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Femtosecond optical Kerr effect measurement using supercontinuum for eliminating the nonlinear coherent coupling effect

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Abstract
A nonlinear coherent coupling effect (NCCE) was observed in a degenerate femtosecond optical Kerr effect (OKE) measurement, which disturbed the analysis of the dynamics of the OKE response. The NCCE in the femtosecond OKE measurement was dependent on the pump power. To eliminate this unwanted influence of the NCCE, we proposed a two-color femtosecond OKE measurement method using supercontinuum, and the feasibility of this method has been demonstrated.

Keywords: femtosecond, optical Kerr effect, nonlinear coherent coupling effect, supercontinuum

(Some figures may appear in colour only in the online journal)

1. Introduction

The optical Kerr gate, making use of the photoinduced birefringence effect, has provided an interesting tool for scientific and potential practical applications, such as time-resolved spectroscopy [1], ballistic-photon imaging [2] and so on [3–6]. The studies on the photoinduced birefringence effect (commonly called the optical Kerr effect) of nonlinear optical materials are of importance not only to provide fundamental information for their applications but also to elucidate the dynamics of their nonlinear response [7, 8]. The procedure of the optical Kerr effect (OKE) measurement is more or less standardized. However, the results are not always consistent because of the experimental complexities [9, 10].

The previous studies show that 'optical Kerr signals' might originate from two kinds of effects in the non-collinear degenerate OKE measurements, the OKE and the coherent effect [11, 12]. The non-collinear OKE experiment is based on specialized pump–probe geometry. When the time separation of the pump and probe pulses is within the field correlation time, the two pulses coherently interfere with each other to form a transient grating [13]. Energy can be scattered from one beam to another, leading to an additional signal to the pure OKE signal. The signal originating from the coherent effect is only limited by the field correlation time of the pulses, rather than by the nonlinear response time of the materials. It could distort the real optical Kerr signals arising from the photoinduced birefringence effect, and in consequence make data analysis ambiguous. Previously, to reduce the influence of the unwanted coherent signals on the measurements of the nonlinear response, the two-color experiment was usually carried out. The frequency-doubled laser was commonly used in the two-color experiment [14, 15]. However, some samples might absorb the frequency-doubled pulse strongly, which would limit the performance of this method.

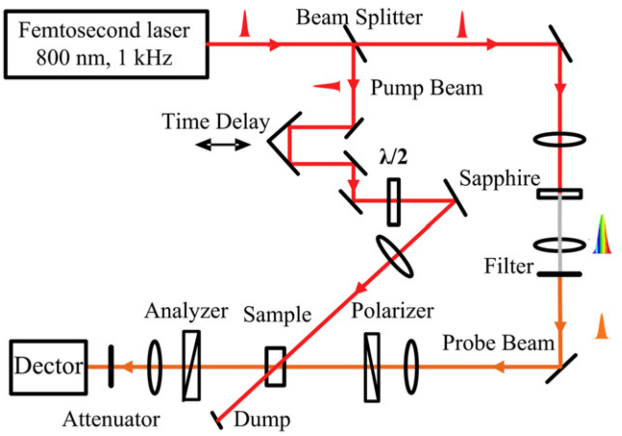

Figure 1. Schematic setup for the two-color OKE measurement using SC.

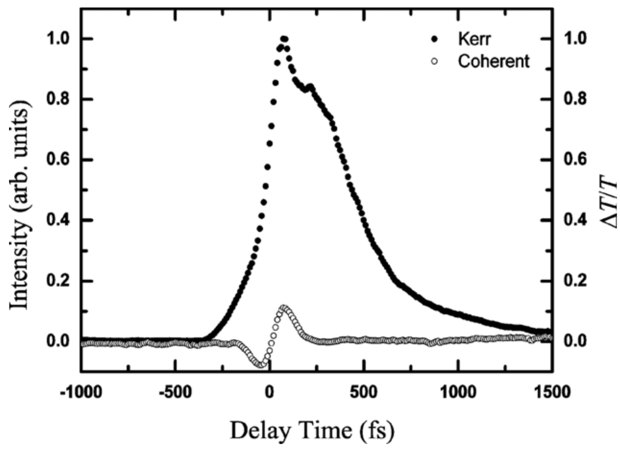

Figure 2. Time-resolved OKE signal (solid circle) and the NCCE signal (hollow circle).

In this paper, we investigate the nonlinear coherent coupling effect (NCCE) in the degenerate OKE measurement and observe that the NCCE had an evident influence on the OKE measurement, which was dependent on the pump power. To reduce this coherent artifact, we proposed a femtosecond two-color OKE measurement method using a supercontinuum (SC), in which a sapphire plate was used to generate the SC. The results show that the influence of the NCCE on the OKE measurement using SC was eliminated.

2. Experiments

A Ti:sapphire laser system (FEMTOPOWER compact Pro), emitting 30 fs, 800 nm laser pulses at a repetition rate of 1 kHz, was used in our experiments. The degenerate femtosecond OKE experiment was performed with the setup in [16]. The CS_2 solution in a quartz cuvette with a path length of 2 mm was used as the sample.

The two-color femtosecond OKE measurement using the SC is illustrated in figure 1. The emitting fundamental laser beam was separated into two parts by means of a beam splitter with a split ratio of 1:4. Passing through an optical delay-line, controlled by a stepping motor, the intense one was as the pump beam. The polarization of the pump beam was controlled by a half-wave plate. The weak beam was focused onto a 3 mm sapphire plate to generate a stable SC. The SC was collimated and passed through a narrowband-pass filter (OPTOSIGMA, CWL = 750 nm) as the probe beam. The narrowband-pass 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 two beams were focused onto the Kerr sample at a crossing angle of 12°. The signals were detected by a photodiode (EOT ET-2020) with the approximate saturation level of 42 mW mm^{-2}. Neutral-density filters were used in front of the photodiode to attenuate the light to avoid saturation.

3. Results and discussion

Using the degenerate OKE setup, we measured the time-resolved OKE signal, in which the pump power was fixed at 10 mW. As shown in figure 2, an ultrafast response component of about 200 fs exists in the leading edge of the time-resolved OKE signal. This result is in disagreement with the usual results [7, 17].

To investigate the physical mechanism of this ultrafast response component, we removed the analyzer from the degenerate OKE setup and measured the pump–probe signal. An energy transfer phenomenon was observed. The transient transmission change $\Delta T/T$ of the probe beam was measured as shown in figure 2. It is a typical NCCE signal for a material with a positive nonlinear index of refraction, in which light is scattered from the beam of higher frequency to that of low frequency [18, 19]. As our pulses have a positive chirp, instantaneously the probe beam has a lower frequency than the pump beam at the positive time delay and gains energy at the expense of the pump beam. On the contrary, the probe beam expended energy. Due to the coherent effect, energy can be scattered from one beam to another, leading to an additional signal to the pure OKE signal as shown in figure 2.

For the further investigation of the influence of the NCCE on the degenerate OKE measurement, we firstly measured the pump power dependence of the OKE signals intensity, when the delay time was adjusted to different values. As shown in figure 3, the triangles, circles and squares indicate the pump power dependence of OKE signals when the delay time was kept at −52 fs, 0 fs and +52 fs, respectively. The delay times of −52 fs and +52 fs correspond to the temporal positions of the valley and the peak of the NCCE signal, respectively.

The pure OKE signal intensity depends on the induced phase shift $\Delta\varphi$ and the polarization angle θ between the polarization planes of the pump and probe beams according to [20]:

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

When the polarization angle θ is fixed at 45°and the induced phase shift $\Delta\varphi$ is small enough, the pure OKE signal is proportional to the square of pump power P_p as follows:

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

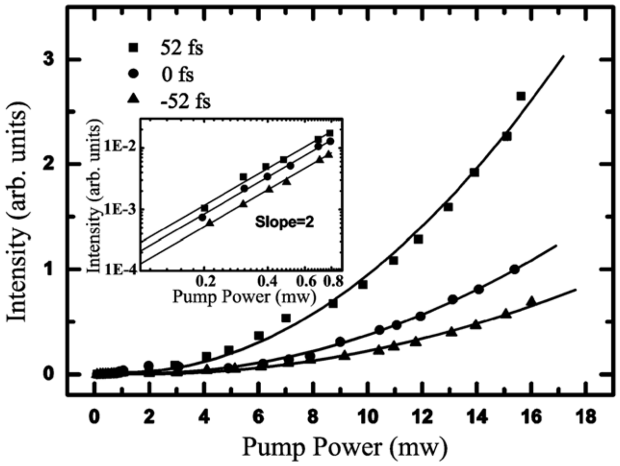

Figure 3. Pump power dependences of OKE signals at time delays of −52 fs (triangle), 0 fs (circle) and 52 fs (square) in the degenerate configuration.

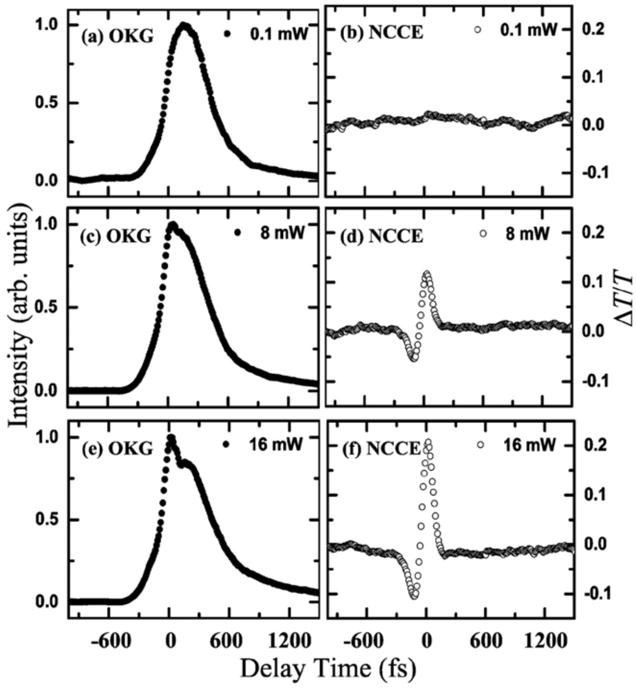

Figure 4. Time-resolved OKE signals and NCCE signals in the degenerate configuration at three different pump powers: 0.1, 8 and 16 mW.

where L_{eff} is the effective length of the sample, $\chi^{(3)}$ 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 data of 0 fs time delay agree well with the theoretical results (solid line) according to equation (2), which is a quadratic curve as shown in figure 3. However, the OKE signals at 52 and −52 fs with the pump power above 1 mW show different pump power dependences because of the additional signals induced by the NCCE. The power exponent of the fitting curve for the experimental data (square) at 52 fs is about 2.35 because of the positive additional signals of the NCCE, while the power exponent of the fitting curve for the experimental data (triangle) at −52 fs is about 1.78 because of the negative additional signals of the NCCE. In addition, the enlarged plots of the data with the pump power below 1 mW are given in the inset in figure 3 with the double logarithmic coordinates. The results show that the detected signal intensities increase quadratically with the pump power below 1 mW, which indicates that the signals mainly arise from the OKE in this condition. However, the detected signals at the time delay of 52 and −52 fs deviate from the quadratic curve more rapidly when the pump power was further increased due to the contribution from the NCCE.

Furthermore, to investigate the influence of the NCCE on the dynamics of the OKE response, we measured the time-resolved OKE signals and NCCE signals at three typical experimental conditions with the pump power of 0.1 mW, 8 mW and 16 mW, respectively. The results are shown in figure 4. The transient transmission changes $\Delta T/T$ of the NCCE signals increased with increasing pump power as shown in figures 4(b) and (d) and (f). When the pump power was kept at 0.1 mW, there is no influence of the NCCE on the degenerate OKE measurement, as shown in figure 4(a). When the pump power was further increased, the distortion of the time-resolved OKE signal gradually became serious and the leading edge of the time-resolved OKE signal became steep, as shown in figure 4(c). When the pump power was increased to 16 mW, a distinct distorted peak formed on the leading edge of the time-resolved OKE signal which was caused by the NCCE as shown in figure 4(e). This would seriously obscure the dynamics of the OKE induced by the femtosecond laser. These results suggest that the influence of the NCCE on the degenerate OKE measurement will increase with the increasing pump power.

In addition, the instantaneous signals in figures 4(c) and (e) may be due to the electronic response, which can be found in the OKE measurement if the cross-correlation width between the pump and probe pulses is short enough (<100 fs). To investigate this possibility, we measured the time-resolved OKE signals of the quartz in the same experimental configuration with two different pump powers (10 and 2 mW) as shown in figure 5. Because the OKE signal of the quartz mainly originates from the electronic response, it can characterize the cross-correlation width between the pump and probe pulses. From figure 5, we can see that both the cross-correlation widths were measured to be about 350 fs (FWHM) because of the dispersion of the optical elements in our experiment and the cross-correlation widths do not vary with the pump powers. The experimental results eliminate the possibility that the instantaneous signals in figures 4(c) and (e) were caused by the electronic response.

In order to eliminate the influence of the unwanted NCCE on the OKE measurement, we filtered the SC to generate a new probe beam to perform the two-color OKE experiment using SC as mentioned above. The new probe beam has a center wavelength of 750 nm with a nearly Gaussian spectral profile. The spectral bandwidth of the probe

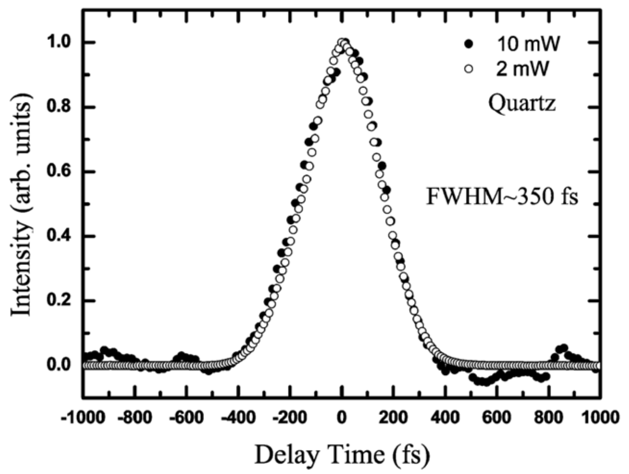

Figure 5. Time-resolved OKE signals of the quartz in the degenerate configuration at two different pump powers: 10 and 2 mW.

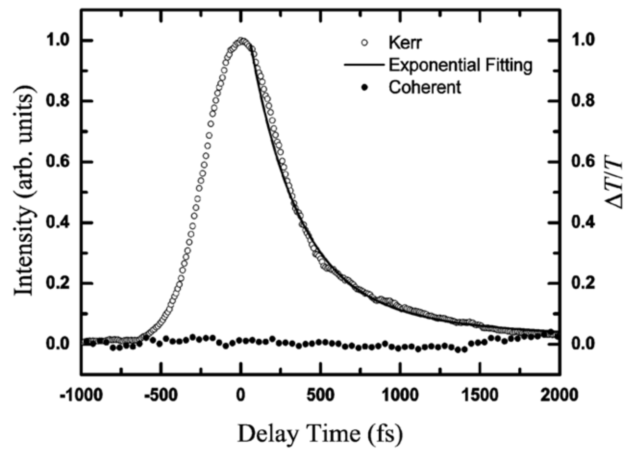

Figure 6. Time-resolved OKE signal (hollow circle) and NCCE signal (solid circle) in the two-color configuration.

beam is measured to be about 24 nm (FWHM) and nearly has no coherent component to the fundamental frequency beam. Measurements on the time-resolved OKE signal and the NCCE signal were conducted in the two-color configuration with a pump power of about 10 mW, as shown by the hollow circle and the solid circle in figure 6.

From figure 6, we can see that the transient transmission change $\Delta T/T$ of the NCCE signal is close to zero, which means that there is no influence of the NCCE on the two-color OKE measurement using SC. We fitted the trailing edge of the time-resolved OKE signal in figure 6 approximately by a dual-exponential function with lifetimes of 300 fs and 1.6 ps with relative amplitude ratios of 7.6:1. The previous femtosecond optical Kerr studies on CS_2 show that the slow response of the OKE for CS_2 is identified with molecular orientation and the fast one is attributed to the electron polarization, molecular librational motion and so on [21, 22]. So the dynamics of the OKE response for CS_2 measured in the two-color OKE experiment using SC are reasonably consistent with these previous reports. The results demonstrated that the influence of the NCCE can be effectively eliminated in the two-color OKE measurement using SC.

It should be noticed that our simulation shown in figure 6 deviates slightly from the measurement data. The reason is that the dual-exponential curve fitting is a simplified approach to investigate the Kerr responses in CS_2. For example, the electron polarization, contributing to the fast exponentially decaying component, is an instantaneous response, which actually follows the intensity profile of the laser pulse cross-correlation. For the detailed molecular dynamic simulation for the femtosecond laser-induced Kerr responses in CS_2 please refer to [21].

4. Conclusions

In conclusion, we have investigated the influence of NCCE on the degenerate femtosecond OKE measurement. The results show that the proportion of the NCCE signal in the measured signal became higher with the increase of the pump power. A two-color femtosecond OKE measurement method using SC has been demonstrated to be applicable to eliminating the influence of the NCCE.

Acknowledgments

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