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

Zhen Tian, Changlei Wang, Qirong Xing, Jianqiang Gu, Yanfeng Li, Mingxia He, Lu Chai, and Qingyue Wang
Center for Terahertz Wave, Ultrafast Laser Laboratory, College of Precision Instrument and Optoelectronics Engineering, and Key Laboratory of Opto-electronics Information and Technical Science (Ministry of Education), Tianjin University, Tianjin 300072, People’s Republic of China

Weili Zhang
School of Electrical and Computer Engineering, Oklahoma State University, Stillwater, Oklahoma 74078, USA

(Received 9 December 2007; accepted 8 January 2008; published online 29 January 2008)

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 (TDS) is the most common setup using electro-optic (EO) crystals4 or photoconductive antennas5 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.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 Nuss11 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 pulses12 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

\[
n_2 = n^{\text{Kerr}}_2 + n^{\text{SHG}}_2 + n^{\text{OR}}_2 + n^{\text{terahertz}}_2 ,
\]

where \( n^{\text{Kerr}}_2 \) is associated with TON, \( n^{\text{SHG}}_2 \) is discovered by DeSalvo et al.15 and \( n^{\text{OR}}_2 \) is discovered by Bosshard et al.16 and Biaggio.17 Caumes et al.8 have clearly shown that \( n^{\text{Kerr}}_2 \) always overcomes the contribution of \( n^{\text{SHG}}_2 \) and \( n^{\text{OR}}_2 \) in the ZnTe crystal and the contribution from the terms \( n^{\text{SHG}}_2 \) and \( n^{\text{OR}}_2 \) can be neglected. Thus only \( n^{\text{terahertz}}_2 \) competes with \( n^{\text{Kerr}}_2 \).

Considering that only either second- or third-order polarizability 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...
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 super-position 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 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 (shown 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 $\gamma=0^\circ - 360^\circ$. Figure 2(a) shows the recorded signal from $10^\circ$ to $90^\circ$ using the balanced detection part and Fig. 2(b) shows the signal of the spectrally resolved two-beam coupling at 90°.

Figure 3 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 $\gamma=0^\circ$ 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 $\gamma=0^\circ$. The signal plotted in solid line in Fig. 3(a) tested at $\gamma=0^\circ$ 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 $\gamma=50^\circ$ 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, we have the nonlinear refractive coefficient concerning TON given by

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

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

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

where $\chi_{1122}$, $\chi_{1221}$, and $\chi_{1111}$ 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 $n_2=7$...
Following Chen et al. the value at zero frequency in spectral domain. Furthermore, Fig. 4 shows the experimental $n_{2}^\text{Kerr}$ as a function of $\gamma$. By removing the fitted $n_{2}^\text{Kerr}$-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 from the original spectra measured in the experiments, the relative Kerr-lens autocorrelation amplitude spectrum is obtained. The signal intensity is plotted in Fig. 4 as the dot-dashed line in Fig. 2(b). Since the value of $n_{2}^\text{Kerr}$ is in proportion to the peak of the autocorrelation signal, we can get the value of $n_{2}^\text{Kerr}$ by measuring the value at zero frequency in spectral domain. Furthermore, Fig. 4(a) shows an experimental $n_{2}^\text{Kerr}$ as a function of $\gamma$.

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 quantifi cational 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.

This work was partly supported by National Key Basic Research Special Foundation of China through Grant No. 2007CB310408, National Natural Science Foundation of China (60578037), National Science Foundation of Tianjin (07ZCGHH01100), NSFC-RFBR program 2007-2008 (60711210198), and “985” program in Tianjin University.