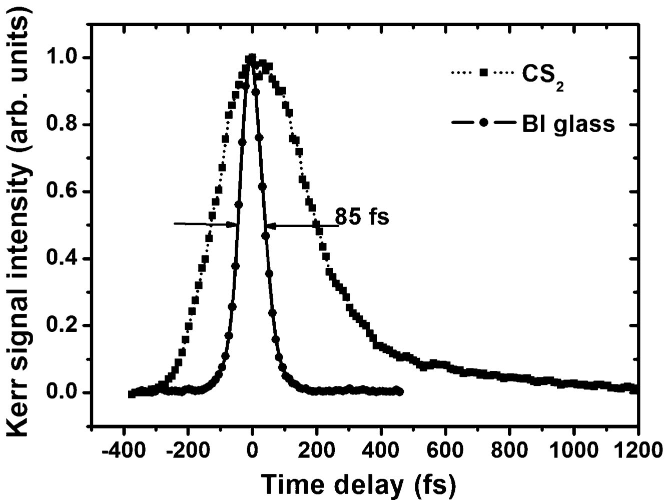

Fig. 1. Time-resolved measurements of OKS signals for BI glass and CS_2.

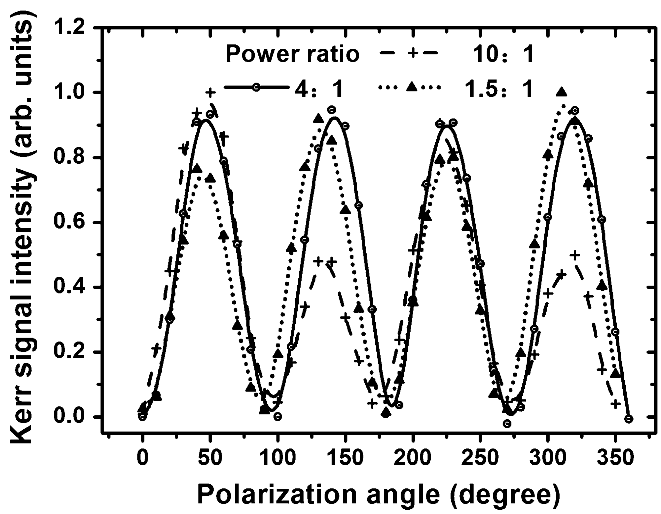

Fig. 2. Polarization dependence of the OKS signals for BI glass on the polarization angle between the two incident beams. The power ratio of the pump beam to probe beam was adjusted to 10 : 1, 4 : 1, and 1.5 : 1, respectively.

solid one refer to CS_2 and BI glass, respectively. As shown in the figure, the decay process of OKS signal of CS_2 had a slow component continuing about 1.2 ps, implying that the ultrafast response of CS_2 should be attributed to molecular orientation relaxation [11]. The full-width at half-maximum of time-resolved optical Kerr signal for the BI glass is estimated to be 85 fs, which has no slow component in the decay process. The symmetric correlation signal width has been estimated to be of 85 fs which was much faster than that of CS_2, and there was no slow component. So the response curve of BI glass was determined by the pulsewidth and the recovery time was faster than 85 fs. This indicates that the origin of the nonlinear response of the BI glass could be attributed to electronic process, the characteristic time of which was much faster than 85 fs.

To identify the origin of the OKS signals, we measured the dependence of signal intensities on the polarization angle between the pump and probe beams of equal wavelength. As shown in Fig. 2, the dashed curve depicts the polarization dependence of the OKS signal for BI glass. The pump–probe delay was set at

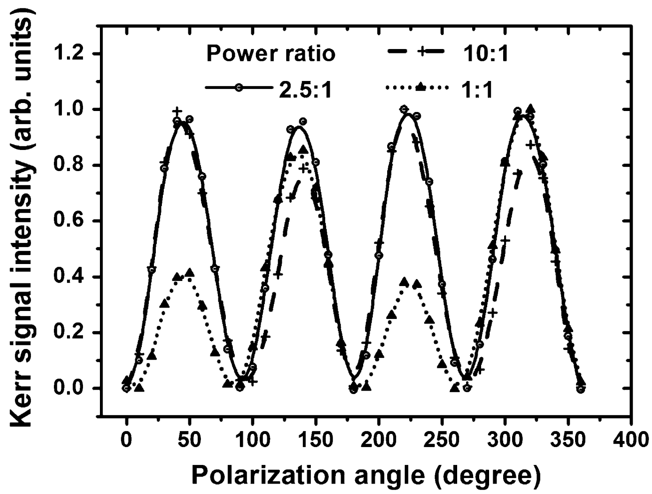

Fig. 3. Polarization dependence of the OKS signals for CS_2 on the polarization angle between the two incident beams. The solid curve shows the dependence for the power ratio of the pump beam to the probe beam fixed at 2.5 : 1, and the dashed and dotted lines refer to that of 10 : 1 and 1 : 1, respectively.

0 fs and the power ratio of the pump beam to probe beam was kept at 10 : 1. From the figure we can see that, the maximum and the minimum of the fitting line occur at $\pi/4 + n\pi/2\,(n = 0, 1, 2, \ldots)$ and $n\pi/2\,(n = 0, 1, 2, \ldots)$, respectively. However, unexpectedly, the peak values occurring at $\pi/4 + n\pi\,(n = 0, 1, 2, \ldots)$ and $3\pi/4 + n\pi\,(n = 0, 1, 2, \ldots)$ are unequal. We measured the polarization dependence of the OKS signals, when the power ratio of the pump and probe beams was adjusted to be about 4 : 1. The results are shown by the solid line in Fig. 2, in which the adjacent peak values were equal to each other. When the power ratio of the pump beam to the probe beam was further decreased to 1.5 : 1, we could obtain the dependence shown by the dotted line in Fig. 2, in which the peak values occurring at $\pi/4 + n\pi\,(n = 0, 1, 2, \ldots)$ were even smaller than the peak values occurring at $3\pi/4 + n\pi\,(n = 0, 1, 2, \ldots)$.

In the photoinduced birefringence effect, the intensity of the probe beam that passed through the analyzer is given as [12]

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

Here, θ is the polarization angle between the probe and pump beams. $\Delta\phi$ is the phase shift, which holds constant when the pump power is invariable. However, as the intensity of LITG effect was related to the coherence of the incident beams, the period of the polarization dependence of OKS signals resulted from LITG should be π, with the minimum values accruing at $n\pi\,(n = 0, 1, 2, \ldots)$ and the symmetry axis of one period of the curve lying at $n\pi + \pi/2\,(n = 0, 1, 2, \ldots)$. From the experimental results, we can see that the polarization dependence for BI glass agreed well with (1), especially for the results when the power ratio of pump beam to probe beam was fixed at 4 : 1. Therefore, the OKS signals for BI glass were attributed to the photoinduced birefringence effect.

In addition, we investigated the polarization dependence of the OKS signals of CS_2, and the results are shown in Fig. 3. From the figure, we can see that the OKS signals of CS_2 show the similar polarization dependence as the BI glass, which are dependent on the intensity ratio of pump beam to probe beam.

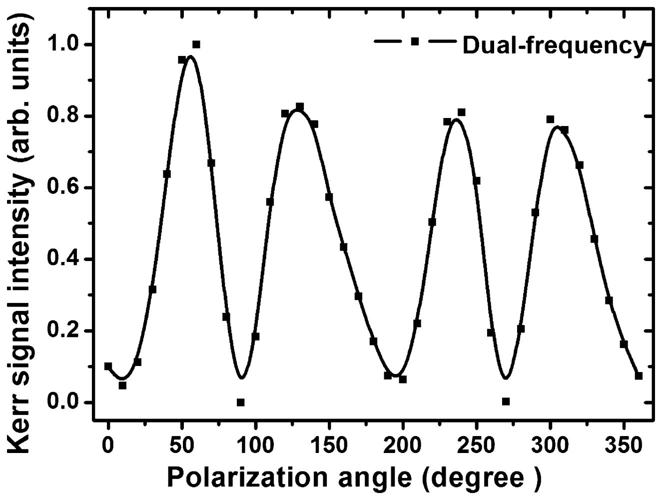

Fig. 4. Polarization dependence of the dual-frequency OKS signals for CS_2.

However, to date, we have not yet understood the mechanism for the intensity-ratio dependence of the OKS signals described above. For the further investigation of the ultrafast process, we conducted the dual-frequency optical Kerr effect, in which an 800-nm pump beam and a 400-nm probe beam were used. Because wavelength of the pump beam was different from that of the probe beam, LITG effect due to the interference between the two incident beams was avoided in the process. As for BI glass, there was a great linear absorption at 400 nm, so it was not able to perform the dual-frequency optical Kerr measurements in the sample. As shown in Fig. 4, the solid line refers to the polarization dependence of the OKS signals of 400 nm in CS_2. The polarization dependence of the OKS signals also indicates that the photoinduced birefringence effect was mainly responsible for the optical Kerr effect.

The following equation was employed to calculate the third-order NLO susceptibilities $\chi^{(3)}$ of the samples [12]:

$$\chi_S^{(3)} = \chi_R^{(3)} (I_S/I_R)^{1/2} (n_S/n_R)^2 \tag{2}$$

where the subscripts of the S and R indicate for the sample and the reference sample of quartz, respectively. I is intensity of the OKS signal when the polarization angle between the pump and probe beams was fixed at $\pi/4$ and the delay time between the pump and probe pulses was fixed at 0. n is the linear refractive-index. By comparison with that of fused quartz of 2.7×10^{-16} cm^2/W [13], the nonlinear refractive-index n_2 of the BI glass was estimated to be 2.93×10^{-15} cm^2/W, which was smaller than that of other BI glass reported in [4]. The decrease of the nonlinear refractive-index is probably due to the reduction of the concentration of Bi_2O_3 in the glass [10].

IV. Conclusion

In summary, we have investigated the ultrafast NLO response of the BI glass using a femtosecond OKS setup. The response time was measured to be less than 85 fs and the nonlinear refractive-index was calculated to be about 2.93×10^{-15} cm^2/W. The dependence of OKS signals on the polarization angle indicated that photoinduced birefringence effect was mainly responsible to the nonlinear response for the BI glass. By adjusting the intensity ratio of pump beam to probe beam, we were able to control the polarization dependence of the OKS signals.

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