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Efficient optical Kerr gate of tellurite glass for acquiring ultrafast fluorescence

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Abstract
We investigated the ultrafast optical Kerr effect of tellurite glass (Te glass) by the femtosecond time-resolved pump–probe technique. The Kerr-gated spectra from a chirped supercontinuum (SC) obtained by a Te glass optical Kerr gate (OKG) were of a narrow bandwidth and wide range of transmittance spectra. Time-resolved violet fluorescence of a ZnO thin film with high temporal resolution was measured using the Te glass OKG. Experimental results showed that Te glass was a good candidate for an OKG medium due to its ultrafast response, large transmittance window, and large nonlinear refractive index.

Keywords: femtosecond, optical Kerr gate, tellurite glass, ultrafast response, nonlinear refractive index

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

1. Introduction

Femtosecond optical Kerr gate (OKG) technique has been widely used in the nonlinear optical measurements due to its precision, sensitivity, and convenient operation [1]. The requirements for OKG configuration of high quality are as follows: (1) ultrafast response, (2) low pump energy, (3) large transmittance window, and (4) high Kerr efficiency. Actually, selecting suitable nonlinear optical materials with nonlinear response of electronic process, large nonlinear refractive index, and large transmittance window as the Kerr media may satisfy the requirements.

Over the last few decades, the development of application of the OKG technique has been a hot research spot. The ordinary applications of OKGs are measurements of the optical nonlinearities and ultrafast responses of materials, e.g. liquids and gases [2, 3], organic or polymeric materials [4, 5], and nonlinear optical glasses [6–8]. In these measurements, the fused silica and CS_2 were used as the standard Kerr media. Typically, the OKG technique has been used for measuring the chirp structure of femtosecond supercontinuum pulses [9, 10], and imaging of shape measurements [11–13]. Since the OKG method was improved on time-resolved spectrum measurements, it has been developed a lot in the application of spectroscopy [14–16]. Especially, time-resolved luminescence spectra in the violet region by using the OKG method become a powerful tool for analyzing the ultrafast dynamics of carriers in semiconductors with large band gaps [17]. However, the Kerr media used in previous experiments were either of small nonlinear refractive index n_2 or of narrow transmittance spectral range. Tellurite glass (Te glass), with ultrafast response, large nonlinear refractive index, and wide range of transmittance spectra, will be very attractive in the application of the OKG technique in the future.

In this paper, the ultrafast optical Kerr effect of Te glass and bismuth glass (Bi glass) were investigated using femtosecond laser pulses. The Kerr-gated spectra obtained from a chirped supercontinuum (SC) using a Te glass OKG were of wider range of transmittance of spectra than those obtained using a Bi glass OKG. To demonstrate the perfor-

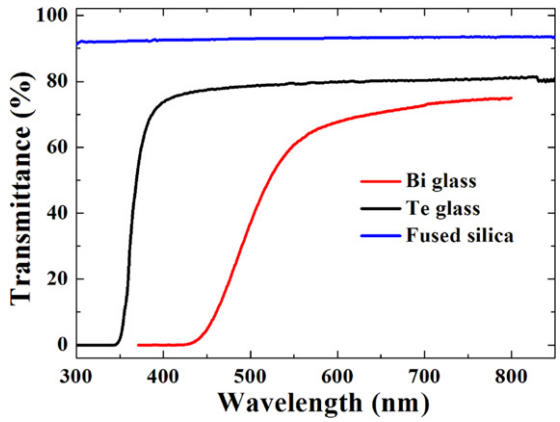

Figure 1. Transmitted spectra of Te glass (black), Bi glass (red), and fused silica (blue).

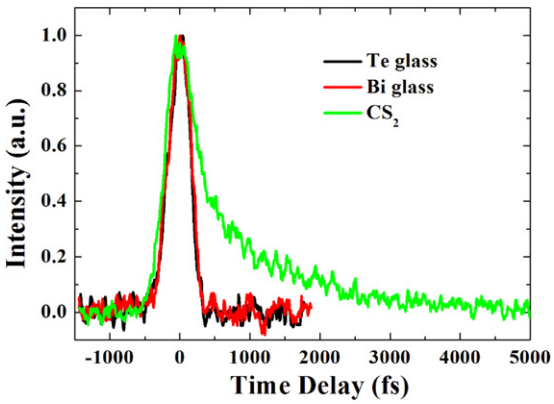

Figure 2. Femtosecond time-resolved optical Kerr signals of Te glass (black), Bi glass (red) and CS_2 (green).

mance of the Te glass OKG in the application of time-resolved fluorescence, the time-resolved EHP luminescence of a ZnO thin film was measured. Time-resolved electron–hole plasma (EHP) luminescence of a ZnO thin film obtained using a Te glass OKG was of higher temporal resolution than that obtained using a CS_2 OKG. The pump power used in the Te glass OKG was one order of magnitude lower than that for fused silica, because the nonlinear refractive index of Te glass was one order of magnitude larger than that of fused silica.

2. Glass samples

The Te glass sample provided excellent transparency from 0.38 up to 6.10 μm; therefore, no resonant absorption was observed at the laser wavelength centered at 800 nm. The Te glass was prepared by the conventional melt-quenching method [18]. Reagent chemical powders of $80TeO_2$–$10ZnO$–$10Na_2O$ with purity $\geq 99.9\%$ are precisely weighed up, homogeneously mixed in a glass bottle, melted in a gold crucible at about 800 °C for 1 h, poured into a brass mold at 220 °C, annealed at 259 °C for 8 h, and then slowly cooled down to room temperature. During the melting process, high melting temperature was chosen to reduce the viscosity of the melted glass; simultaneously, the powders were mixed uniformly by further shaking the crucible in the process of melting. The preparation of Bi glass has been described in detail elsewhere [19].

The linear transmitted spectra of Te glass, Bi glass and fused silica are shown in figure 1. We can see that the absorption edge is located at about 380 nm for Te glass and 500 nm for Bi glass, respectively. The transmittance loss for Bi glass above 550 nm was mainly due to the reflection and the scattering effect from bismuth particles dispersed in glass, and the absorption could be negligible [20]. No absorption is observed from 300 to 850 nm for fused silica.

3. Experiments and discussions

3.1. Ultrafast Te glass and Bi glass OKG

The instrument response function of the OKG system was tested by measuring the Kerr signal of a probe beam with the same wavelength as the pump beam. A multi-pass amplified Ti:sapphire laser system was employed, which emitted 30 fs, 800 nm laser pulses at a repetition rate of 1 kHz. Using the OKG setup [21], we measured the response time of the Te glass, Bi glass and CS_2 at the wavelength of 800 nm, respectively. The thicknesses of Te glass and Bi glass were both 1 mm, and the CS_2 was placed in a 1 mm thick fused silica cell.

The results are shown in figure 2, from which we can see that the nonlinear response times of the Te glass and Bi glass were nearly the same. The full widths at half maximum (FWHMs) of the pulse correlation for the Kerr signals of both the Te glass and Bi glass were about 300 fs. The pulse duration of the fundamental pulse was broadened due to the group velocity dispersion in the optical path. In our previous reports, an 85 fs width optical Kerr gate was obtained using Bi glass as the Kerr medium by further compressing the pulse width at the sample [21]. From the OKG measurement of EHP luminescence of ZnO film, the OKG with 300 fs width was narrow enough to acquire the time-resolved EHP luminescence of ZnO film. Hence, no further compression was performed in our experiments. It could be expected that the FWHM of the Kerr signals measured in Te and Bi glasses might be narrower if the fundamental pulses were compressed further. The OKG curves for Te and Bi glass showed symmetric profiles without slow decay components. The nonlinearities of the Te glass and Bi glass were mainly attributed to the electronic processes. The response times of the electronic processes in Te glass and Bi glass were several femtoseconds, which could be detected by even shorter laser pulses. The OKG decay curve of CS_2 showed two response components, which lasted about 330 fs and 1960 fs, respectively. The former was attributed to the electronic response, and the latter was attributed to the decay process of the molecular reorientation of CS_2.

3.2. Acquisition of Kerr-gated spectra from the chirped supercontinuum

The experimental setup of selecting gated spectra from a chirped SC using an OKG was shown in [10]. The multi-pass

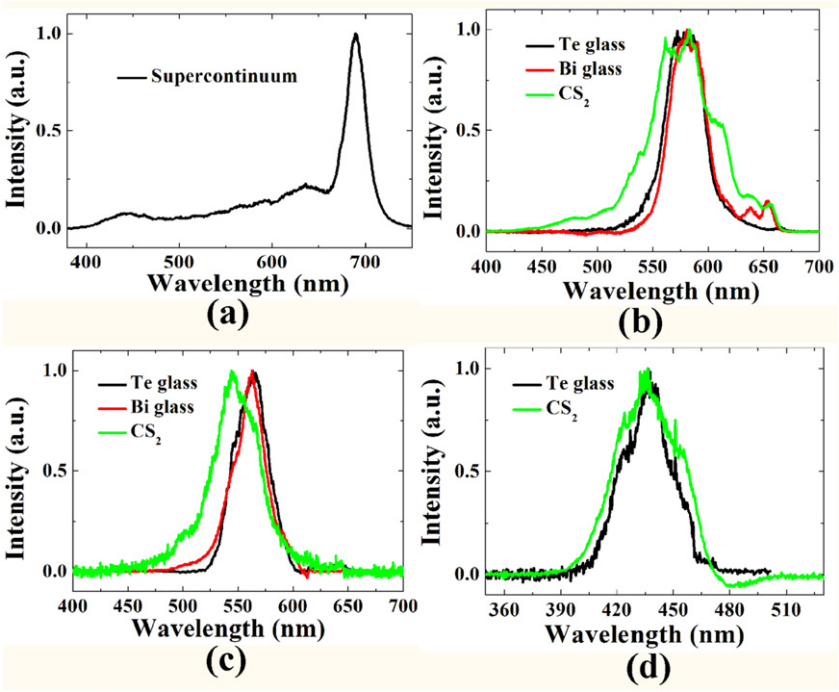

Figure 3. (a) Spectrum of SC generated in the sapphire plate and (b)–(d) Kerr-gated spectra from SC at different time delays. The neighboring intervals among (b)–(d) were about 160 fs and 1100 fs, respectively. The Kerr media were Te glass (black), Bi glass (red), and CS_2 (green).

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 was focused into a sapphire plate with 3 mm thickness by a 10 cm focal-length lens and generated the SC, which served as probe beam. The other beam, which passed through a time-delay device, served as the pump beam. A polarizer was placed behind the Kerr medium in a cross-Nicol configuration, so that the probe beam could not pass through the polarizer without the pump beam. A half-wave plate was introduced into the optical path of the pump beam. To optimize the intensity of the Kerr signals, the polarization plane of the pump beam was rotated by 45° from that of the probe beam. Stable SC probe pulses were produced by focusing a properly attenuated femtosecond laser beam and then collimated into the Kerr medium after filtering the long-wave region. Figure 3(a) shows that the spectrum of the SC ranges from 400 to 730 nm. The SC was up-chirped mainly by group velocity dispersion in the lenses, the air, and the sapphire itself.

Figures 3(b)–(d) show the Kerr-gated spectra obtained at different time delays between the pump beam and the supercontinuum. From figures 3(b) and (c), we can see that both of the gated spectra obtained by the Te glass and Bi glass OKGs have symmetrical distributions with FWHM of 30 nm, which are much narrower than those obtained by the CS_2 OKG. Besides, the band tailing is observed obviously in the gated spectra obtained by the CS_2 OKG due to its slow nonlinear response. The gated spectra at shorter spectral range are shown in figure 3(d). The obtained Kerr-gated spectrum using Te glass is still much narrower than that of CS_2 at 435 nm. However, no gated spectrum signal was detected at the wavelength shorter than 450 nm by using Bi glass as the Kerr medium.

The Kerr-gated spectra obtained by Te glass OKGs have narrower bandwidth and more symmetrical distribution than those of CS_2. By comparing the gated spectra obtained by Te glass and Bi glass, the Te glass OKG exhibited a much wider transmitted spectral range. The Te glass OKG, which showed the superiorities of ultrafast nonlinear response and the strong capability of capturing spectra in the violet, can be used to capture the transient fluorescence in the violet region.

3.3. Acquisition of time-resolved spectra of violet fluorescence with high temporal resolution and low pump power using a Te glass OKG

To demonstrate performance of the Te glass OKG in the application of time-resolved fluorescence in the violet region (∼400 nm), the time-resolved two-photon absorption-induced EHP luminescence of a ZnO thin film was measured. The ZnO film was fabricated by the radio frequency magnetron sputtering technique [22].

In this measurement, the 3 mm sapphire plate was replaced with a ZnO film with 2 μm thickness. A low-pass filter was placed behind the ZnO film to remove the fundamental beam. The ZnO film luminescence, which served as the probe beam, was polarized by a polarizer and then passed through the Kerr medium. When the probe beam and the pump beam met at the Kerr medium temporally and spatially, the Kerr signal of luminescence of the ZnO film was detected by a photomultiplier tube.

The luminescence of the ZnO film sample was excited at a fluence of 200 mJ cm^{-2}. This yields carrier density

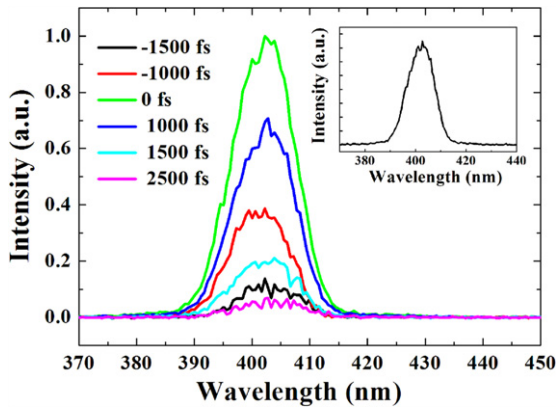

Figure 4. Time-resolved luminescence spectra of EHP emission of ZnO film for different delay times using Te glass OKG. The inset shows the time-integrated luminescence spectrum of the EHP emission.

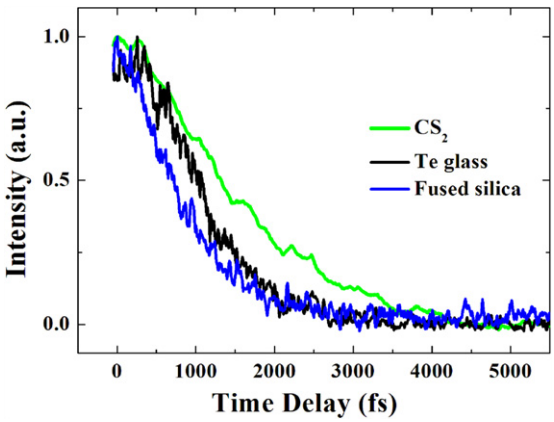

Figure 5. Decay time of EHP emission of ZnO film measured by OKGs of Te glass (black), CS_2 (green) and fused silica (blue), respectively.

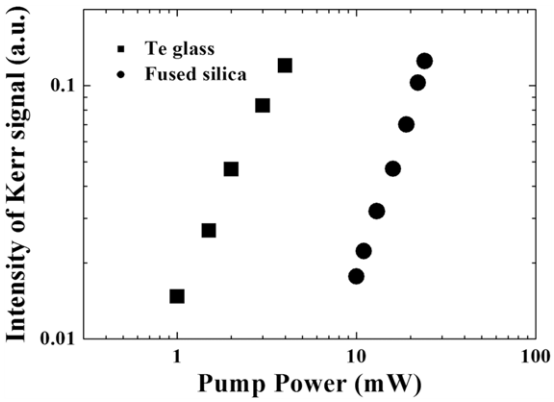

Figure 6. Intensity dependence of Kerr-gated EHP luminescence on pump power by using Te glass and fused silica as Kerr media, respectively.

higher than the Mott transition density. The excitonic state became unstable due to screening of the Coulomb attraction between electrons and holes, and the exciton was ionized. Therefore, luminescence of the high-density EHP state was observed [23]. The inset of figure 4 shows the time-integrated luminescence spectrum of the EHP emission of the ZnO film. The central wavelength of the EHP luminescence was located at 402 nm, which was longer than the wavelength of the intrinsic luminescence. The EHP luminescence band accompanied by a red-shift was thought to be bandgap renormalization [24], which showed a cooling process from a hot to a quasi-thermalized electron–hole system. The time-resolved luminescence spectra of the EHP emission using Te glass as the Kerr medium are shown in figure 4. The onset wavelength of each Kerr-gated spectrum was about 417 nm, which was in good agreement with that obtained from the time-integrated EHP emission spectrum (the inset of figure 4). This indicated that the observed time-resolved luminescence spectra of the EHP emission came from a single origin.

The decay time of the EHP emission of the ZnO film was measured using OKGs, where the Te glass, fused silica, and CS_2 were used as the Kerr media, respectively. The decay times of EHP emission measured by the Te glass OKG and fused silica were nearly the same, while the decay times measured by the CS_2 OKG were significantly different, as shown in figure 5. By using the Te glass and fused silica as the Kerr media, the decay times of the EHP luminescence of the ZnO film were both about 2.5 ps, which were in agreement with previously reported results [22]. A much longer decay time was observed by using CS_2 as the Kerr medium. The decay time was strongly broadened to about 4.5 ps, which misinterpreted the true characteristic of the EHP emission. The temporal broadening of the decay time of the EHP emission using CS_2 as the Kerr medium was due to the slow decay of molecular reorientation in CS_2. The Te glass OKG showed the application of high temporal resolution in acquiring time-resolved luminescence of EHP emission in the violet region.

Figure 6 shows the intensity dependence of Kerr signals of the EHP emission on the pump power by using Te glass and fused silica as Kerr media, respectively. In this measurement, the thicknesses of Te glass and fused silica were both 1 mm, and the time delay was kept at the peak of the Kerr signal of the EHP luminescence. The central wavelength of the pump beam was centered at 800 nm. We can see that the intensity of the Kerr signal increased with the pump power in both Te glass and fused silica. The EHP luminescence transmittance (I_k) of the OKG measurement is given by [15]

$$I_k = I_0 \sin^2(\Delta\phi/2) \approx I_0(\Delta\phi/2)^2 \propto n_2^2 I_{\text{Pump}}^2,$$

where n_2 is the nonlinear refractive index of the Kerr medium and I_{Pump} is the intensity of pump beam. The intensity of the Kerr signal is proportional to the square of I_{Pump}. The slope of the Kerr signal intensity measured by the Te glass OKG was close to 2 at low pump power (below 2 mW), while it was smaller than 2 at high pump power. The decrease of slope was due to the pump power loss at high pump power, which could be attributed to the multi-photon absorption. To acquire the Kerr signal with the same intensity, the pump power used in the Te glass OKG was about one order of magnitude smaller than that of fused glass. The significant decrease of pump power by using Te glass as the Kerr medium was attributed to its large nonlinear refractive index,

$n_2 \sim 10^{-15}$ cm^2 W^{-1} [25–27], which is one order of magnitude larger than that of fused silica [28]. In the Te glass, the high optical nonlinearity may be attributed to the empty 5d orbitals [29] or the electron lone pair at the Te atom [30].

The OKG method has been widely used in many optical fields. The good quality of the OKG was mainly dependent on a Kerr medium with ultrafast response, large nonlinear optical refractive index, and wide transmission spectra in the visible and infrared region. In the above experiments, Te glass showed superiorities which met the requirements of the OKG well. Compared with the Bi glass discussed in this context, and other recently reported glasses, such as tungstate oxide based glass [31], chalcogenide glass [32], and sodium borosilicate glass [33], the nonlinear response times were of the same magnitude due to the electronic response processes in these glasses. The nonlinear susceptibility $\chi^{(3)}$ of Te glass was of the same magnitude as that of tungstate oxide based glass, and smaller than that of chalcogenide glass and sodium borosilicate glass. However, the Te glass showed a much larger transmittance window from 0.38 up to 6.1 μm, which promised the Te glass to be a good candidate for photonic applications in future.

4. Conclusion

In summary, we investigated the ultrafast optical Kerr effect of Te glass and Bi glass, respectively. The Kerr-gated spectra from SC obtained using a Te glass OKG showed narrower bandwidth and a large transmittance window. The decay time with high temporal resolution of the EHP luminescence of the ZnO film was measured using the Te glass OKG. The pump power used in the Te glass OKG was about one order of magnitude lower than that used in the fused silica OKG due to its larger nonlinear refractive index.

Acknowledgments

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