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Application of Optical Kerr Gate with $SrTiO_3$ Crystal in Acquisition of Gated Spectra from a Supercontinuum *

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We investigate the optical Kerr effect of $SrTiO_3$ (STO) crystal, of which the nonlinear response time was measured to be less than 200 fs, while the nonlinear refractive index is estimated to be 2.16×10^{-15} cm^2/W. Using the optical Kerr gate (OKG) technique with an STO crystal as the Kerr medium, we obtain narrow-bandwidth and symmetric gated spectra from a supercontinuum generated in distilled water by a femtosecond laser. The experimental results show superiority compared with the gated spectra obtained using OKG with CS_2 as the Kerr medium, demonstrating that STO crystal is a promising OKG medium.

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With the wide use of the femtosecond laser, the ultrafast optical Kerr gate (OKG) technique has been developed as a key tool to investigate the ultrafast fluorescence spectroscopy, high time-resolved imaging, ballistic light imaging, and Kerr gated fluorescence microscopy, etc.[1−4] The OKG technique can provide an ultrafast measurement of broad wavelength range, ultrafast switching time, and high precision. As the key factor to construct an OKG configuration, a suitable Kerr medium should be of a large nonlinearity and ultrafast switching time. Over the last few decades, great efforts have been made in the research and development of nonlinear optical materials with large nonlinearities, high transparency, and fast response time.[5−7]

Supercontinuum generated from the propagation of ultrashort laser pulses in transparent media has found numerous applications in such diverse fields as spectroscopy, pulse compression, and design of tunable ultrafast femtosecond laser sources.[8,9] It is well known that supercontinuum has a strong chirp owing to dispersion of the material in which supercontinuum is generated. Until now, the self-diffraction frequency-resolved optical gating (SD-FROG) method,[10] two-photon absorption[11] and the optical Kerr gate (OKG) method[12] have been used to investigate chirp characteristics. Compared with other methods, the OKG technique has many advantages such as no need for satisfaction of the phase-matching condition or high intensity of the probe pulse.

$SrTiO_3$ (STO) is a promising wide band gap material and appears to be excellent in the field of electronic industry. As STO crystal provides a good lattice match to most materials with the perovskite structure, it has been widely used for special optical windows and as high quality sputtering targets.[13,14] Over the last few years, very little work has been devoted to its nonlinear optical properties, whereas STO crystal has been mainly used for dielectric devices and optical substrates. In the previous reports, Deng *et al.* investigated the nonlinear optical properties of STO thin films as well as their optical limiting behaviors.[15] In 2004, Nakamura and Kanematsu measured the nonlinear refractive index of STO crystal using z-scan technique, which was found to be highly efficient and suitable as a Kerr gate material.[16] In 2011, Yu *et al.* compared the OKG property of STO crystal and other crystalline media.[17] Because they used a collinear OKG arrangement, the group delay dispersion (GDD) effect would limit the performance of the STO crystal, as has been discussed in Ref. [16].

In this Letter, utilizing a femtosecond laser, we investigate the optical Kerr effect in STO crystal. The switching time of STO crystal is measured to be less than 200 fs, and the nonlinear refractive index is estimated to be 2.16×10^{-15} cm^2/W. Using the optical gate with the STO crystal as the Kerr medium, we obtain narrow-bandwidth and symmetric gated spectra from the supercontinuum generated in distilled water by a femtosecond laser. The experimental results show that the gated spectra obtained using OKG with the STO crystal as the Kerr medium have superiority,

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compared to those obtained with CS_2, demonstrating that STO crystal is a promising OKG material.

In our experiment, ultrafast optical Kerr measurements were carried out on a nondoped STO crystal plate (Furuuchi Chemical Co.). The sample was polished to be about 1 mm thick with (100) surface. A 1-mm-thick fused silica (FS) plate and liquid CS_2 filled in a fused silica cell with path length of 1 mm were used as the reference samples. A laser with 30-fs pulse duration at 800 nm operating at 1 kHz was employed in our experiments. The output of the laser was split into two beams, one was used as the gating beam, and the other was used as the probe beam. The pump power was adjusted to be about 15 mW and the power ratio of the pump and probe pulse was adjusted to be about 15:1. The Kerr sample was positioned between a polarizer and an analyzer in a cross Nicole polarizer configuration. The two beams were focused into the sample at an angle of 10° and the spots of the focused beams were carefully spatially overlapped. The pump beam was focused into the sample with a $1/e^2$ beam radius of about 150 µm. To avoid the continuum generation, the input pump power was kept below 20 mW, with a maximum fluence of about $0.056\,\mathrm{J/cm^2}$. A computer controlled delay line was introduced into the optical path of the gating beam. The OKG signal was detected by a photomultiplier tube (PMT). To optimize the OKG signals, the polarization of the gating beam was rotated by 45° with respect to the linear polarization of the probe beam.

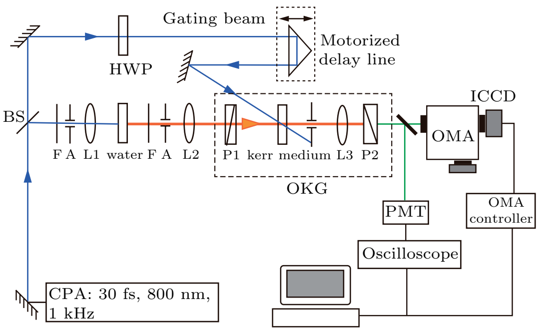

Fig. 1. Scheme of acquisition of gated spectra using OKG technique. BS: beam splitter, HWP: half-wave plate, P: polarizer, A: aperture, L: lens.

The experimental setup of acquisition of gated spectra using the OKG technique is schematically represented in Fig. 1. To generate the supercontinuum, the fundamental probe pulse was focused into a 10-mm cell filled with distilled water using a 100-mm lens. A short-wave pass filter was used to remove the 800 nm light and the infrared part. The supercontinuum was focused into the Kerr medium along with the gating pulse. When the Kerr gate was opened by the gating pulse, gated spectra passing through the analyzer were collected by lenses and finally detected by a PMT and an optical multi-channel analyzer (OMA).

The open circles and closed triangles in Fig. 2 show the time-resolved optical Kerr signals for STO crystal and fused silica, respectively. The full width at half-maximum (FWHM) of time-resolved Kerr signals for both materials was estimated to be 200 fs, which had no slow component in the decay process. The temporal profile of the Kerr signals in STO crystal can be well fitted using the Gaussian function, as shown by the dashed line in Fig. 2. As the laser pulse was broadened by the elements in the optical path, the pulse width measured by autocorrection at the sample was estimated to be about 200 fs. This indicates that the response curve of STO crystal could be determined by the pulse width and the origin of the nonlinear response of the STO crystal would be attributed to the electronic process. The solid line in Fig. 2 shows the time-resolved optical Kerr signals for CS_2, which has a slow component lasting about 1.5 ps.[18] The open squares in the inset of Fig. 2 show the optical Kerr signals as a function of the power of the gating light. The solid line in the inset shows a quadratic dependence of optical Kerr signals on the gating pulse energy.[5,7]

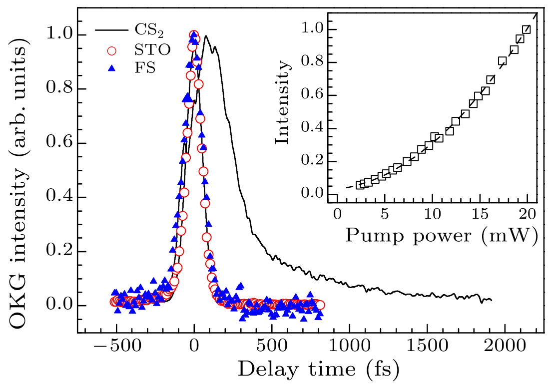

Fig. 2. Time-resolved measurements of optical Kerr signals for the STO crystal (open circles), fused silica (closed triangles) and CS_2 (solid line), respectively. The open squares in the inset show the Kerr signals as a function of the power of the gating light.

The OKG signal intensity of STO crystal was measured to be about 54 times larger than that of fused silica at the same pump power. This value is much larger than the results measured by Yu *et al.*, as a collinear two-color arrangement was used in their experiments, and the transmittance of the OKG was greatly affected by the GDD effect.[19] The following equation was employed to calculate the third-order nonlinear refractive index n_2 of the Kerr sample:[5,7]

$$n_2 = n_{2,R}\Big(\frac{(T_{\rm OKG}/T_0)_S}{(T_{\rm OKG}/T_0)_R}\Big)^{1/2}\frac{L_{\rm eff,R}}{L_{\rm eff,S}}, \qquad (1)$$

where the subscripts of the S and R indicate for STO crystal and the reference sample, respectively. $T_{\rm OKG}$ is the transmittance of the OKG setup, T_0 is the transmittance of the sample, and $L_{\rm eff}$ is the effective length of the sample. The nonlinear refractive-index n_2 of the STO crystal was estimated to be $2.16\times10^{-15}\,\mathrm{cm^2/W}$ by comparison with that of fused silica of $2.5\times10^{-16}\,\mathrm{cm^2/W}$ at 804 nm, which was used as a calibration standard.[19] However, as the STO crystal has a large nonlinear absorption coefficient, the intense pump light might be greatly absorbed due to the two-photon absorption effect. This would decrease the induced nonlinear phase shift in the OKG measurements. Hence, the difference between the nonlinear refractive index of the STO crystal and reference sample seems much smaller than that reported in Ref. [16], in which a z-scan technique was used to evaluate the nonlinearity of the sample.[16]

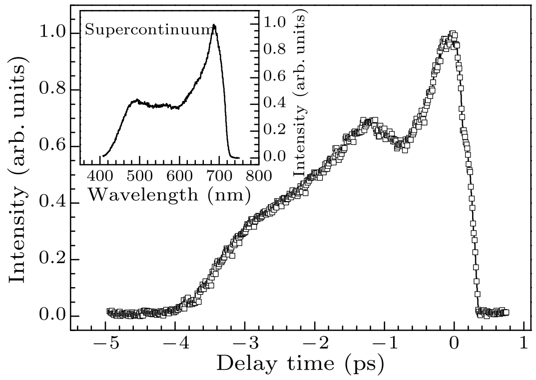

Fig. 3. Temporal profile of supercontinuum pulse generated in distilled water measured by OKG with STO crystal. The inset shows the supercontinuum spectrum.

Because of the large nonlinearity and fast response time, STO crystal was used as the Kerr gate medium and a series of gated spectra were obtained from the supercontinuum using the OKG method. To reduce the GDD effect, the interacting angle between the pump and probe beams was fixed at 10°. A supercontinuum generated in distilled water by a femtosecond laser was used as the probe light. After passing through a short-wave pass filter, the probe light ranged from 400 nm to 720 nm, as shown in the inset of Fig. 3. The squares in Fig. 3 indicate the temporal profile of the supercontinuum pulse measured using the OKG method. The duration of the supercontinuum was estimated to be about 4 ps, as the chirped pulse was broadened by the water and optical elements in the setup due to the group velocity dispersion (GVD) effect.

Figure 4(a) show the Kerr gated spectra using STO crystal as the Kerr medium. By finely controlling the delay time between the gating and probe pulses, the central wavelength of each spectrum was adjusted to 480 nm, 500 nm, 550 nm, 600 nm, 650 nm, and 700 nm, respectively. Figure 4(b) show the Kerr gated spectra using CS_2 as the Kerr medium. From the figure, we can see that the gated spectra obtained using OKG with STO crystal have much narrower bandwidth and better symmetry than those obtained with CS_2. As shown in Fig. 2, the Kerr signal for STO crystal shows no slow response component compared with that for CS_2, hence, it is easy to understand the narrow-bandwidth and symmetric properties of the gated spectra. To obtain even narrower gated spectra using the OKG with STO crystal, the supercontinuum was temporally broadened by passing through a 10-mm quartz. Figure 4(c) show the gated spectra from temporally broadened supercontinuum, from which we can see that the bandwidth of the gated spectra became much narrower. The average bandwidth of the gated spectra was estimated to be about 16 nm.

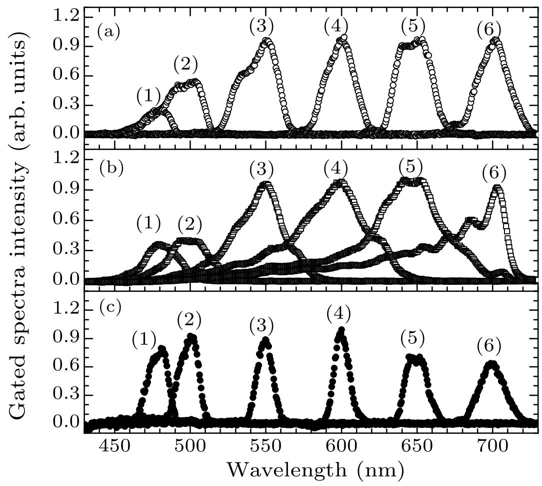

Fig. 4. Kerr gated spectra at different delay times. The spectra shown by (1), (2), (3), (4), (5), and (6) are centered at 480 nm, 500 nm, 550 nm, 600 nm, 650 nm, 700 nm, respectively. (a) STO crystal as the Kerr medium, (b) CS_2 as the Kerr medium, (c) STO crystal as the Kerr medium, while the supercontinuum was broadened by a 10-mm quartz.

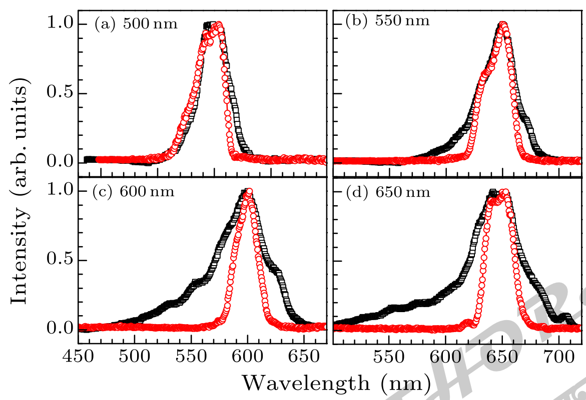

Fig. 5. Gated spectra obtained from supercontinuum using CS_2 and STO crystal as the Kerr medium, respectively. The gated spectra shown in (a), (b), (c), and (d) are centered at 500 nm, 550 nm, 600 nm, and 650 nm, respectively.

Figure 5 shows the four pairs of gated spectra selected from Fig. 4, which were centered at 500 nm, 550 nm, 600 nm, and 650 nm, respectively. From the figure, we can see that the gated spectra for CS_2 show much broader bandwidths and exhibit obvious band tailing compared with those for STO crystal. For the gated spectrum centered at 500 nm, however, STO crystal does not show obvious superiority compared with CS_2. Two factors are attributed to such phenomena. First, the supercontinuum shows low chirp characteristic in the short-wave region.[10−12] Second, the probe light intensity decreases sharply with the decreasing wavelength from 480 nm, as shown by the inset of Fig. 3. Hence, no component could experience the relaxation process of the birefringence induced in CS_2. Generally, the gated spectra obtained by OKG with STO crystal show better symmetry and have narrower bandwidth compared to those obtained with CS_2.

In conclusion, we have investigated the optical Kerr effect of STO crystal, the switching time is measured to be less than 200 fs. The response time of STO crystal is comparable with that of fused silica, while the nonlinear refractive index is estimated to be as large as $2.16 \times 10^{-15}\,\mathrm{cm^2/W}$. Using OKG technique with a STO crystal as the Kerr medium, we obtain narrow-bandwidth and symmetric gated spectra from the supercontinuum generated in distilled water by a femtosecond laser. The experimental results show superiority compared with the gated spectra obtained by OKG with CS_2. The average bandwidth of the gated spectra will be even narrower if the supercontinuum is temporally broadened.

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