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The Influence of Coherent Transient Energy Transfer on Femtosecond Time-Resolved Z-Scan Measurements *

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We investigate the influence of the coherent effect on femtosecond time-resolved Z-scan measurements using a degenerate pump-probe Z-scan technique. The time response of the light-induced transient grating (LITG) of Bi_2O_3-B_2O_3-SiO_2 (BI) glass shows a valley-peak variation, which has an obvious influence on the time-resolved Z-scan measurements. The valley-peak variation of the LITG signals with the delay time in BI glass is due to coherent transient energy transfer between positively chirped pump and probe pulses. The influence of the LITG effect on the closed-aperture time-resolved Z-scan measurements could be reduced by adjusting the position of the aperture transversely.

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The Z-scan technique is an accurate and sensitive tool for determining nonlinear refraction and absorption for a wide variety of materials.[1−3] This technique enables us to measure both the sign and magnitude of third-order nonlinear susceptibility χ^3. By use of a pump and delayed-probe configuration, this method can be used to study the relaxation time of optically induced nonlinearities and distinguish the nonlinearities having different temporal responses, which is called the time-resolved Z-scan.[4−6] However, in the degenerate pump-probe configuration, coherent effects such as the light-induced transient grating (LITG) effect occur when the two beams overlap in time and space within the sample.[7−9] Coherent effects make it hard to extract information about the response and nonlinear refractive index of the material, especially in materials that have an ultrafast response.[10−12] There exists extensive literature concerning coherent effects, especially the LITG.[7,8,13,14] There are, however, arguments on the coherent effects left in time-resolved measurements using degenerate pump-probe configurations.[15,16] In our previous work, the influence of the LITG effect on femtosecond pump-probe optical Kerr measurements has been clarified in Bi_2O_3-B_2O_3-SiO_2 (BI) glass, which shows large optical nonlinearity[9,17,18] and an ultrafast electronic response of less than 90 fs.[10]

In this Letter, we study the influence of the LITG effect on the non-collinear femtosecond time-resolved Z-scan measurements using an 800 nm femtosecond laser. BI glass is used as the nonlinear medium in our experiments. We find that the time response of signals resulting from the LITG effect shows a valley-peak variation, which has an obvious influence on the time-resolved Z-scan measurements. The valley-peak variation of the LITG signals with the delay time in BI glass is due to the coherent transient energy transfer between positively chirped pump and probe pulses. The influence of the LITG effect on the closed-aperture time-resolved Z-scan measurement could be reduced by adjusting the position of the aperture. After reducing the influence of the LITG, the nonlinear response time of the BI glass is estimated to be less than 175 fs.

In our experiments, the experimental setup of the non-collinear femtosecond time-resolved Z-scan was similar with the optical Kerr shutter (OKS) setup used in Ref. [10], except that the analyzer was replaced with an aperture. The focusing and detecting parts of the experimental setup are illustrated schematically in Fig. 1. A multi-pass Ti:sapphire amplifier laser system was employed in experiments, which delivered a train of 800 nm, 30 fs laser pulses at a repetition rate of 1 kHz. Using such a low repetition rate laser, the refractive index change induced by the thermal effect can be neglected.[5] The laser beam was split into two beams to provide the pump and probe beams. The pump and probe powers were set at 1 mW and 0.1 mW, respectively. The two beams were focused into the sample noncollinearly by two lenses with a focal length of 15 cm, respectively. The spots of the focused beams were adjusted carefully to overlap well in the sample. An optical delay line was introduced into the path of the pump beam to control the time delay between the two beams. The polarization of the pump beam was controlled by a half-wave plate. In the time-resolved

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Z-scan experiments, the dependence of signals on the polarizations of the pump and probe beams was measured using the parallel and orthogonal configurations, in which the polarizations of the two beams were set to be parallel and perpendicular, respectively. In addition, the time-resolved Z-scan was performed using a closed aperture and an open aperture in order to distinguish the nonlinear refractive effect and the LITG effect. In the closed-aperture case, the aperture linear transmittance was 0.4. The measured signals were characterized with the change of the normalized transmittance of the probe beam ΔT. Z denotes the distance between the focal plane of the probe beam and the sample. The positive and negative Z directions are defined as the probe beam focus in front of the sample and behind the sample, respectively. At a fixed time delay, if neglecting the LITG effect, the closed aperture signal ΔT as a function of the sample position Z is shown in Fig. 1. When the medium with the positive (negative) nonlinear refractive index is used, ΔT is positive (negative) for the positive Z while negative (positive) for the negative Z. By shifting the lens for the probe beam longitudinally, the probe beam focus was set in front of the sample or behind the sample to measure the time-resolved signals.

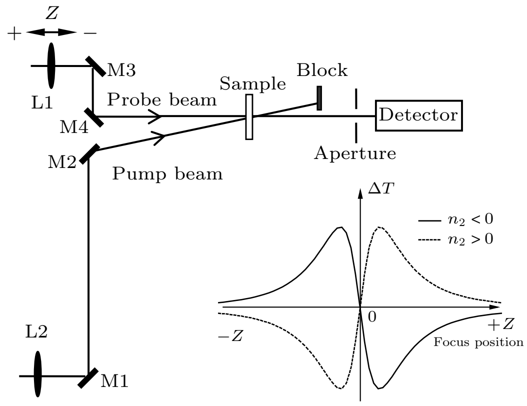

Fig. 1. Schematic setup of the non-collinear time-resolved Z-scan experiments. Z refers to the distance of the probe beam focus from the sample. Inset: the Z-scan pattern for the material with a different sign of the nonlinear refractive index.

First, the transmittance of the probe beam was measured as a function of the time delay between the pump and probe pulses. The polarizations of pump and probe beams were set to be parallel. The probe beam focus was set in front of the sample and adjusted carefully to obtain signals with the largest magnitude. As shown in Fig. 2, the time response curves for both closed aperture and open aperture show no slow component. Because the BI glass has a positive nonlinear refractive index,[17,18] the transmittance change of the probe beam ΔT due to self-focusing should be positive in the closed-aperture time-resolved Z-scan. In Fig. 2, however, with the increase in the time delay, the closed-aperture signals firstly decrease to −0.15, then increase to 0.31, and finally decrease to zero. The open-aperture signals exhibit a similar trend but smaller magnitude. According to the open-aperture single beam Z-scan measurement, no obvious nonlinear absorption was observed in the BI glass. Hence, such a valley-peak variation was probably attributed to the LITG effect rather than nonlinear absorption, which had large influence on the time-resolved Z-scan measurement.

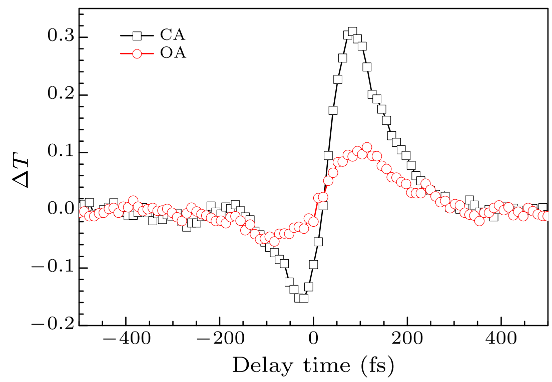

Fig. 2. Time-resolved Z-scan of the BI glass obtained with parallel polarizations of the pump and probe beams. CA, closed aperture; OA, open aperture. The probe beam focus was set in front of the sample.

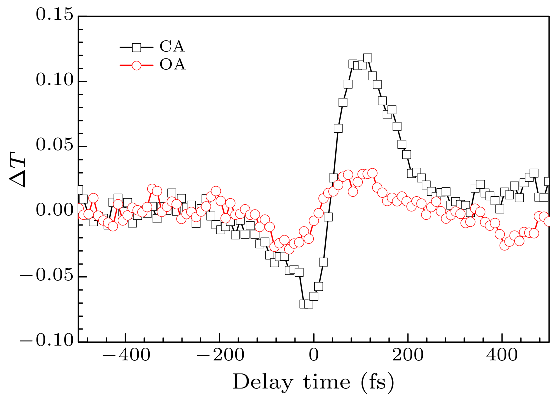

Fig. 3. Time-resolved Z-scan of the BI glass obtained with orthogonal polarizations of the pump and probe beams. CA, closed aperture; OA, open aperture. The probe beam focus was set in front of the sample.

To decrease the influence of the LITG, the polarizations of pump and probe beams were set to be orthogonal to measure the time-resolved Z-scan signals. However, as shown in Fig. 3, the influence of the LITG still existed. Compared with that in Fig. 2, the time responses of the BI glass in orthogonal polarization configuration show similar valley-peak variations, except that the magnitudes of the probe transmittance change decrease. When the probe beam focus was set behind the sample, the time response of the closed-aperture signals became a valley (refer Fig. 4) while that of the open-aperture signals still retained a similar valley-peak variation.

According to the results described above, the closed-aperture time-resolved Z-scan was mainly due to the self-focusing effect. The open-aperture time-resolved Z-scan signals should be mainly attributed to the LITG effect. As is well-known, pump and probe pulses can induce the third-order nonlinear polarization associated with both electric fields and χ^3. The third order nonlinear polarization was spatially modulated due to the interaction between pump and probe pulses and an index grating could be induced in Kerr media, even for the perpendicularly polarized pump and probe pulses.[19−21] Therefore, the signal arising from the LITG effect was not eliminated by setting perpendicular polarizations of pump and probe pulses. The closed-aperture time-resolved Z-scan measurements modulated by the LITG effect showed a similar valley-peak variation, which was much easier to observe for the positive transmittance change of the probe beam ΔT due to self-focusing effect than the negative one.

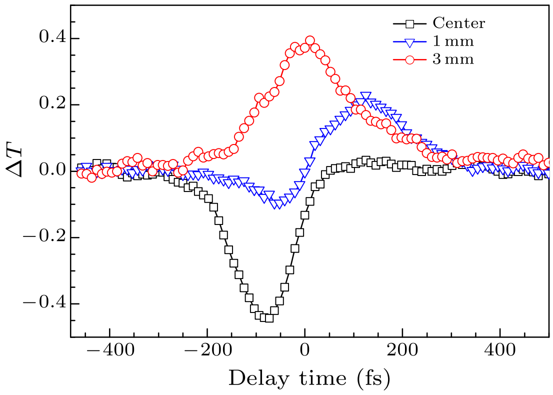

Fig. 4. Time-resolved closed-aperture Z-scan of the BI glass for the different aperture radial positions. The polarizations of the pump and probe beams are set to be parallel. The probe beam focus is set behind the sample.

The valley-peak time response variation can be attributed to the transient energy transfer resulting from the refractive LITG induced by chirped pulses in Kerr media.[19] The energy transfer between the pump and probe beams depends on their frequencies in the temporal-nonlocal response Kerr media (TNRKM).[22] In the TNRKM with a positive nonlinear refractive index, the energy is transferred from the high-frequency component to the low-frequency component. The energy transfer efficiency depends on the frequency detuning of both beams. The time response of the valley-peak variation was because of the variation of frequency detuning in the overlapping region of the pump and probe beams with respect to their delay time. Pump and probe pulses reaching the sample had positive chirp introduced by the optical system. When the probe beam temporally overlapped with the pump beam in the minus time delay, the frequency of the probe beam was higher than that of the pump beam, and therefore the energy was transferred from the probe beam to the pump beam, and the intensity of the probe beam decreased. Conversely, the energy was transferred from the pump beam to the probe beam, and the intensity of the probe beam increased. The control of energy transfer direction has been realized experimentally by using chirp-controlled pulses.[23] However, the transient energy transfer should be restrained to extract the dynamics of other nonlinearities, such as the self-focusing effect and the optical Kerr effect. A two-color optical Kerr measurement method has been proposed to eliminate the influence of the energy transfer on the time-resolved optical Kerr measurement.[24]

To reduce the influence of the LITG effect on the degenerate time-resolved Z-scan measurements, we studied the dependence of the time response of the closed-aperture signals on the position of the aperture by transversely adjusting the position of the aperture for the center of the probe beam, while keeping the aperture size constant. The probe beam focus was set behind the sample. For the self-focusing effect, the time response of the closed-aperture signals should be a valley and a peak, respectively, when the aperture was in the center of the probe beam and at the edge.[25,26] Figure 4 shows the results for different radial positions of the aperture in the parallel polarization configurations. When the aperture was in the center of the probe beam, the closed-aperture signals showed a time response with a valley. When the aperture was 1 mm away from the center of the probe beam, the signals showed a time response with valley-peak variation. For the position of 3 mm, the time response became only a peak, similar to that of the self-focusing component. The full-widths at half-maximum (FWHM) of the time response were estimated to be 175 fs and 135 fs for the aperture at 3 mm and center, respectively. The time-resolved measurements in orthogonal configuration were also performed, and the similar position dependence of the time response was observed. According to the above results, the time response for the position of 3 mm, which showed only a peak, indicated that the influence of the LITG effect can be neglected. However, the asymmetry of the time response for the center position indicated that the contribution of LITG to the time-resolved Z-scan signals can not be neglected, which made the measured value of the response time of the BI glass become shorter than the real value. The aperture position dependence indicates that the contribution of the LITG effect to the closed-aperture signals decreased when the aperture was away from the center of the probe beam. This is probably because the edge light of the probe beam is more sensitive to the self-focusing than at the center.[24,25] Therefore, we conclude that the influence of the energy transfer resulting from the LITG effect on the closed-aperture time-resolved Z-scan measurements could be reduced by setting the aperture at the edge of the probe beam. After reducing the influence of the LITG effect, the nonlinear response time of the BI glass was estimated to be less than 175 fs, which was attributed mainly to the electronic polarization process. This value is larger than that in Ref. [10], because the duration of pulses arriving at the sample was longer in this experiment, which was broadened to be about 125 fs due to the group velocity dispersion in the optical path.

In summary, we have investigated the influence of the LITG effect on the non-collinear femtosecond time-resolved Z-scan measurements. The transient energy transfer occurs due to the LITG induced by positively chirped pulses in the BI glass. The time response of the transient energy transfer resulting from the LITG effect shows a valley-peak variation, which has an obvious influence on the time-resolved Z-scan measurements. The influence of the LITG effect on the closed-aperture time-resolved Z-scan measurements could be reduced by setting the aperture at the edge of the probe beam.

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