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Influence of Self-Diffraction Effect on Femtosecond Time-Resolved Single-Shot Optical Kerr Measurements *

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Femtosecond time-resolved single-shot optical Kerr gating (OKG) measurements are performed by focusing the probe pulse and using a cylindrical lens to introduce a spatially encoded time delay with respect to the pump pulse. By measuring the pump power and polarization dependence of the OKG signals in CS_2*, the contribution of self-diffraction effect which is independent of the nonlinear response time of the material is directly observed on the rising edge of the time-resolved OKG signals. The influence of the self-diffraction effect on the optical Kerr signal could be controlled effectively by varying the polarization angle between pump and probe pulses.*

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Various kinds of ultrafast transient phenomena of materials have been extensively studied after the invention of ultrashort pulsed lasers.[1−8] Femtosecond time-resolved pump-probe technique based on optical Kerr gating (OKG) measurements has been proven to be one of the most convenient nonlinear optical measurements due to its high precision, ultrafast switching time and broad wavelength range.[9−13] However, the conventional pump-probe OKG technique is limited to materials without reversible photoreaction due to the fact that the measurement requires many repetitions of pump-probe pulse sequences with different delay times to record the temporal signals in a region of interest. To measure the OKG signals in materials with some photoinduced irreversible reactions, many techniques based on single-shot detection have been proposed to date.[14−20] In these techniques, the probe pulses are manipulated to have a spatially encoded time delay, and as a result they have the capability for single-shot measurements without scanning the optical delay.

In the pump-probe OKG measurements, an intense linearly polarized pump pulse passing through the nonlinear material will cause the refractive index change of the sample. A phase shift occurs between the probe field components polarized in parallel and perpendicular to the polarization plane of the pump pulse when the probe pulse overlaps with the pump pulse temporally. When the pump and probe pulses with equal wavelength are used in the pump-probe OKG experiments, OKG signals usually originate from two kinds of effects: the optical Kerr effect and the self-diffraction effect that results from laser-induced transient gratings (LITG).[21,22] Some studies have demonstrated that when femtosecond pulses are used in OKG measurements, Kerr signals result mainly from self-diffraction by the LITG.[21−23] As the self-diffraction effect is only limited by the duration of the pump laser pulse, the temporal behavior of the OKG signal is the field correlation time of the pump and probe pulses, while not the nonlinear optical response time of the materials. To acquire time-resolved OKG signals with a higher signal-to-noise ratio, it is important to control the contribution of self-diffraction effect to the time-resolved single-shot OKG measurements.

In this Letter, we investigate the self-diffraction and optical Kerr effects of CS_2 in femtosecond time-resolved single-shot OKG measurements by focusing the probe pulse using a cylindrical lens to introduce a spatially encoded time delay with respect to the pump pulse. The whole responses and relaxation of the OKG signals are obtained in single-shot OKG measurements. By measuring the pump power and polarization dependence of the OKG signals in CS_2, the contribution of self-diffraction effect which is independent of the nonlinear response time of the material was directly observed on the rising edge of the time-resolved OKG signals. The influence of the self-diffraction effect on the optical Kerr signal could be controlled effectively by varying the polarization angle between pump and probe pulses.

Figure 1 shows the experimental setup for femtosecond time-resolved single-shot OKG measurements. A Ti:sapphire amplifier system emitting 65 fs laser pulses centered at 800 nm at a repetition rate of 1 kHz with horizontally linear polarization was used. The laser beam was split into a pump and a probe

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beam by a beam splitter. The pump pulse passed through a $\lambda/2$ plate to change the polarization angle between the pump and probe pulses. The pump and probe beams were focused into a 2 mm cuvette filled with CS_2 by cylindrical lenses with focal lengths of 40 mm and 15 mm, respectively. The directions of the main axis of the elliptical pump and probe light spots were perpendicular to the experimental stage. The spot sizes of the focused pump beam were estimated to be 10 mm and 1 mm at the vertical and horizontal directions respectively, while those of the probe beam were estimated to be 10 mm and 0.5 mm. The peak energy density of the pump pulse was estimated to be below $0.34 \times 10^4\,\mathrm{MW/cm^2}$. The collimated pump and probe beams intersect with a finite angle of ~13° at the vertical direction. To ensure the sufficient interaction length of pump and probe beams, the crossed angle was set at ~2° at the horizontal direction. To avoid generating supercontinuum, the sample was placed about 4 mm behind the focal points of the pump and probe beams. The interaction area of probe and pump beams on the sample is much smaller than the spot size of the elliptical pump beam. Thus the pump intensity at the interaction area could be considered to be homogeneous and the influence of inhomogeneous intensity on the optical Kerr signal could be ruled out. Since the pump beam reaches different portions of the sample at different time, a time delay between the probe and pump beams was spatially encoded across the sample at the vertical direction. After passing through the sample, the polarization of the probe light changes, and only the components polarized parallel to the analyzer (A) placed behind the sample can be extracted. Finally, a real-time imaging of the time-resolved OKG signals could be obtained along the direction of the focal line of probe pulse. To record the image of OKG signals, we used a high-spatial-resolution CCD camera imaging overlapping plane of the pump and probe beams (P0). Due to the finite vertical crossing angle, the arrival time of the pump and probe pulses differ, depending on the position on the sample. As a result, the probe beams at different portions on the sample experience different-delayed pump beams. In other words, the delay time is mapped spatially across the sample plane. The mapping between the time coordinate t and the spatial coordinate y can be readily seen to be

$$t = \frac{y \times \sin\theta}{c}, \tag{1}$$

where y is the overlap length of probe and pump beams on the sample, θ is the vertical angle between the pump and probe beams, and c refers to the speed of light in air. For the pulse lengths of tens of femtoseconds, the resolution of the pump–probe single-shot technique mainly depends on the thickness of the sample. The relevant principle and numerical computations have already been reported.[24]

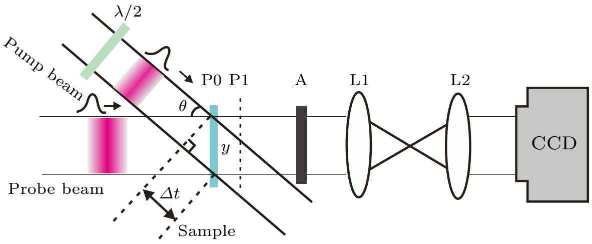

Fig. 1. Schematic diagram of the experimental setup, L1, L2: lens with a focal length of $f = 150\,\mathrm{mm}$, make up a $4f$ imaging system. A: analyzer, y: overlap length of probe and pump beams, Δt: delay time, P0: the imaging plane of the imaging system; and P1: the plane at 2 cm behind P0.

Firstly, we measured the single-shot time-resolved OKG signals by spatial-temporal transformation of the recorded signal images. Figures 2(a) and 2(b) show the normalized femtosecond time-resolved OKG signals in CS_2 when the pump powers were fixed at 40 mW and 60 mW, respectively. The polarization angle between the pump and probe pulses was kept at 45°. Under our experimental conditions, the contribution of empty cuvette to OKG signal was not ruled out. The insets show the signal images recorded by using a CCD camera. The fitted red solid line in Fig. 2 shows the decay process consisting of a fast decay and a slow decay which were attributed to the electron contribution and re-orientation contribution, respectively. The fast and slow decay times were estimated to be 0.25 ps and 1.2 ps respectively, agreeing well with the results measured by the conventional pump-probe OKG technique.[23,25] According to Eq. (1), the interaction area of probe and pump beams on the sample is estimated to be 4 mm at the vertical direction which is much smaller than the spot size of the elliptical pump beam. Thus the pump intensity at the interaction area could be considered to be homogeneous and the influence of inhomogeneous intensity on optical Kerr signal could be ruled out. In comparison of the temporal behavior of OKG signals at different pump powers we can see that there is a fast peak at the rising edge of the time-resolved signal and the peak at the rising edge becomes stronger with increasing the pump power. As has been demonstrated in some previous reports, the self-diffraction effect could easily take place in time-resolved OKG measurements when the pump and probe pulses with equal wavelength are used.[26−31] The temporal behavior of the self-diffraction effect is determined by the duration of the laser pulse rather than the nonlinear response of the material. Hence, the fast components of the OKG signals shown in Fig. 2 could be attributed to the self-diffraction effect. The temporal behavior of the OKG signal was obviously distorted due to the contribution of self-diffraction effect.

To further study the influence of the self-diffraction effect, the imaging setup of the signal-shot OKG measurements was adjusted to image the parallel plane (P1) about 2 cm behind the overlapping plane (P0) of the pump and probe beams. Figure 3(a) shows the recorded image of plane P1, and several light spots separated from the OKG light spot are observed. In the OKG measurements, the superposition of two coherent pulses yields a spatially modulated distribution of the energy density leading to the creation of the LITG and the diffractions of the incident beams.[27] As shown in Fig. 3(a), the imaged light spots are divided into upper and lower parts, corresponding to the diffraction spots of the probe and pump beams, respectively.

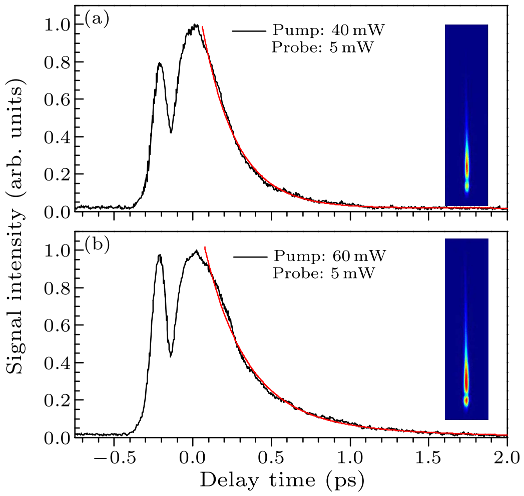

Fig. 2. Time-resolved OKG signals of CS_2 with pump powers fixed at (a) 40 mW and (b) 60 mW. The polarization angle between the pump and probe beams was kept at 45°. The inset shows the images of the transmitted probe beam at the imaging plane.

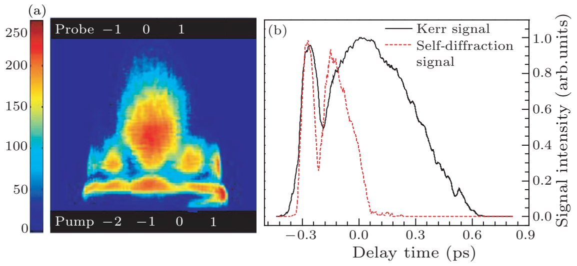

Fig. 3. (a) Self-diffraction signal separated from the optical Kerr signal. The upper and lower parts of diffraction orders correspond to the probe and pump beams, respectively. (b) Temporal behaviors of self-diffraction and optical Kerr signals.

As the 0 order of the probe light will experience the light-induced birefringence effect induced in CS_2, the diffraction order 0 is submerged by the middle luminous area which is responsible to the optical Kerr effect. Figure 3(b) shows the temporal behavior of self-diffraction (pump 0 order, and probe 1 order) and Kerr signal (probe 0 order, and pump −1 order), derived from the data shown in Fig. 3(a). The red and black lines in Fig. 3(b) show the temporal behaviors of self-diffraction signal and optical Kerr signal, respectively. Due to the fact that the optical Kerr effect in CS_2 mainly originates from the re-orientation of molecule, the response time is slower than the pulse duration, and the rising edge is slower. As the intensity of optical Kerr signal is still very weak when the contribution of self-diffraction vanishes, there is a dip in the trace between the peaks of self-diffraction effect and optical Kerr effect. We can see that the self-diffraction signal is independent of the response of the material, while the 0 order of probe light shows a slow decay process. As all the self-diffraction orders are blended into the Kerr signals in the OKG measurements, the contribution of the self-diffraction effect will influence the temporal behavior of the OKG signals.

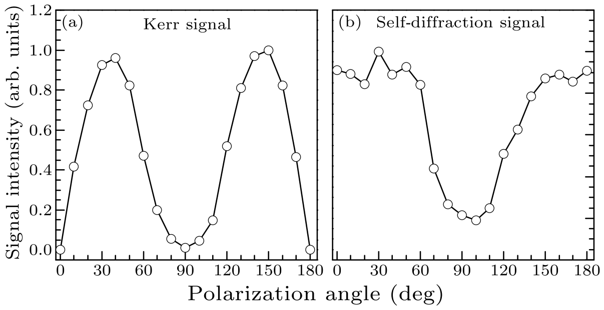

Fig. 4. The dependence of (a) the optical Kerr signal and (b) the self-diffraction signal on the polarization angle between the pump and probe beams.

To control the contribution of the self-diffraction effect in the OKG measurements, we investigate the polarization dependence of the OKG and self-diffraction signals, and the results are shown in Fig. 4. We can see from Fig. 4(a) that the period of polarization dependence of optical Kerr signal is $\pi/2$, with the maximum and minimum values occurring at $n\frac{\pi}{2}+\frac{\pi}{4}$ and $n\frac{\pi}{2}$ $(n=0,1,2\ldots)$, respectively. The experimental results that the polarization dependence of optical Kerr signal agree well with previous reports, and the OKG signal intensity can be expressed by[32]

$$I_{\mathrm{OKG}}=I_0\sin^2(2\theta)\sin^2(\Delta\varphi/2), \tag{2}$$

where I_{OKG} is the OKG signal intensity passing through the analyzer, I_0 is the probe beam intensity, and θ is the polarization angle between the pump and probe beams.

However, as the intensity of LITG effect is related to the coherence of the incident beams, the period of the polarization dependence of self-diffraction signal should be π. Figure 4(b) shows the polarization dependence of the self-diffraction signal, comparatively. The period of polarization dependence of the self-diffraction signal is π, with the maximum and minimum values occurring at $n\pi$ and $n\pi+\pi/2$ $(n=0,1,2,\ldots)$, respectively. Considering the different polarization dependences of the OKG and self-diffraction signals, it is possible to control the influence of self-diffraction effect on the optical Kerr signal by adjusting the polarization angle.

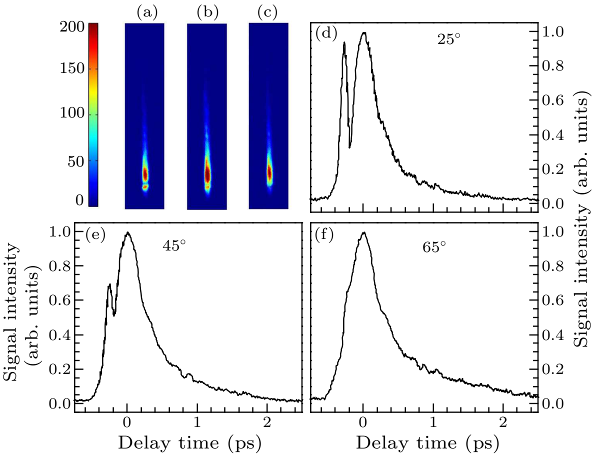

Fig. 5. The recorded signal images with polarization angle kept at (a) 25°, (b) 45° and (c) 65°. (d)–(f) The time-resolved OKG signals of CS_2 by spatial-temporal transformation of the recorded signal images in (a)–(c).

Figures 5(a)–5(c) show the recorded OKG signal images with the polarization angle kept at 25°, 45° and 65°, while Figs. 5(d)–5(f) show the time-resolved OKG signals by spatial–temporal transformation of Figs. 5(a)–5(c), respectively. Comparing the temporal behaviors of OKG signals at different polarization angles we can see that the contribution of the self-diffraction effect in the OKG measurements is decreased by increasing the polarization angle between the pump and probe beams. The fast peak at the rising edge of the time-resolved OKG signals almost disappears when the polarization angle is kept at 65°, as shown in Fig. 5(f). It should be noted that the self-diffraction effect is suppressed at the sacrifice of reduction of the OKG signal by increasing the polarization angle from 45° to 65°. The self-diffraction effect can be suppressed completely when the polarization angle is kept at 90°, while the OKG signal vanishes as well. The experimental results suggest that the contribution of the self-diffraction effect in the OKG measurements can be efficiently controlled when the polarization angle is kept around 65° in femtosecond time-resolved single-shot OKG measurements.

In conclusion, femtosecond time-resolved single-shot optical Kerr gating measurements have been performed by focusing the probe pulse using a cylindrical lens to introduce a spatially encoded time delay with respect to the pump pulse. By measuring the pump power and polarization dependence of the OKG signals in CS_2, the contribution of self-diffraction effect, which is independent of the nonlinear response time of the material, is directly observed on the rising edge of the time-resolved OKG signals. The self-diffraction signal is separated by adjusting the imaging setup to image the parallel plane behind the overlapping plane of the pump and probe beams. We investigate the different polarization angle dependence of self-diffraction and optical Kerr signals using this method. The influence of the self-diffraction effect on the optical Kerr signal could be controlled effectively by varying the polarization angle between pump and probe pulses.

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