Acquisition of gated spectra from a supercontinuum using ultrafast optical Kerr gate of lead phthalocyanine-doped hybrid glasses

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Abstract: We demonstrated the selection of the chirped supercontinuum using an ultrafast optical Kerr gate (OKG) of lead phthalocyanine (PbPc)-doped hybrid glasses. Using the OKG, narrow bandwidth and symmetrical gated spectra were obtained continuously from the chirped supercontinuum generated in a sapphire plate with a femtosecond laser. Experimental results show that the obtained Kerr-gated spectra using the PbPc-glass have many advantages comparing with that using CS_2.

OCIS codes: (190.4400) Nonlinear optics, materials; (190.3270) Kerr effect; (190.7110) Ultrafast nonlinear optics; (320.2250) Femtosecond phenomena; (320.7100) Ultrafast measurements.

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1. Introduction

Supercontinuum (SC) generation from the propagation of ultrashort laser pulses in transparent media is a well-known phenomenon [1,2] and is very attractive for some possible applications [3,4,5]. This phenomenon has been observed in lots of gaseous [6,7], liquid [8] and solid [9] transparent optical medium and found extensive applications in femtosecond time-resolved spectroscopy [10], atmospheric sensing [11,12], broadband spectrum lidar [13], high time-resolved imaging [14,15,16], lightning control [17] , and so on. It is well known that the SC has a strong chirp owing to dispersion by the material in which it is generated. Until now, the sum-frequency generation cross-correlation method [18], two-photon absorption [19] and the optical Kerr gate (OKG) technique [20,21] were used to investigate its chirp characteristics. Comparing with other methods, the OKG technique has many advantages, such as no need of satisfaction of the phase-matching condition or high intensity of the probe pulse. Based on this technique and the chirp characteristics of the SC, Minoshima et al. proposed a method for simultaneous three-dimensional (3-D) imaging [14]. This method makes full use of the advantages of the OKG, but also avoids scanning of the laser. It is applicable for imaging of the shape of moving objects, or surface testing and inspection. However, when going further into the femtosecond region, there is an inherent trade-off between high sensitivity and fast response in all the application based on the OKG technique.

Among the available nonlinear optical materials, metallophthalocyanines (MPcs) seem to be preferable candidates for the above applications, due to their large nonlinearities, high transparency, and fast response time. Generally, MPcs with two-dimensional conjugated π-electrons delocalization exhibit large third-order NLO susceptibility due to the modified chemical and molecular structures [22,23]. There is considerable interest in synthesizing bulk optical materials containing MPcs by the sol-gel method to increase the laser-materials

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interaction length for compact-sized optical devices. Sol-gel processing is a significant technique for developing the organic-inorganic hybrid nanocomposites at low temperatures, at which organic molecules may not be decomposed. Thus, the nonresonant-type bulk materials with MPcs synthesized by sol-gel method are expected to have large NLO susceptibilities, ultrafast response time, low optical losses, and long interaction length [24]. Using Kerr media with ultrafast response time, which result in narrow and symmetrical Kerr-gated spectra, may characterize the chirped structure of SC more exactly or improve spatial resolution in simultaneous 3-D imaging using chirped ultrashort light pulses.

In this paper, we demonstrated the selection of the chirped SC using an ultrafast OKG of lead phthalocyanine (PbPc)-doped hybrid glass, the nonlinear response time of which was measured to be less than 120 fs. Using the OKG, narrow bandwidth and symmetrical gated spectra were obtained continuously from the chirped SC generated in a sapphire plate with a femtosecond laser. Experimental results show that the obtained Kerr-gated spectra using the PbPc-glass have many advantages comparing with that using CS_2.

2. Experiments

The PbPc-doped bulk material was synthesized by hydrolysis condensation of the organically modified precursors vinyltriethoxysilane $H_2C{=}CHSi(OC_2H_5)_3$(VTES) in ethanol under acid (HCl)-catalyzed hydrolysis and basic-catalyzed condensation. Details of preparation processing were described in the literature [25]. The PbPc dye was dissolved in DWF(N, N-dimethyl formamide)/ethanol mixed solvent and sonicated for about 1 h and then introduced into sol-gel precursor solution at different initial concentration of 1.25×10^{-4} mol/l corresponding to 0.06 wt.%. The third-order nonlinear refractive index of the PbPc-glass was measured to be about 2.4×10^{-13} esu, which only was 1.40 times larger than that of fused quartz [26] due to the low concentration of PbPc**.** It can be expected that the third-order nonlinear refractive index of the PbPc-glass can be increased by increasing the concentration of PbPc. The derived PbPc-doped gel glasses were yellowish and cylindrical transparent material with 0.6 mm in thickness. Two weak linear absorption peaks at 601 and 688 nm were observed in the range of 450-700 nm and no obvious linear absorption at 800 nm [27].

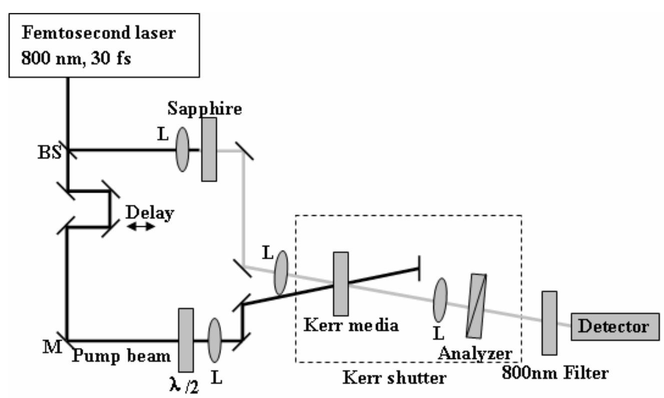

Fig. 1. Experimental setup for picking gated spectra from the chirped supercontinuum by optical Kerr gate. BS: beam splitter; L: lens; M: mirror.

The experimental setup is shown in Fig. 1. The multi-pass amplified Ti:sapphire laser, which emitted 30 fs, 800 nm laser pulses at a repetition rate of 1 kHz, was split into two beams. One beam which had 30% of the total power (10 mW) was focused into a sapphire with 3 mm thickness by a 10 cm focal-length lens and generated the SC served as probe beam. The other beam (pump beam), which had 70% of the total power, passed through a time-delay device and a $\lambda/2$ plate to control the path length and polarization of the pump beam, respectively. Both beams were focused by lenses before they passed through the Kerr media.

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The centers of the pump beam and SC spatially overlapped at the Kerr media at an angle of 25°. A polarizer was placed behind the sample in a cross-Nicol configuration, so that the SC could not pass through the polarizer without the pump beam. The SC light transmitted after the Kerr media was collimated with a 100 mm focal-length lens before passing through the analyzer. The polarization of the pump beam was rotated by $\lambda/2$ plate from that of the SC to optimize the intensity of the Kerr signals. A time-delay device, which was controlled by a computer, was used to adjust the timing of pulse collisions. The Kerr media we used here were CS_2 and the PbPc-glass. A low-pass filter was used to remove the 800 nm beam and the infrared contribution from the SC. As time-delay device moving, the temporal signals and spectral signals were detected by an optoelectronic diode and a spectroscope, respectively.

3. Results and discussion

Using the OKS setup in Ref. [28], we measured the response time of the PbPc-glass and CS_2 at the wavelength of 800 nm, respectively. The results are shown in Fig. 2, from which, we can see that the nonlinear response time of the PbPc-glass is less than 120 fs and much faster than that of the reference sample CS_2 solution.

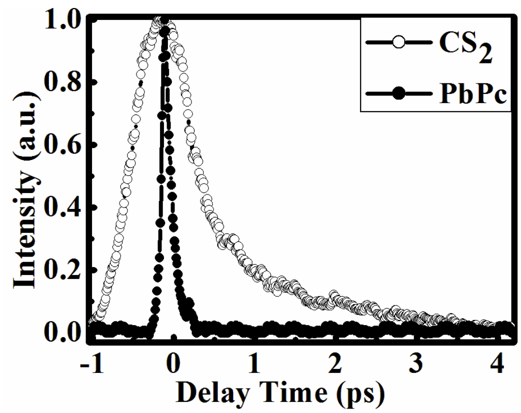

Fig. 2. Time-resolved measurements of Kerr signals for the PbPc-glass (closed circles) and CS_2 (open circles).

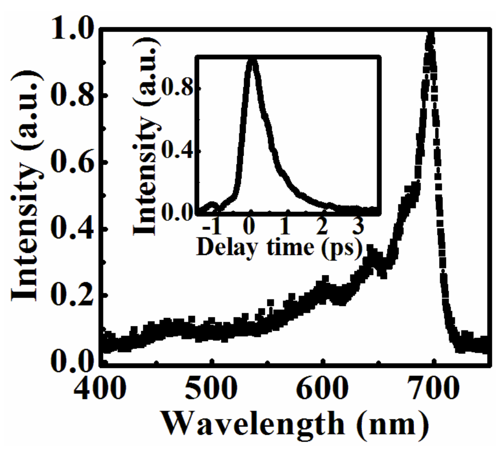

Fig. 3. Supercontinuum generated in a sapphire plate and time-resolved measurements (inset) for the supercontinuum duration using OKG with the PbPc-glass.

Figure 3 shows the SC generated in the sapphire plate with a femtosecond laser, and the

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spectrum of the SC ranges from 450 to 700 nm with a temporal span of 2.8 ps. The temporal span for the SC pulses was measured using OKG with the PbPc-glass, and the result is shown in the inset of Fig. 3. The generated SC is up-chirped mainly by group velocity dispersion in the sapphire itself, the lenses, the Kerr media, and the air [29]. Figure 4(a) and 4(b) show the Kerr-gated spectra of SC when CS_2 and PbPc were used as Kerr media, respectively. The peak wavelengths of the Kerr-gated spectra are 480 nm, 500 nm, 550 nm, 600 nm, 650 nm, and 700 nm, respectively, which were detected at different delay times of the pump laser. The neighboring intervals of gated spectra at 480 nm, 500 nm, 550 nm, 600 nm, 650 nm, 700 nm for CS_2 in Fig. 4(a) were 0.40, 0.50, 0.35, 0.20, 0.15 ps, respectively, and that for PbPc-glass in Fig. 4(b) were 0.40, 0.60, 0.40, 0.25, 0.20 ps, respectively.

From Fig. 4, we can see that all the Kerr-gated spectra have a long trailing edge, and the obtained gated spectra using OKG of the PbPc-glass have much narrower bandwidth and better symmetry than that of CS_2. This can be understood according to Fig. 2. From Fig. 2, we can see that the Kerr signals of the PbPc-glass shows no slow component and the full width at half maximum (FWHM) of the pulse correlation is 120 fs, which is much faster than that of CS_2. Thus, the obtained Kerr-gated spectra using the PbPc-glass have many advantages comparing with that using CS_2.

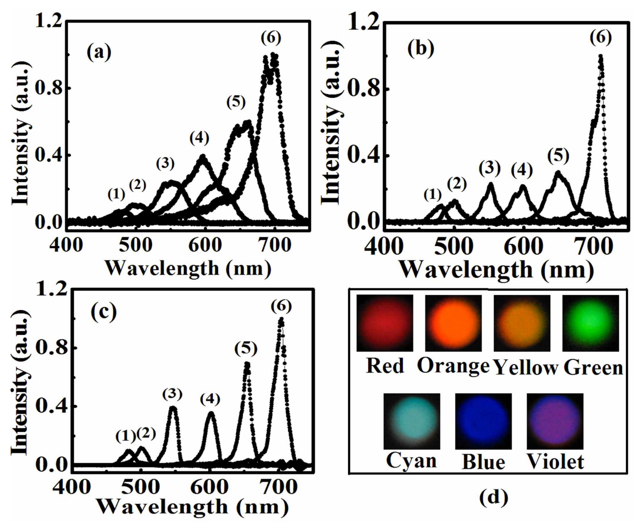

Fig. 4. Kerr-gated spectra at different delay times. The peak wavelengths for the spectra of (1), (2), (3), (4), (5), and (6) are 480 nm, 500 nm, 550 nm, 600 nm, 650 nm, 700 nm, respectively. (a) CS_2 as Kerr media, (b) PbPc-glass as Kerr media, (c) PbPc-glass as Kerr media, where the SC was broadened by a 40 mm long quartz. (d) Beam patterns of the obtained Kerr-gated spectra at different delay times.

Furthermore, we temporally broadened the SC by passing it through a quartz with 40 mm thickness to get much narrower in SC spectral selection by using the PbPc-glass. The results are showed in Fig. 4(c). The neighboring intervals of gated spectra at 480 nm, 500 nm, 550 nm, 600 nm, 650 nm, 700 nm are 1.00, 1.45, 1.30, 0.90, 0.65 ps, respectively. From Fig. 4(c), we can see that the spectral bandwidth of the gated spectra become much narrower when the SC was broadened by quartz. Figure 4(d) shows the beam patterns of the obtained Kerr-gated spectra at different delay times.

To further compare the difference of the obtained gated spectra using CS_2 and the PbPc-glass, we evaluated the FWHM of the Kerr-gated spectra shown in Fig. 4 with Gaussian curve fit method. Figure 5 shows the FWHM of the gated spectra presented in Fig. 4(a), (b), (c) and the gated spectra at the peak wavelength of 550 nm for the three cases. From Fig. 5(a), we can clearly see the FWHM of the Kerr-gated spectra by using CS_2 as Kerr media ranges from 30 nm to 60 nm, while the bandwidths of the gated spectra for the PbPc-glass are narrower than

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30 nm. When the PbPc-glass was used and the SC was broadened by the 40 mm long quartz, the FWHM of the Kerr-gated spectra reduced to below 20 nm. From Fig. 5(b), we can clearly see the shape of the obtained gated spectra using the PbPc-glass is more symmetrical and the spectral bandwidth is 23 nm in FWHM. The obtained gated spectrum using CS_2 exhibits obvious band tailing and spectral bandwidth of 43 nm. When the PbPc-glass was used and the SC was broadened by the 40 mm long quartz, the FWHM of the Kerr-gated spectra reduced to 17 nm.

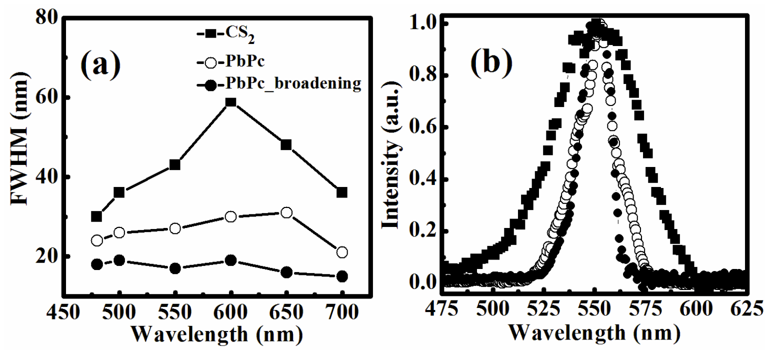

Fig. 5. Comparison of the obtained Kerr-gated spectra using CS_2 (closed squares), PbPc-glasses (open circles), and PbPc-glasses where the SC was broadened (closed circles). (a) Variation of FWHM of the Kerr-gated spectra as a function of the wavelength. (b) Comparison of the Kerr-gated spectra at the peak wavelength of 550 nm.

Due to the group velocity dispersion, the fundamental pulse duration in our experiments broadened to about 350 fs at the samples, which made the Kerr gate broaden. The broadened Kerr gate resulted in that there performed only a factor of ~2 between the gated spectral bandwidths of PbPc-glass versus CS_2. It can be predicted that the bandwidth of the obtained gated spectra using the PbPc-glass can decrease significantly if the fundamental pulse is compressed further. That means we could improve the time resolution and the longitudinal resolution in the 3-D imaging based on the OKG of the glass. In addition, this ultrafast-response and strong-signal Kerr gate can be used to capture the transient fluorescence and handle optical signals in future integrated optical systems.

4. Conclusion

In this paper, the ultrafast nonlinear optical response of the PbPc-glass was investigated using a femtosecond OKS setup. We demonstrated the selection of the chirped supercontinuum using an ultrafast OKG of the PbPc-glass, the nonlinear response time of which was measured to be less than 120 fs. Using the OKG of the PbPc-glass, narrow bandwidth and symmetrical gated spectra were obtained continuously from the chirped SC generated in a sapphire plate with a femtosecond laser.

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