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Elimination of the coherent effect in the optical Kerr measurement of bismuth glass using supercontinuum

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A strong coherent effect was observed in the monochromatic optical Kerr effect (OKE) experiment of a Bi_2O_3-B_2O_3-SiO_2 oxide glass (BI glass). To eliminate the influence of the coherent effect, we proposed a simple femtosecond two-color OKE measurement method with wavelength tunability using a supercontinuum. In the two-color OKE measurement the third order nonlinear susceptibility of the BI glass was measured to be 1.64×10^{-13} electrostatic units (esu), in which the influence of the coherent effect was eliminated. The result is smaller than that obtained in a previous monochromatic OKE experiment. In addition, we discuss the discrepancies between the result obtained in this paper and those reported in other studies. © *2011 American Institute of Physics*. [doi:10.1063/1.3597787]

I. INTRODUCTION

Nonresonant-type bismuth glasses containing high concentrations of Bi_2O_3 are preferable candidates for the optical Kerr gate applications,[1–3] because of their excellent optical properties, such as large nonlinearities and ultrafast response time. The third-order nonlinear susceptibility is one of the most important parameters in characterizing the properties of bismuth glasses for these applications.[4–8] Precise experimental determination of the third-order nonlinear susceptibility of bismuth glasses is fundamentally important.

The optical Kerr effect (OKE) measurement is a common method to characterize the third-order optical nonlinearities of optical materials. The procedure for OKE measurement is more or less standardized. Results are not always consistent, however, because of the complexities of the experiment.[9,10] The third-order nonlinear susceptibility of a bismuth glass was measured using femtosecond monochromatic OKE measurement in our previous work.[11] However, a coherent effect often accompanies the OKE signal, especially using a femtosecond laser.[12] When the time interval between a pump and a probe pulse is within the field correlation time, the two pulses will interfere with each other to form a transient grating in the noncollinear monochromatic OKE experiment.[13] Energy can be scattered from one beam to another, leading to an additional signal beyond the pure OKE signal. Therefore, it is important to eliminate the influence of the unwanted coherent effect on the OKE measurements. A two-color OKE experiment can be carried out, in which a frequency-double laser is generally used.[14,15] However, some samples might absorb the frequency-double pulse strongly, for example bismuth glasses have strong absorption around 400 nm, which will limit the performance of this method. The other excellent solution is to use an optical parametric

amplifier (OPA) with wide wavelength tunability in the two-color experiment,[16] but the OPA is complex and not a common laboratory apparatus. In our previous works,[17,18] we have concentrated on the application of the Bi_2O_3-B_2O_3-SiO_2 (BI) oxide glass optical Kerr gate to control the gated spectra with narrow bandwidth from a supercontinuum (SC). As we found that the coherent effect would disturb the OKE signal, we noticed that a two-color OKE measurement method with wavelength tunability using the SC could be used to eliminate the coherent effect in the OKE measurement.

In this study, a strong coherent effect was observed in the monochromatic OKE measurement of a BI glass. To eliminate the influence of the coherent effect on the OKE measurement, we proposed a simple femtosecond two-color OKE measurement with wavelength tunability using the SC. Using the two-color OKE measurement the third-order nonlinear susceptibility of the BI glass was measured to be 1.64×10^{-13} electrostatic units (esu), smaller than what we obtained in a previous monochromatic OKE experiment.[11] In addition, we also discussed the discrepancies between the results obtained in this paper and those reported in other studies.

II. EXPERIMENTS AND MATERIAL

A BI glass sample with a composition of 70% $BiO_{1.5}$, 10% SiO_2, 10% B_2O_3, and 0.15% GeO_2 (mol. %)was prepared by the melting method. The details about the preparation of the sample are given in Ref. 19. The thickness of the glass sample was about 1.5 mm.

The two-color OKE measurement using the SC is illustrated in Fig. 1. A Ti:sapphire laser system with a repetition rate of 1 kHz, pulse duration of 30 fs, and an average power of 1 W at 800 nm was used in our experiments. The emitted fundamental laser beam was separated into two parts by means of a beam splitter. The weak beam was focused into a 3 mm sapphire plate to generate a stable SC. The SC was

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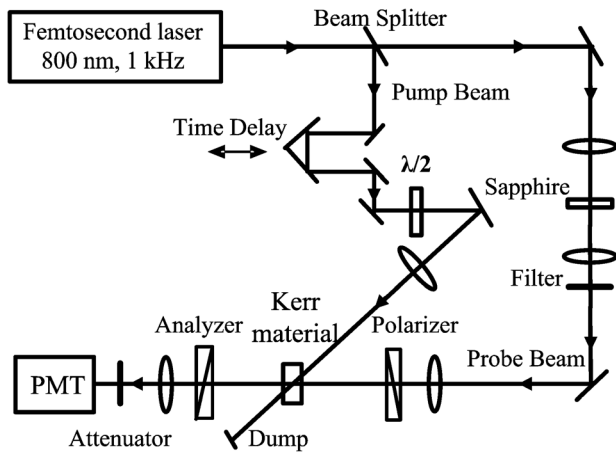jinhaisi@mail.xjtu.edu.cn.

FIG. 1. Schematic setup for the two-color optical Kerr measurements using SC.

collimated and passed through a narrow bandpass filter. The transmitted pulse, with a nearly Gaussian spectral profile, was acquired as the probe beam. The narrow bandpass filter was chosen to ensure that the transmitted beam was not absorbed by the sample and had no coherent component to the fundamental frequency beam. The sample was positioned between the polarizer and the analyzer in a cross-Nicole configuration set on the probe beam path. Passing through an optical delay-line, enabling a temporal resolution of about 5.2 fs, the intense beam was finally focused onto the Kerr material as a pump beam. The polarization of the pump beam was controlled by a half-wave plate. The OKE signals were detected by a photomultiplier tube (PMT). For the monochromatic OKE measurement the weak beam was directly focused onto the nonlinear optical material as the probe beam without passing through the sapphire plate.

III. RESULTS AND DISCUSSION

To investigate the influence of the coherent effect on the Kerr signal, we first measured the coherent effect in both of the OKE measurement configurations mentioned above, fixing the polarization angle of the pump beam relative to the probe beams at 0° by the half-wave plate. The result is shown in Fig. 2(a). From Fig. 2(a), we can see that the energy transfer between the pump beam and the probe beam was caused by the coherent effect in the monochromatic configuration (open circles), which is different from that in two-color configuration (solid circles). The monochromatic signal is a typical two-beam-couple signal for a material with a positive nonlinear index of refraction, in which light is scattered from the beam of high frequency to that of low frequency leading to an additional signal beyond the OKE signal.[20] There was no coherent effect observed in the two-color configuration, as shown in Fig. 2(a).

Then we studied the influence of the coherent effect on the OKE signals in the monochromatic configuration. The dependence of the OKE signals on pump power at the time delays of $+80$ and -80 fs, which correspond to the temporal positions of the peak and valley in Fig. 2(a), were measured as shown in Fig. 2(b). When the pump power is below 1

mW, the signal intensities at both time delays coincide well (Fig. 2(b)). When the pump pulse power is further increased, the signal intensities deviate from each other and increase more rapidly at 80 fs than that at -80 fs. The reason is that the influence of the coherent effect on the OKE signals becomes larger with increasing pump pulse power, which distorts the real OKE signal seriously.

Furthermore, we investigated the nonlinearity of the BI glass in the two-color OKE measurement configuration. The new probe beam filtered from the SC is shown in Fig. 3. From Fig. 3, we can see that the new probe beam had a center wavelength of 750 nm with a nearly Gaussian spectral profile and nearly no coherent component to the fundamental frequency beam.

Under the two-color OKE measurement configuration, we measured the time-resolved OKE signal of the BI glass. The results are shown in Fig. 4, from which we can see that the full-width at half maximum of the time-resolved OKE signal of the BI glass is estimated to be below 247 fs and no slow decay is observed. In the two-color OKE experiment, the third-order optical nonlinearities usually result from the nonresonant electronic process and resonant processes, such
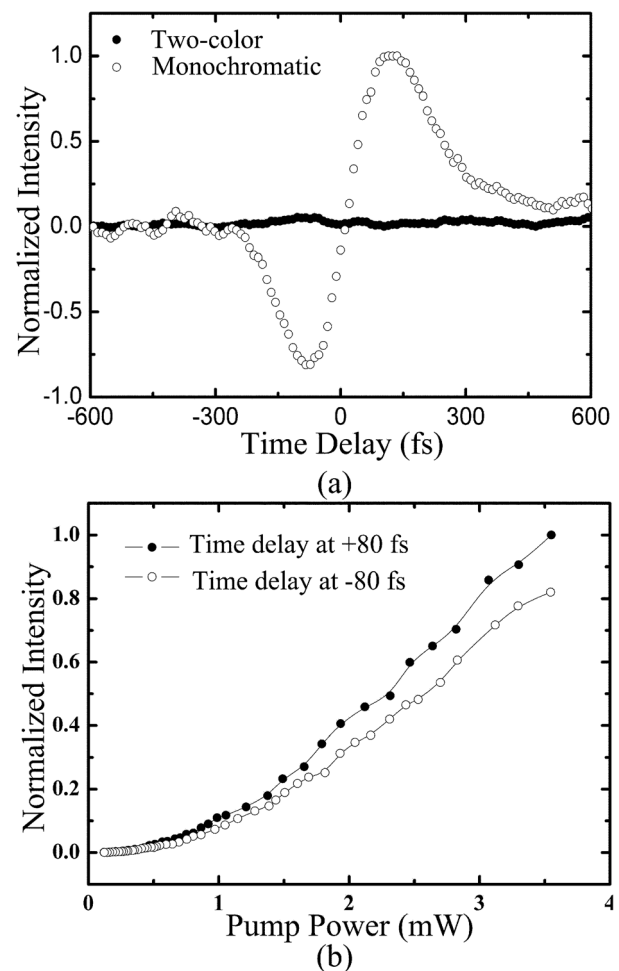

FIG. 2. (a). Coherent signals of the OKE experiments: the open circles plot the monochromatic configuration and the solid circles plot the two-color configuration; (b). Dependence of the OKE signals on pump pulse power at the time delays of $+80$ fs (solid) and -80 fs (hollow) in the monochromatic configuration.
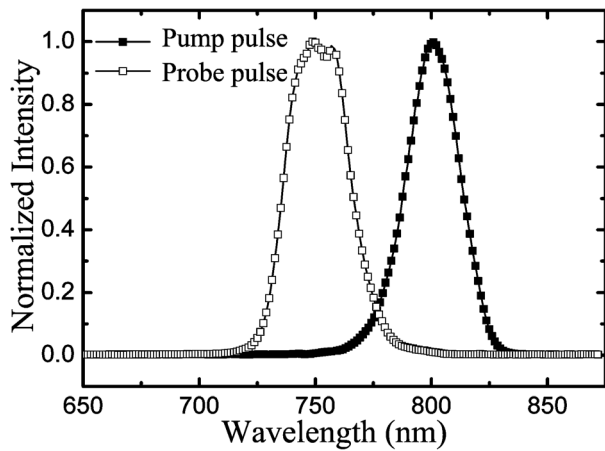
FIG. 3. Spectra of the pump and probe beams in the two-color OKE experiments using SC.

as resonant Raman and two-photon processes.[21] For the inorganic solid media, however, the resonant Raman and two-photon processes generally lead to slower decay because both the excited electric state relaxation and the Raman vibrational relaxation are on the picosecond time scale or longer.[22,23] Therefore, we conclude that the mechanism of the third-order optical nonlinearity in our experiment can be mainly attributed to the nonresonant electron polarization. That is probably because the difference between the pump and probe frequencies in our experiment is 833 cm^{-1} and far from the nearest Raman spectral peak (890 cm^{-1}) of the BI glasses.[24] Under the same conditions, the time-resolved OKE signals of the reference samples quartz and CS$_2$ were also measured. The quartz plate had the same optical length as the BI glass. The CS$_2$ solution filled a glass cuvette with a path length of 1 mm. From Fig. 4, we can see that the curve of the quartz was almost the same as that of the BI glass because the dominant mechanism of its OKE is also electron polarization. However, the curve of the CS$_2$ has an asymmetrical decay tail with a decay time of 1.6 ps which is attributed to the molecular orientation relaxation. These results were in accord with previous reports and indicated the reliability of our experiment.[25]
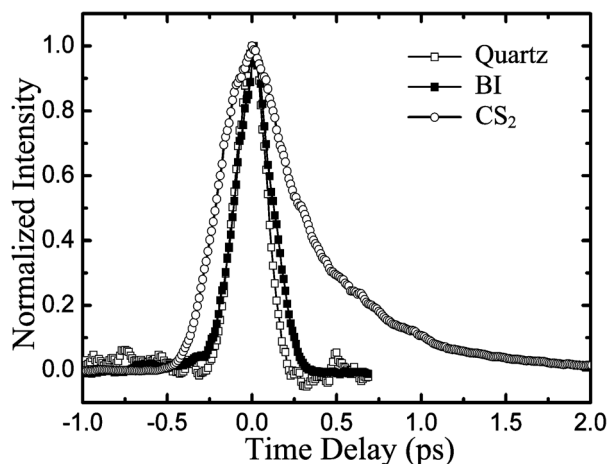
Finally, we investigated the dependence of the OKE signals on the pump beam power. The signal intensity depends on the induced phase shift $\Delta\varphi$ and the angle θ between the polarization planes of the pump and probe beams according to Ref. [22]:

$$I = I_0 \sin^2(2\theta) \sin^2\left(\frac{\Delta\varphi}{2}\right) \qquad (1)$$

When the polarization angle θ between the pump and probe beams is fixed at 45° and the induced phase shift $\Delta\varphi$ is small enough, the optical Kerr effect signal is proportional to the square of P_p as follows:[26]

$$I/I_0 = \left(\frac{3 L_{\text{eff}} \chi^{(3)}}{\lambda_p c\varepsilon_0 r^2 n_0^2}\right)^2 P_p^2 \qquad (2)$$

where L_{eff} is the effective length of the sample, $\chi^{(3)}$(SI system of units) is the third-order susceptibility, λ_p is the wavelength of the probe beam, and P_p and r are the pump power and the beam radius at the focal point, respectively. The experimental and theoretical results based on Eq. (2), which agreed well with each other, are shown in Fig. 5.

Following the standard reference measurement,[27] we employ the following equation to calculate the third-order nonlinear optical susceptibilities of the glass sample:

$$\chi_S = \chi_R \left(\frac{(I/I_0)_S}{(I/I_0)_R}\right)^{\frac{1}{2}} \left(\frac{n_{0S}}{n_{0R}}\right)^2, \qquad (3)$$

where the subscripts S and R indicate the sample and the reference sample, respectively. Using the third-order susceptibility $\chi_R = 6.58 \times 10^{-15}$ esu (~2.48×10^{-16} cm^2/W) and linear refractive index $n_{0R} = 1.45$ of the fused quartz,[28] the third-order susceptibility of the BI glass sample was estimated to be 1.64×10^{-13} esu for $n_{0S} = 2.13$. It has the same order as the theoretical value of 6.29×10^{-13} esu according to Miller's Rule.[29]

It should be indicated that there were some different results reported in previous studies on the third-order

FIG. 4. Time-resolved OKE signals of the BI glass, the CS$_2$, and the quartz. Solid squares: BI glass; Circles: CS$_2$; Open squares: quartz.
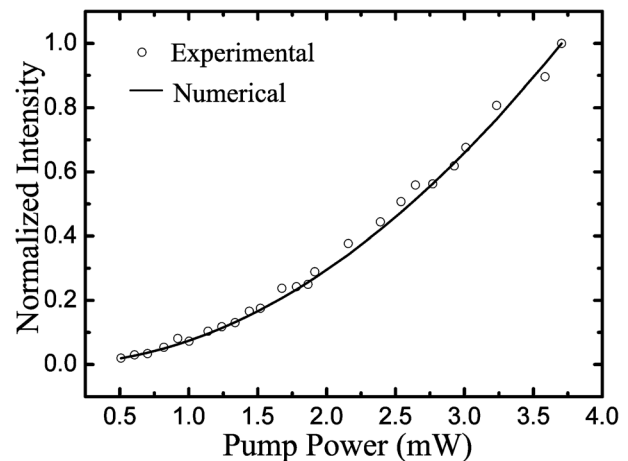

FIG. 5. Dependence of the OKE signals on pump pulse power at the time delay of 0 fs in the two-color OKE experiment. Circles: experiment results; Solid line: calculated curve from Eq. (3).

susceptibility of bismuth glasses. For example, the third-order susceptibilities of series of bismuth glasses were measured to be $\sim 10^{-12}$ esu and 9.2×10^{-13} esu ($\sim 1.6 \times 10^{-14}$ cm^2/W), respectively, using a Z-scan method and a monochromatic OKE measurement.[11,30] The discrepancies should be attributed to different moduli of the third-order susceptibility $\chi_{ijkl}^{(3)}$ for different approaches, i.e., $\left|\chi_{xxxx}^{(3)}\right|$ for the Z-scan measurement and $\left|\chi_{xxxx}^{(3)} - \chi_{xxyy}^{(3)}\right|$ for the OKE measurement. On the other hand, a possible coherent effect in the monochromatic OKE measurement could lead to an additional signal beyond the pure OKE signal, which caused the measured third-order susceptibility to be bigger than that obtained in this paper.

IV. CONCLUSIONS

In conclusion, we have investigated the influence of the coherent effect on the OKE signals for BI glass in a monochromatic femtosecond OKE experiment. To eliminate the coherent effect, we proposed a simple two-color OKE experiment using SC. The third-order nonlinear susceptibility of BI glass sample was estimated to be 1.64×10^{-13} esu, in which the influence of the coherent effect was eliminated. The result is smaller than that obtained in the monochromatic OKE experiment previously. In addition, we also discussed the discrepancies between the results obtained in this paper and those reported in other studies.

ACKNOWLEDGMENTS

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