Quantitative analysis of Kerr nonlinearity and Kerr-like nonlinearity induced via terahertz generation in ZnTe

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Kerr nonlinearity and Kerr-like nonlinearity induced via terahertz generation and the electro-optical effect in ZnTe crystal are investigated. In general, these nonlinear effects are concomitant and difficult to quantitatively analyze in the time domain. Even *Z*-scan technique, which is a simple and sensitive single-beam method to determine both the sign and magnitude of the nonlinear refractive index as well as the nonlinear absorption coefficient of a given material, cannot quantitatively analyze the contribution of each nonlinear effect directly. A method is proposed in the spectral domain to distinguish between Kerr nonlinearity and Kerr-like nonlinearity. Experimental results agree with theoretical analysis. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2838446]

In the past 20 years, there has been considerable interest in generating and detecting terahertz radiation by femtosecond pulse lasers.[1,2] Terahertz time-domain spectroscopy[3] (TDS) is the most common setup using electro-optic (EO) crystals[4] or photoconductive antennas[5] as terahertz emitter and detector. More recently, among the EO crystals, ZnTe has been regarded as the primary material for terahertz generation based on optical rectification excited by femtosecond laser pulses at ~800 nm, and thus has been extensively investigated from both the fundamental and the applied points of view.[4,6–10]

When generating and detecting terahertz, pulse is performed synchronously using one piece of ZnTe crystal, both third-order nonlinear (TON) susceptibility called Kerr nonlinearity and cascaded second-order nonlinear (SON) effects called Kerr-like nonlinearity occur in the crystal. Auston and Nuss[11] have mentioned higher order nonlinearity (Kerr effect) in terahertz generation besides second-order nonlinearity. Our group has carried out an investigation of Kerr effect and two-photon absorption in the ZnTe crystal using *Z*-scan technique with femtosecond laser pulses[9] and discussed the phase control of few-cycle coherent terahertz radiation pulses using Kerr effect.[10] However, for a long time, Kerr-like nonlinearity in the ZnTe has been overlooked. Recently, researchers have been paying increasing attention to this phenomenon. Caumes *et al.*[8] reported Kerr-like nonlinearity resulting from the propagation of terahertz radiation in ZnTe, and demonstrated its competition with a third-order optical Kerr effect. Based on their experiment, He *et al.*[12] pointed that in a (110) oriented ZnTe crystal, the nonlinear refractive coefficient n_2 is dominated by the Kerr-like nonlinearity, while n_2 in a (111) oriented ZnTe crystal mainly comes from the combined contribution of the Kerr-like nonlinearity and Kerr nonlinearity. However, the views of Caumes *et al.* and those of He *et al.* are contradictory, for Caumes *et al.* thought that Kerr nonlinearity and Kerr-like nonlinearity are comparable and He *et al.* thought that Kerr-like nonlinearity dominates in a (110) oriented ZnTe crystal. And so far no other theoretical and experimental data are available to discuss this issue. It is necessary to clarify this phenomenon, because otherwise Kerr signal may be regarded to be terahertz signal. Furthermore, Kerr-like nonlinearity also gives a potential way for all-optical switches and related applications.[13] It is, however, difficult to quantitatively evaluate the contribution of each nonlinear effect mentioned above to n_2 alone due to their simultaneity. In this paper, we have proposed a spectral model to quantitatively distinguish between Kerr nonlinearity and Kerr-like nonlinearity induced via terahertz generation in the ZnTe. We show that in a (110) oriented ZnTe crystal, both Kerr nonlinearity and Kerr-like nonlinearity play important roles and the nonlinear refractive coefficient n_2 has been measured by use of spectrally resolved two-beam coupling.[14]

According to Caumes *et al.*,[8] the nonlinear refractive coefficient n_2 of ZnTe crystals can be written as

$$n_2 = n_2^{\mathrm{Kerr}} + n_2^{\mathrm{SHG}} + n_2^{\mathrm{OR}} + n_2^{\mathrm{terahertz}}, \tag{1}$$

where n_2^{Kerr} is associated with TON, n_2^{SHG} is discovered by DeSalvo *et al.*[15] and n_2^{OR} is discovered by Bosshard *et al.*[16] and Biaggio.[17] Caumes *et al.*[8] have clearly shown that n_2^{Kerr} always overcomes the contribution of n_2^{SHG} and n_2^{OR} in the ZnTe crystal and the contribution from the terms n_2^{SHG} and n_2^{OR} can be neglected. Thus only $n_2^{\mathrm{terahertz}}$ competes with n_2^{Kerr}.

Considering that only either second- or third-order polarization is responsible for the temporal evolution of the small birefringence induced phase change due to either Kerr nonlinearity or Kerr-like nonlinearity, the linearly polarized probe beam becomes slightly elliptical as a result of the phase change after its propagation in the crystal. This elliptical polarization is recorded by the detection unit in the experiment and gives rise to the signal

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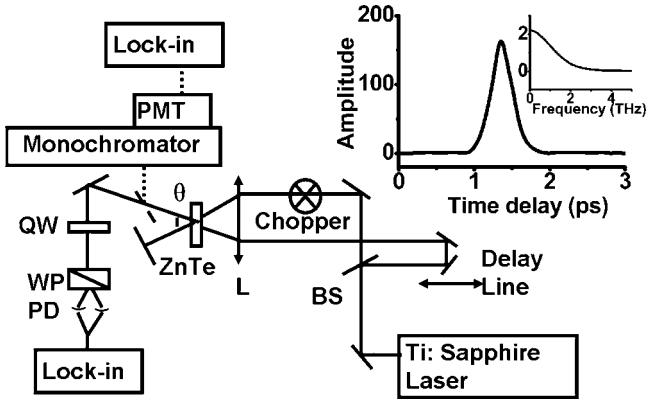

FIG. 1. Right: femtosecond Kerr-lens autocorrelation: time-domain trace and corresponding amplitude spectra, Left: the pump-probe experimental setup.

$$E(\tau) = E^{\text{Kerr}}(\tau) + E^{\text{terahertz}}(\tau), \tag{2}$$

where $E^{\text{Kerr}}(\tau)$ and $E^{\text{terahertz}}(\tau)$ are connected with the SON and TON polarizations, respectively. When we perform Fourier transform to Eq. (2), the spectrum obtained is the superposition of the terahertz spectrum and the autocorrelation trace of the pump beam based on the Kerr effect. If $E^{\text{terahertz}}(\tau)$ is negligible, the signal $E(\tau)$ will be the autocorrelation trace of the pump pulses[18] based on the Kerr effect, as shown in the top right corner of Fig. 1, and the inset is its corresponding spectra, whose value at zero frequency is in proportion to the peak of the autocorrelation signal in the time domain at a time delay $\tau=0$. We can regard the spectrum at zero frequency of the signal obtained in the experiments as being dependent on the Kerr nonlinearity only.

To evidence this proposal, we built a modified pump-probe setup (showed in the left of Fig. 1). The part of the setup plotted in solid line measures the Kerr signal and terahertz signal based on Pockels cell effect and the part plotted in dotted line measures nonlinear refractive coefficient n_2 based on spectrally resolved Kerr gate effect. The setup is a combination of balanced detection and spectrally resolved two-beam coupling. The parameter of the femtosecond laser is the same with Ref. 7. In the experiments, by rotating the ZnTe crystal around the (110) orientation, equivalent to changing the laser polarization relative to the crystallographic axis, we recorded the signal in time domain for every 10° rotated within the range of $\gamma(\gamma=0\sim360°)$. Figure 2(a) shows the recorded signal from $10°\sim90°$ using the balanced detection part and Fig. 2(b) shows the signal of the spectrally resolved two-beam coupling at 90°.

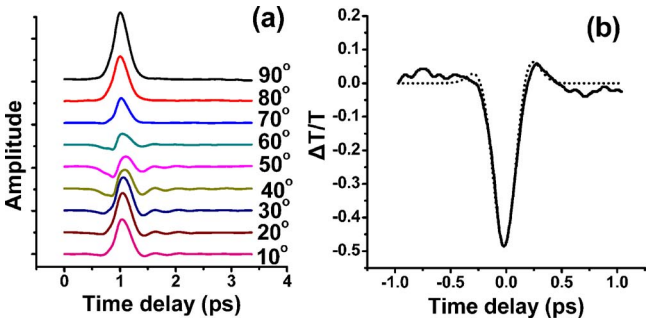

FIG. 2. (Color online) (a) The recorded signal using balanced detection part from 10°–90°, (b) the recorded signal (solid line) using spectrally resolved two-beam coupling at 90°, and the dotted line is theoretical fit data by setting the nonlinear refractive coefficient $n_2=7\times10^{-18}$ m^2/W and the nonlinear absorption coefficient $\beta=38$ cm/GW.

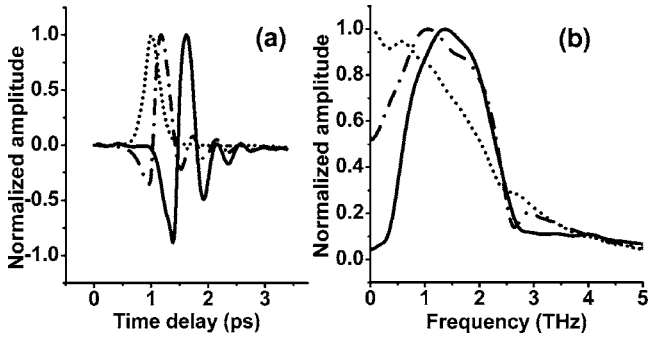

FIG. 3. Three typical signals measured in the experiments (a) in the time domain and (b) their corresponding spectra. Autocorrelation signal of femtosecond Kerr-lens only (dotted line) and terahertz signal only (solid line). Competition between femtosecond Kerr-lens autocorrelation signal and terahertz signal (dash dotted line).

Figure 3(a) shows three typical signals measured in the experiments in the time domain and their corresponding spectra are plotted in Fig. 3(b). The signal plotted in dotted line in Fig. 3(a) is measured at $\gamma\sim0°$ that is almost the same as the autocorrelation trace of the femtosecond laser pulses, as shown in Fig. 1. It hardly contains any terahertz component because there is almost no terahertz radiation excited at $\gamma\sim0°$. The signal plotted in solid line in Fig. 3(a) tested at $\gamma\sim0°$ is a typical terahertz signal with tiny Kerr nonlinearity contribution. The signal plotted in dash dotted line in Fig. 3(a) shows the signal for $\gamma\sim50°$ with considerable competition between both terahertz radiation and Kerr nonlinearity. The Kerr nonlinearity is plotted in Fig. 4(a) as the squares for each data point at zero frequency recorded when every 10° is rotated. Following Owyoung,[19] we have the nonlinear refractive coefficient concerning TON given by

$$n_2 = C_1 + C_2\left[1 - \frac{1}{2}(4\sin^2\gamma - 3\sin^4\gamma)\right], \tag{3}$$

$$C_1 = \frac{12\pi}{n_0}(2\chi_{1122} + \chi_{1221}), \tag{4}$$

$$C_2 = \frac{12\pi}{n_0}(\chi_{1111} - 2\chi_{1122} - \chi_{1221}) \tag{5}$$

where χ_{1122}, χ_{1221}, and χ_{1111} are independent TON tensor coefficients. The fit to Eq. (3) is shown as the solid line in Fig. 4(a). The nonlinear refractive coefficient n_2 is also given by spectrally resolved two-beam coupling. The fit to the signal of the spectrally resolved two-beam coupling at 90° by setting the nonlinear refractive coefficient $n_2=7$

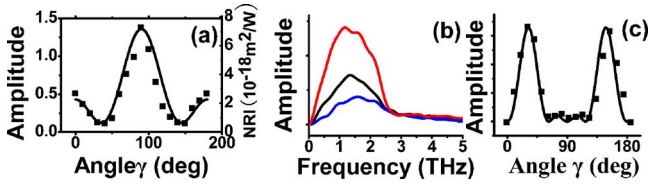

FIG. 4. (Color online) (a) Femtosecond Kerr-lens autocorrelation signal at zero frequency by rotating the ZnTe crystal every 10° along the (110) direction and nonlinear refractive index coefficient at every degree. (b) The pure terahertz spectra minus Kerr-lens autocorrelation amplitude spectra. (c) Terahertz signal intensity as a function of angle.

$\times 10^{-18}$ m^2/W and the nonlinear absorption coefficient $\beta = 38$ cm/GW is shown as the dotted line in Fig. 2(b). Since the value of n_2^{Kerr} is in proportion to the peak of the autocorrelation signal, we can get the value of n_2^{Kerr} by measuring the value at zero frequency in spectral domain. Furthermore, Fig. 4(a) shows an experimental n_2^{Kerr} as a function of γ.

By removing the fitted n_2^{Kerr}-related spectra according to the relative Kerr-lens autocorrelation amplitude spectrum from the original spectra measured in the experiments, the pure terahertz spectra are separated. Figure 4(b) shows the separated terahertz spectra. Then, we perform inverse Fourier transform to the terahertz spectra obtained, and terahertz time domain intensity signal is obtained. The signal intensity is plotted in Fig. 4(c) as the squares as a function of angle γ. Following Chen *et al.*,[20] terahertz signal can be determined by

$$I_{\mathrm{signal}} \propto \sin^2[2(\gamma - \varphi)] \times \sin^2 \gamma \times (1 + 3\cos^2 \gamma) \times (1 + 3\sin^2 \phi), \quad (6)$$

where $\tan \phi = 2 \cot \gamma$, $2 \tan \phi = -\tan 2\varphi$. The fit to Eq. (6) is shown as the solid line in Fig. 4(c). Following the results of He *et al.*,[12] we know that the nonlinear refractive coefficient n_2 thus measured in a (110) oriented ZnTe crystal is a sum of n_2^{THz} and n_2^{Kerr} and they are comparable.

In conclusion, we have proposed a spectral domain method to distinguish between various nonlinear contributions during terahertz generation in ZnTe. Based on a modified pump-probe setup, we measured a series of time domain signals consisted of terahertz and Kerr and their corresponding spectra. According to the spectra at zero frequency of the signal, we got the Kerr signal as a function of laser polarization angle. Then, utilizing spectrally resolved two-beam coupling, we measured the quantificational Kerr nonlinear refractive coefficient. At last, by removing Kerr spectra from the original spectra and performing inverse Fourier transform to the terahertz spectra obtained, we obtained the pure terahertz time domain intensity signal. So the competition of Kerr nonlinearity and Kerr-like nonlinearity was observed. The experimental results agree well with the theoretical analysis. Our experiments demonstrated that our method is practical and suitable for other EO materials used in terahertz time-domain spectroscopy.

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