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Ultrafast nonlinear optical properties of Bi_2O_3–B_2O_3–SiO_2 oxide glass

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Abstract

The ultrafast nonlinear optical properties of Bi_2O_3–B_2O_3–SiO_2 oxide glass were investigated using a femtosecond optical Kerr shutter (OKS) at wavelength of 800 nm. The nonlinear response time of this Bi_2O_3-doped glass was measured to be <90 fs. The nonlinear refractive-index n_2 was estimated to be 1.6×10^{-14} cm^2/W. Measurements for the dependence of Kerr signals on the polarization angle between the pump and probe beams showed that the Kerr signals induced by 30-fs pulse laser arose mainly from the photoinduced birefringence effect.

1. Introduction

High nonlinear (NL) optical materials with fast response time and large NL susceptibilities have attracted much attention for applications in the future high-bit-rate handling of optical signals and integrated optical systems [1–3]. Generally, photonic materials for such applications have to present large nonlinearity, low optical loss, high transparency over large spectral ranges, high damage threshold to light irradiation, good thermal stability and processability. More specifically, the third-order optical nonlinearity of such NL materials is the most significant property for realization of all-optical switching operation. Certainly, the large third-order nonlinearity can be observed near resonant wavelength in organic polymers or metals-doped glasses, but unfortunately these resonant-type materials have a large optical absorption and a slower response time in the limited wavelength range where large third-order nonlinearity is obtained. Thus, much effort has invested to the development of nonresonant-type materials that combine ultrafast response times and high optical nonlinearities for such particular applications [4–6]. Among these available NL optical materials, inorganic homogeneous glasses seem to be preferable candidates for constructing these optical systems, due to their large nonlinearities, high transparency, and fast response time [7–9]. For instance, it has been reported that chalcogenide glasses present great efficiency in optical Kerr shutter (OKS) experiment with response time less than 200 fs and third-harmonic generation susceptibility $\chi^{(3)}$ was measured to be 1.4×10^{-11} esu [6]. More recently, ultrafast nonlinear response time about 155 fs has been observed in antimony polyphosphate glasses using an optical OKS setup with 100-fs pulse laser [7].

In this letter, we investigated the nonlinear response of the nonresonant-type Bi_2O_3–B_2O_3–SiO_2 (here denoted by BI) oxide glass containing high concentrations of Bi_2O_3. The nonlinear response time was measured to be less than 90 fs using a femtosecond OKS setup, which is the fastest response time observed in nonlinear optical glasses, to our knowledge. The nonlinear refractive-index n_2 was estimated to be 1.6×10^{-14} cm^2/W, presenting large nonlinearity as large as that of chalcogenide glasses. In addition, by the aid of measurements for the dependence of Kerr signals on the polarization angle between the pump and probe

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beams, we found that for molecular systems such as the organic molecules, the Kerr signals observed when using a 30-fs pulse laser arose mainly from the photoinduced birefringence and not from a laser-induced grating as observed when using a 200-fs pulse laser. The different mechanism responsible for Kerr signals was ascribed as being due to the dependence of laser-induced transient grating and laser-induced birefringence on pulse width of femtosecond lasers.

2. Experiments

The BI glass sample was prepared by the melting method. The raw material of it contained 70%-$BiO_{1.5}$, 10%-SiO_2, 10%-B_2O_3, and 0.15%-GeO_2 (mol%). The mixture of raw material were melted in Pt crucible at high temperature in air, then the melt were poured onto a stainless-steel plate at room temperature to remove strain in the glass. There is no evident absorption above the absorption edge of 450 nm. More details on the samples preparation were given elsewhere [9]. The BI glasses with higher concentration of Bi_2O_3 showed a larger third-order nonlinearity but the same decay properties [9], thus a 0.8-mm thick bulk BI glass was selected as a sample to investigate the NL optical properties of the BI glasses.

The OKS experimental setup is shown in Fig. 1. To perform the ultrafast OKS experiments, a Ti:sapphire laser operating at 800 nm and delivering 30-fs pulses with a 1 kHz repetition rate was employed. The laser beam was split into two beams, one was used as the pump beam and the other as the probe beam. The probe beam passed through a time-delay device and a $\lambda/2$ plate to control the path length and polarization of the beam, respectively. The pump and probe beams were focused into the sample by a lens with 100-mm focal length. The sample was placed between the $\lambda/2$ plate and a polarizer in a cross-Nicol configuration. In other words, only Kerr signals generated through the third-order nonlinearity could pass through the polarizer when the pump and probe beams overlapped spatially and temporally on the sample. The polarization plane of the probe beam was rotated by $\pi/4$ against the pump beam to optimize the intensity of Kerr signal by adjusting the $\lambda/2$ plate. The Kerr signal was detected by an optoelectronic diode.

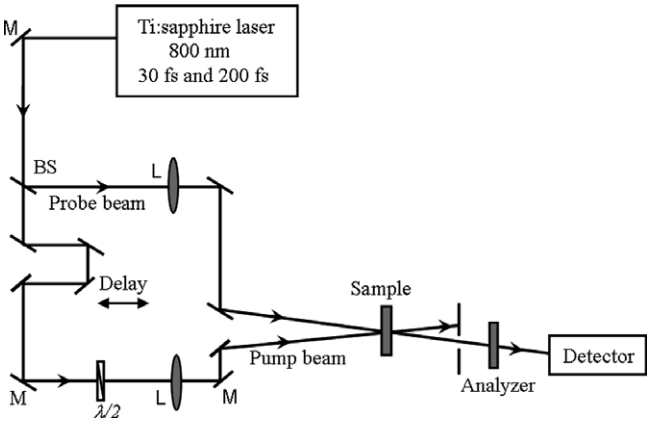

Fig. 1. Experimental setup for OKS using femtosecond laser pulses at 800 nm. BS, beam splitter; F, filter; L, lens; and M, mirror.

3. Results and discussion

Fig. 2 shows the dependence of Kerr signal intensity of the BI glass sample on delay time between the pump and probe pulses using 30-fs pulse laser. The solid curve and dotted one refer to the BI glass and reference sample CS_2, respectively. The full width at half-maximum of the pulse correlation for the Kerr signals of the BI glass was 90 fs, which was the fastest response observed in nonlinear optical glasses so far. For the assumed hyperbolic secant pulse shape, the symmetric correlation signal width of 90 fs, which showed no slow component and which was much faster than that of reference sample CS_2, implied that the response curve was determined by pulse width and that the recovery time of the BI glass is faster than that. This indicates that real excitation due to multi-photon absorption which causes free carrier absorption is negligible, and the origin of nonlinearity may be attributed to electronic processes either alone or in combination with other processes whose characteristic times are shorter than 90 fs. The nonlinear refractive-index n_2 was calculated to be 1.6×10^{-14} cm^2/W by comparison with that of fused quartz of 2.7×10^{-16} cm^2/W [10] which was used as a calibration standard. This value compares favourably with those for chalcogenide glasses.

Some studies have showed that Kerr signal was mainly related to self-diffraction generated by the ultrafast laser-induced transient grating (LITG) in OKS experiments using 200-fs laser pulses [11,12]. To discriminate the origin of optical Kerr signals of the BI glass in our experiment using 30-fs pulses, we measured the dependence of Kerr

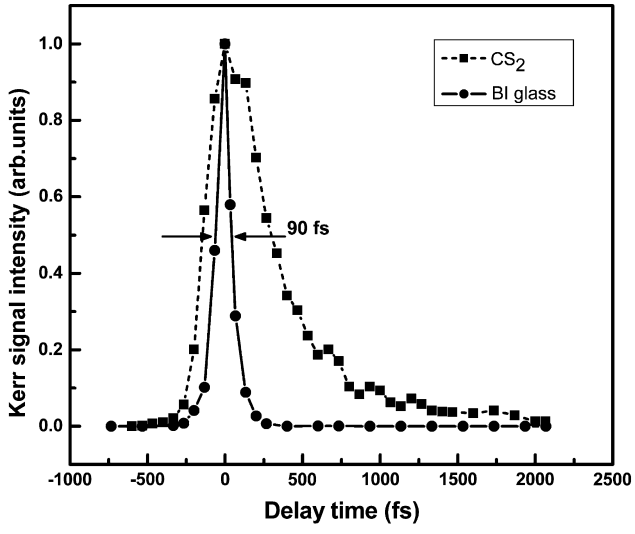

Fig. 2. Time-resolved measurements of Kerr signals for the BI glass and CS_2.

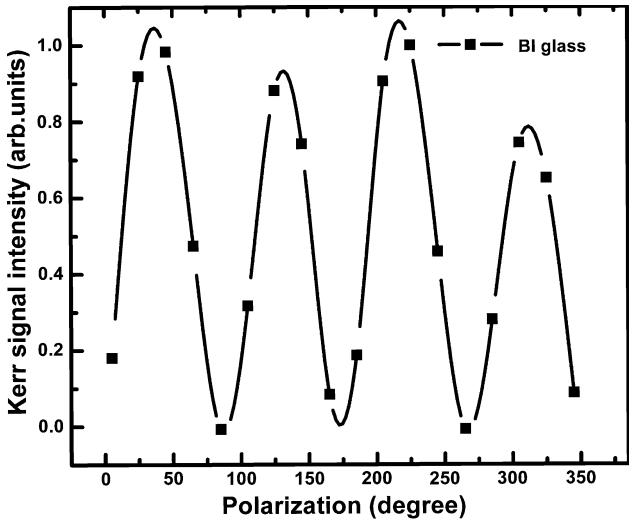

Fig. 3. Dependence of Kerr signal intensity on polarization angle between the pump beam and probe beam for the BI glass induced by 30-fs pulses.

signal intensity on the polarization angle between the probe and pump beams when the pump–probe delay time was set at 0. As shown in Fig. 3, the period of the dependence of Kerr signals on the polarization is $\pi/2$, and the maximum value of Kerr signals occurs at $\pi/4 + n\pi/2$ ($n = 0, 1, 2, \ldots$).

In OKS configuration 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 second polarizer. In that case, the Kerr signal intensity is given as:

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

where θ is the intersecting angle between polarization of the probe and pump beams, $\Delta\phi$ is the phase shift. $\Delta\phi$ holds constant, while the pump power is invariable. The Kerr signal intensity I depends on the intersecting polarization angle θ. Comparing the results shown in Fig. 3 with Eq. (1), we can conclude that the photoinduced birefringence effect was responsible for the Kerr signals of the BI glass when the 30-fs pulse laser was used.

In order to confirm the experimental results shown in Fig. 3, we used 200-fs and 30-fs pulse lasers, respectively, in optical Kerr experiments for lead phthalocyanine (PbPc)-doped gel glass with 0.6-mm thickness, and measured the dependence of Kerr signal intensity on the polarization angle between the pump and probe beams, where the delay time was set at 0 fs. The results are shown in Fig. 4. From Fig. 4a, we can see that when 200-fs pulses laser is used, the maximum probe transmittance occurs at $n\pi$ ($n = 0, 1, 2, \ldots$), and the period of the relation curve is π. While the maximum probe transmittance occurs at $\pi/4 + n\pi/2$ ($n = 0, 1, 2, \ldots$), and the period of the relation curve is $\pi/2$ for 30-fs pulse laser (Fig. 4b). These results can be explained as follows: when 200-fs pulse laser was used, the LITG was generated in PbPc-doped gel glass and diffracted the incident pulses itself to yield Kerr signals.

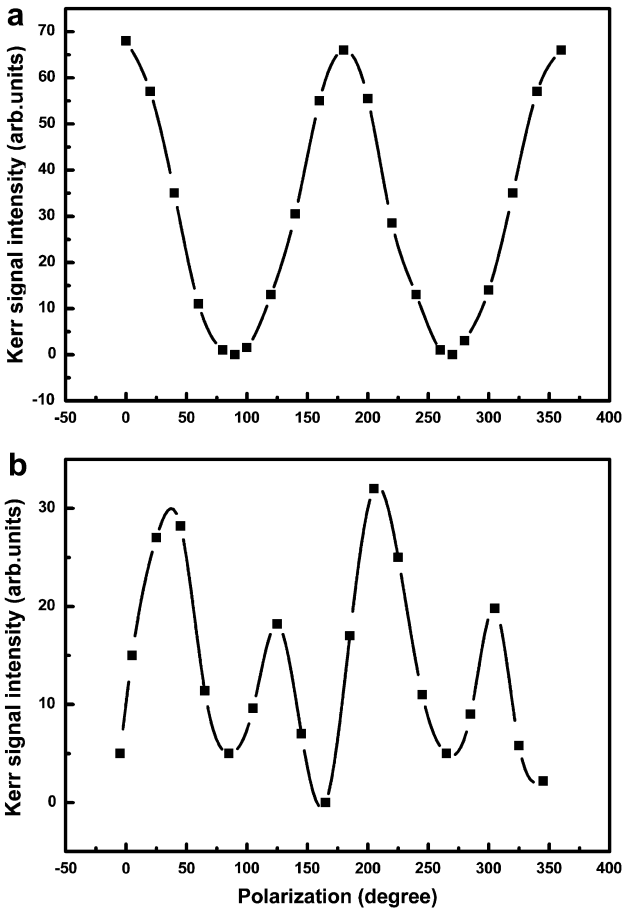

Fig. 4. Dependence of Kerr signal intensity on polarization angle between the pump beam and probe beam for PbPc samples using different femtosecond lasers: (a) for 200-fs pulse laser and (b) for 30-fs pulse laser.

The interference pattern could be translated into such corresponding LITG effect [13]. As we know, femtosecond lasers have a broad wavelength distribution. The monochromaticity of 30-fs pulses are much worse than that of 200-fs pulses, meaning that when 30-fs pulse laser was used LITG effect generated by the interference between two incident beams was sharply cut down, and contribution of the photoinduced birefringence effect to the Kerr signals became dominant. When a femtosecond laser whose pulse width is about 100 fs is used, the Kerr signals probably result from both the LITG and photoinduced birefringence.

In addition, it should be explained that LITG usually occurs in molecular systems such as the organic molecules, in which molecular population induced by the pump and probe beams leads to transient refractive-index modulation. Third-order nonlinearity of the BI glasses used in our experiments resulted mainly from the pure electronic polarization, and the molecular population could be negligible. Therefore, LITG can not be formed in the BI glasses even using the 200-fs laser, and the Kerr signals will arise from the birefringence effect related to the pure electronic polarization.

4. Conclusions

In summary, we investigated the ultrafast nonlinear optical response of the BI oxide glass using a femtosecond OKS setup. The response time was measured to be <90 fs. The nonlinear refractive-index n_2 was estimated to be 1.6×10^{-14} cm^2/W. Additionally, the experimental results for the dependence of Kerr signal intensity on the polarization angle between two incident beams showed that photoinduced birefringence effect related to electronic processes was the main contribution to Kerr signals of BI glass.

Acknowledgements

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