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Femtosecond nonlinear optical property of a TeO₂–ZnO–Na₂O glass and its application in time-resolved three-dimensional imaging

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ABSTRACT

Compared with bismuth glasses and chalcogenide glasses, tellurite glasses have a wider transparency window. Especially, these glasses with excellent transparency in visible region could offer a larger dynamic range for the time-resolved simultaneous three-dimensional imaging based on the optical Kerr gate (OKG). We investigated the ultrafast nonlinear optical properties of a TeO₂–ZnO–Na₂O glass using a femtosecond optical Kerr gate at wavelength of 800 nm. The nonlinear refraction n_2 and the ultrafast nonlinear response time of the tellurite glass were estimated to be $4.56 \times 10^{-15} \text{ cm}^2/\text{W}$ and less than 200 fs, respectively. We also demonstrated the time-resolved simultaneous three-dimensional imaging using the tellurite glass as the Kerr medium, which indicated that tellurite glasses may also be good candidates as the Kerr media for the time-resolved imaging based on the OKG.

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1. Introduction

Large third-order optical nonlinearity of nonlinear optical materials is the most important property for the photonic applications [1–4]. Although some materials have large nonlinearity, such as organic liquids and semiconductors with quantum well structures [5,6], these materials also have strong absorption, limiting their application. Recently, it has emerged that heavy-metal oxide glasses seem to be preferable candidates to photonic applications due to their large optical nonlinearities, ultrafast response time, and wide transparency window [7–11]. For instance, it has been reported that bismuth glasses have been used as the Kerr media in a time-resolved simultaneous three-dimensional (3-D) imaging based on the optical Kerr gate (OKG) due to their large optical nonlinearity and ultrafast response time, which could offer higher signal-to-noise ratio and better temporal resolution for such application [8,12]. However, bismuth glasses have a limited transmission bandwidth in the visible region, the absorption edges of which are about 500 nm [13]. It somewhat restricts the dynamic range of the time-resolved simultaneous 3-D imaging using bismuth glasses as the OKG media [12].

Compared with bismuth glasses, apart from the large optical nonlinearity and the ultrafast response time, tellurite glasses have a wider transparency window [14–16]. Therefore, these glasses

could offer a larger dynamic range for the time-resolved simultaneous 3-D imaging based on the OKG. In addition, tellurite glasses have larger refraction index [14], very low phonon energy [17], and good thermal and mechanical stability [15]. These excellent optical properties of tellurite glasses have been extensively exploited in the fields of nonlinear optical processing and fiber based optical amplification [18–21].

In this paper, we investigated the ultrafast nonlinear optical properties of a TeO₂–ZnO–Na₂O (hereafter denoted by Te) glass using a femtosecond OKG measurement at wavelength of 800 nm. The nonlinear refraction n_2 of the Te glass was estimated to be $4.56 \times 10^{-15} \text{ cm}^2/\text{W}$. The ultrafast nonlinear response time was measured to be less than 200 fs. For its application, we demonstrated the time-resolved simultaneous 3-D imaging using the Te glass as the Kerr medium, which indicated that tellurite glasses may be good candidates as the Kerr media for the time-resolved imaging based on the OKG [12,22,23].

2. Experiments and materials

The Te glass sample with composition of 80TeO₂–10ZnO–10Na₂O was made by the traditional melt-quenching method. Reagent chemical powders with purity $\geq 99.9\%$ were precisely weighed up, homogeneously mixed in a glass bottle, melted in a gold crucible at about 800 °C for 1 h, poured onto a brass mold at 220 °C, annealed at 259 °C for 8 h, and then slowly cooled down to room temperature. The Te glass samples were cut into specimens

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with the size of $10 \times 10 \times 1 \text{ mm}^3$ and were polished to optical quality. The linear refractive index of the sample was measured to be 2.00873 at the wavelength of 800 nm.

The linear transmission spectrum of the Te glass was measured as shown in Fig. 1. The enlarged linear transmission spectrum from 300 nm to 500 nm was shown in the inset of Fig. 1. From Fig. 1, we can see that the absorption edge is below 400 nm, above which there is no clear absorption. The transmittance curve is quite flat at the range of 800–2000 nm, which indicates that the Te glass has good optical quality and effect of OH^{-1} on optical property is well eliminated [15]. Therefore, the Te glass presents a wide transparency window of 400–2000 nm. It suggests that the Te glass has a large applicable laser wavelength range from the visible to the near-infrared wavelength region. Especially, the Te glass with excellent transparency in the visible region could offer a large dynamic range for the time-resolved simultaneous 3-D imaging based on the OKG [12].

Measurements of the third-order optical nonlinearities and the ultrafast response of the Te glass were performed by use of the OKG measurement [13,24]. A Ti:sapphire laser system with a repetition rate of 1 kHz, pulse duration of 30 fs and an average power of 1 W at 800 nm was used in our experiments. The angle between the two beams was about 12° . The pump beam was time delayed using a computer-controlled optical delay-line, enabling a temporal resolution of about 5.2 fs. The OKG signals were detected by a photomultiplier tube (PMT) and recorded by a computer. Using the OKG measurement setup, the femtosecond time-resolved OKG signal of the Te glass, the reference sample CS_2 was measured as shown in Fig. 2. The CS_2 solution was filled in a

quartz cuvette with a path length of 1 mm. The solid circle and the hollow square refer to the Te glass and CS_2 , respectively. The signal intensities for these media are normalized by their peak intensities.

3. Results and discussions

From Fig. 2, we can see that the time-resolved OKE signal of the Te glass was symmetrical and the full width at half-maximum (FWHM) of the signal was about 200 fs, which was another ultrafast response observed in nonlinear optical glasses [24]. In our experiments, the duration of the laser pulses was expanded because of the dispersion of the optical elements. The symmetric correlation signal width of 200 fs, showing no slow component, implied that the response curve was determined by pulse width and the recovery time of the Te glass is faster than the pulse width. This indicates that the dominant mechanism of nonlinearity was attributed to electronic polarization. On the other hand, the curve of CS_2 has an asymmetrical decay tail approximately 1.6 ps, attributed to the molecular orientation relaxation, which was in accordance with previous reports and indicated the reliability of our experiment [24,25].

When we measured the OKE signals induced in the sample, an analyzer perpendicular to the polarization plane of the probe beam was positioned before the detector. The signal intensity depends on the induced phase shift $\Delta\varphi$ and the angle θ between the polarization planes of the pump and probe beams according to [26]

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

The OKE signal shows a maximum when the polarization angle θ between the pump and probe beams is fixed at 45° . Using the effective length of the sample L_{eff} and the wavelength of the probe beam λ_p , the phase shift $\Delta\varphi$ is given by [26]

$$\Delta\varphi = \frac{2n_2 L_{\text{eff}} P_p}{r^2 \lambda_p} \quad (2)$$

where P_p and r represent the pump power and the beam radius at the focal point, respectively. Kerr nonlinear refractive index n_2 corresponds to the third-order nonlinear coefficient. If the phase shift $\Delta\varphi$ is small enough, substituting Eq. (2) into Eq. (1) we could get

$$I/I_0 = \left(\frac{2n_2 L_{\text{eff}}}{\lambda_p r^2}\right)^2 P_p^2 \quad (3)$$

therefore, following the standard reference measurement, the nonlinear refractive index n_2 of the sample can be evaluated by the equation

$$n_{2S} = n_{2R} \left(\frac{(I/I_0)_S}{(I/I_0)_R}\right)^{\frac{1}{2}} \quad (4)$$

where the subscripts S and R indicate the sample and the reference sample of fused quartz, respectively. According to Eq. (4) nonlinear refractive-index n_2 of the Te glass was estimated to be $4.56 \times 10^{-15} \text{ cm}^2/\text{W}$, where $n_{2R} = 2.48 \times 10^{-16} \text{ cm}^2/\text{W}$ at about 800 nm. The result agreed well with that in Ref. [17].

Furthermore, we demonstrated the application of the Te glass in the time-resolved simultaneous 3-D imaging which was proposed by Minoshima et al. [12]. The similar setup and imaging target were used in our experiment. In order to get higher signal-to-noise ratio and better time resolution, a 1-mm Te glass sample was chosen as the Kerr medium. In the measurement, a chirped supercontinuum was used as the imaging beam, which was collimated and expanded to illuminate a transparent object with

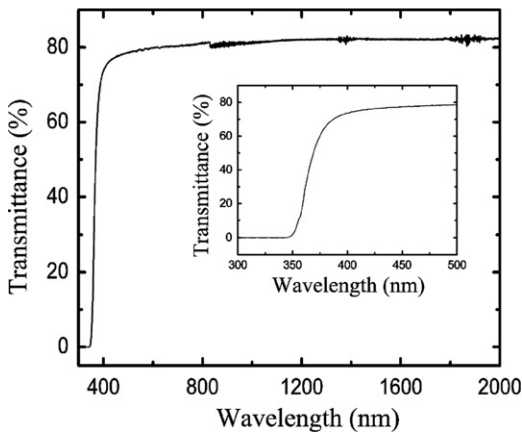


Fig. 1. Transmission spectra of the Te glass. Inset: the enlarged transmission spectra from 300 nm to 500 nm.

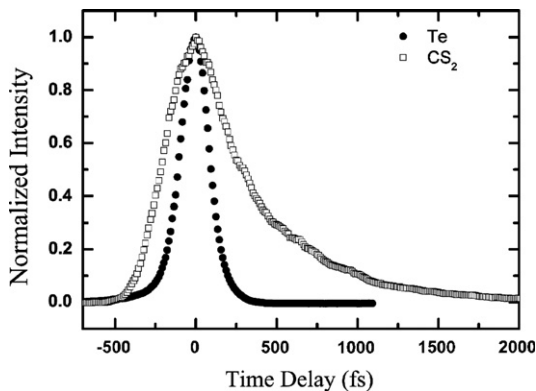


Fig. 2. Femtosecond time-resolved OKE signals of the samples. Solid square: Te glass; Hollow square: CS_2 .

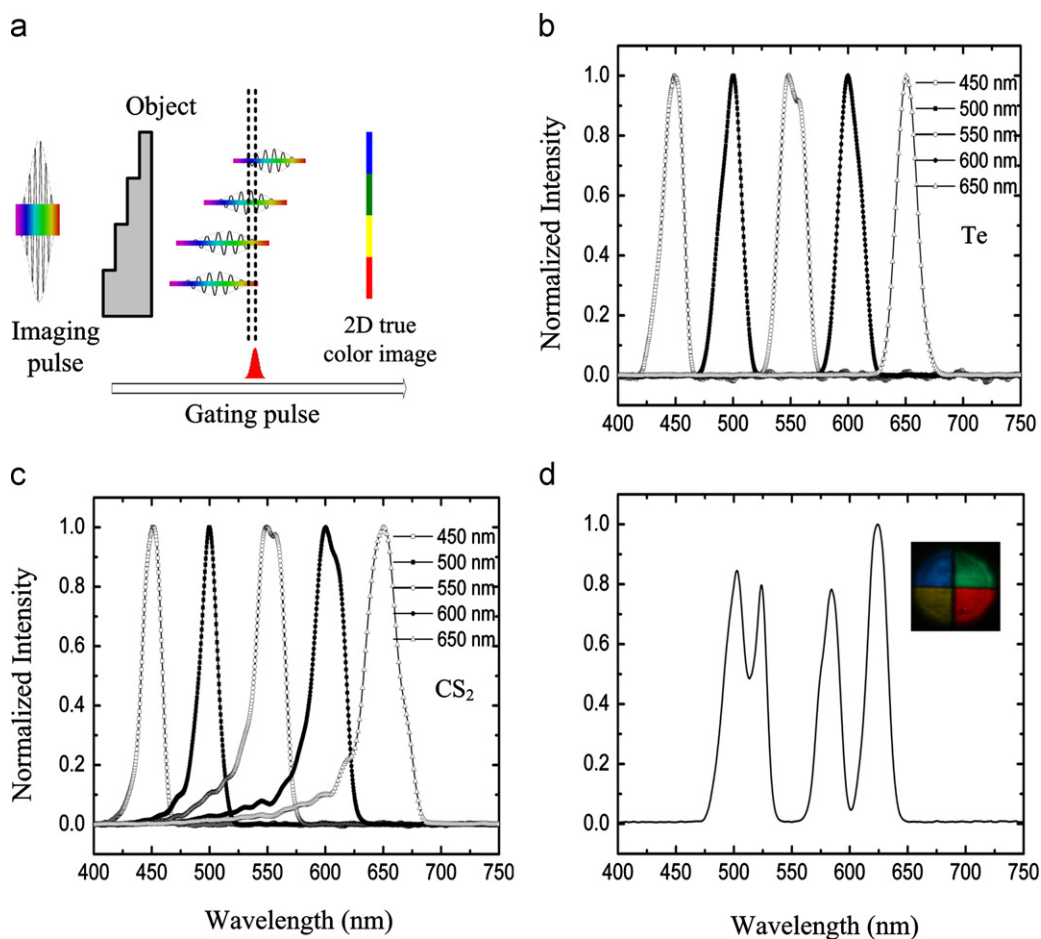


Fig. 3. (a) Schematic of time-resolved simultaneous 3-D imaging using OKG; (b) the Kerr-gated spectra from the SC at five selected wavelengths obtained using the Te glass; (c) the Kerr-gated spectra from the SC at five selected wavelengths obtained using CS₂; (d) The Kerr-gated spectrum and corresponding Kerr-gated true 2-D color image of the object at a certain time delay using the Te glass as the Kerr medium. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

four flat steps as schematically shown in Fig. 3(a). The spectrum of the supercontinuum ranges from 400 nm to 750 nm. Passing through the object, the imaging beam had different optical paths for different regions of the object. By adjusting the time delay between the gating pulse and the supercontinuum, the OKG could transmit a time-sliced supercontinuum with four wavelengths, which was detected by a color CCD. The spectral information was also monitored by a microspectrometer synchronously. To block light at the fundamental 800 nm wavelength, two cut-off filters were placed before the CCD and the microspectrometer, respectively. The transmitted beam showed a color distribution according to the surface structure, so we could obtain a real-time colored 3-D map of the object. From Fig. 3(a), we can see that the longitudinal dynamic range of this technique is mainly limited by the pulse duration of the supercontinuum. Several tens of centimeters are capable to be measured by extending the pulse duration of the supercontinuum to nanosecond region. Fig. 3(a) also indicates that acquiring the gated spectra of narrower bandwidth and better symmetry by use of a fast OKG could offer better temporal and longitudinal spatial resolution in this technique. Theoretical calculation shows the gating pulse of about 200 fs is used in our experiment, the resolvable optical path of 36 μm can be obtained. The longitudinal resolution is dependent on the optical path difference corresponding to the different regions on the object surface. For the reflection-type imaging, the optical path difference is twice the step height on the object surface, while for the transmission-type imaging, the optical path

difference is equal to the product of the step height and the refractive index difference between air and the object.

We firstly studied the characteristics of Kerr-gated spectra acquired from the chirp supercontinuum using the Te glass and reference Kerr medium CS₂, respectively, in which the object was removed. A series of Kerr-gated spectra at five different wavelengths are obtained as shown in Fig. 3(b) and (c). The peak wavelengths of the Kerr-gated spectra are 450 nm, 500 nm, 550 nm, 600 nm, and 650 nm, which were detected at different delay times of the pump laser. The bandwidths of Kerr-gated spectra obtained using the BI glass are narrower 25 nm in FWHM and kept nearly invariant. While the gated spectra bandwidths for CS₂ increase from about 20 nm to 31 nm in FWHM with the increase of the wavelength due to the chirp characteristics of the supercontinuum. Moreover, the Kerr-gated spectra of CS₂ exhibit obvious band tailing. The results suggest that the time-resolved 3-D imaging using the Te glass could offer better temporal and longitudinal spatial resolution compared with CS₂.

Furthermore, we measured the Kerr-gated spectrum and corresponding Kerr-gated true 2-D color image of an area of four flat steps at a certain time delay using the Te glass as the Kerr medium. The steps are composed of four pieces of glass, the thicknesses of which were 165 μm , 330 μm , 495 μm , and 660 μm , respectively. From Fig. 3(d), we can see that there are four distinguishable peaks in the Kerr-gated spectrum and the Kerr-gated image clearly shows four color regions, i.e., blue, green, yellow, and red region, presenting the difference of the step

heights of the object. These results indicate that tellurite glasses may also be good candidates as the Kerr media for the time-resolved imaging based on the OKG [12,22,23].

4. Conclusion

In summary, we investigated the ultrafast nonlinear optical properties of the Te glass using the femtosecond OKG measurement at wavelength of 800 nm. The nonlinear refraction n_2 of the Te glass was estimated to be $4.56 \times 10^{-15} \text{ cm}^2/\text{W}$. The ultrafast nonlinear response time of the Te glass was measured to be less than 200 fs. In addition, we demonstrated the time-resolved simultaneous 3-D imaging using the Te glass as the Kerr medium, which indicated that tellurite glasses may be good candidates as the Kerr media for the time-resolved imaging based on the OKG.

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