The THz Kerr Effect in Liquids and Ferroelectric Crystals

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Abstract: We have observed THz-pulse-induced optical birefringence in liquids and ferroelectric crystals. The polarization modulation is dependent on the square of the applied THz pump field which may exceed 100 *kV/cm* levels. As in many all-optical Kerr effect measurements, there is a fast electronic response which may be followed by a slower reorientational response. The electronic response may be used to map out the intensity profile of the THz pulse. THz Kerr effect measurements may enable study of dipole dynamics in relaxor ferroelectric crystals.

Keywords: Terahertz Spectroscopy, Kerr Effect, nonlinear spectroscopy, molecular reorientation

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1. Introduction

Recent advances in high-field THz pulse generation [1-3] have led to the development of nonlinear THz spectroscopies [4-11]. Large electronic nonlinearities have been observed in a variety of semiconductor systems [4-10]. In particular, nonlinear transmission measurements have shown free carrier absorption saturation at high THz field strength at which free carriers, accelerated by the THz field, undergo scattering into lower-mobility side valleys of the conduction band that have substantially lower THz absorption [4,7,10]. Subsequent THz-pump/THz-probe studies measured the relaxation dynamics of the absorption saturation process [4,10]. Nonlinear transmission and THz-pump/THz-probe measurements on the low bandgap semiconductor InSb traced the dynamics of free carrier multiplication by impact ionization [5,6] and have shown evidence of lattice phonon excitation resulting from relaxation of the hot carriers in such systems. Similarly, lattice mediated nonlinearities have been observed in ferroelectric crystals [11], where driving anharmonic crystal motions with a THz field have led to self-phase modulation and harmonic generation in the THz range.

In this paper, we present results from THz-pump/optical-probe studies on liquids and ferroelectric crystals. The THz pump field introduces an optical anisotropy in the sample which is measured by polarization analysis of an optical probe field. By analogy to the optical Kerr effect, we call this the THz Kerr effect.

2. Experimental Methods

The experimental setup is shown in Fig. 1. Single-cycle THz pulses with energies exceeding 1.5 μJ were generated by the tilted pulse front technique [1]. The generated field was collimated and focused onto the sample where the THz intensity exceeded 50 MW/cm^2. A rectangular Infrasil cuvette with a path length of 5 *mm* was used for liquid samples.

A weak 800 *nm* probe beam was passed through the sample collinearly with the THz pulse at a

polarization of 45 degrees with respect to the THz polarization. A combination of a quarter-wave plate and a Wollaston prism was used to analyze the change in refractive index. Two balanced photodiodes and a lock-in amplifier were used to record pump-probe data. In order to assess the field strength and temporal shape of the THz field at the sample position, electro-optic sampling [12] with a 0.1 *mm* ZnTe crystal was conducted.

Since in our experiment, the THz excitation pulse and the optical probe pulse travel through the sample together, it is important to achieve velocity matching. The time-domain signal is optimal if the phase velocity of the THz pulse causing the birefringence and the group velocity of the optical pulse probing the polarization are identical. In this case the optical pulse envelope remains fixed at the same part of the THz wave cycle or at the same delay relative to the THz pulse as the two pulses propagate through the thick sample. The intrinsic matching of the optical (group) refractive index and the THz refractive index in our samples allows us to use long path lengths, thus enhancing measurement sensitivity to small refractive index changes.

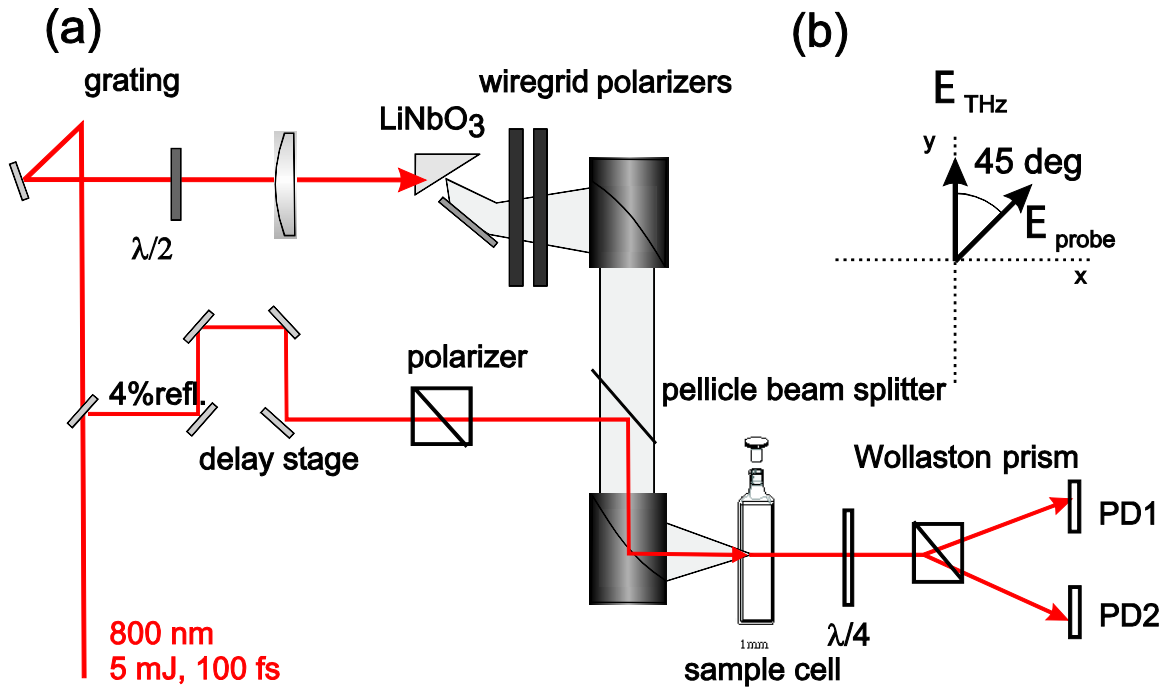

Fig. 1 (a) Experimental setup. THz radiation was generated by the tilted pulse front method and focused onto the sample in a 5 *mm* quartz cell. Light pulses with 800 *nm* wavelength and 100 *fs* duration were used to probe the sample, and their induced depolarization was analyzed. (b) The probe polarization at the sample was at 45 degrees with respect to the THz polarization.

3. Results and Discussion

Fig. 2a shows normalized THz Kerr effect scans for CS_2, CH_2I_2, benzene, CCl_4 and chloroform. All the liquids Kerr responses show a fast electronic component that follows the square of the THz pump field (shown under the $CHCl_3$ curve). CS_2 and CH_2I_2 exhibited the strongest THz Kerr effect signals, and showed slow decays due to their orientational contributions to the polarizability [13,14]. CCl_4 has no orientational component to its signal

because of its symmetry. $CHCl_3$ should show orientational signal but the level is apparently too low to detect above noise in our current measurements. Fig. 2b shows the expected quadratic scaling of THz Kerr effect signal level with THz pump field strength.

THz Kerr effect responses were also observed in a 200-micron thick crystal of the relaxor ferroelectric $KTa_{1-x}Nb_xO_3$ (KTN) with x = 0.09 at room temperature, in the paraelectric phase (Fig. 3). Unlike the liquids discussed above, where the optical and THz refractive index values are comparable, velocity-matching was not possible in KTN due to the large mismatch in optical and THz index. This raises the possibility that the THz Kerr effect seen in this material is a fast electronic response and that the signal decays as the THz pump field propagates and is absorbed by the crystal. As such, the origins of this effect are under further study.

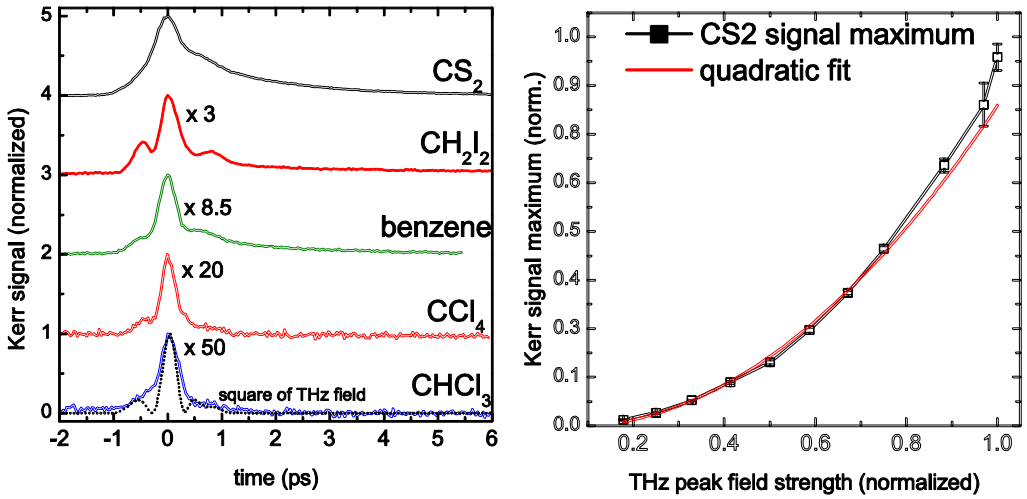

Fig. 2 (a) THz Kerr signals obtained from five different liquids. The dotted line indicates the square of the electric field measured by electro-optic sampling with ZnTe. (b) The magnitude of the Kerr signal (shown for CS_2) scales quadratically with the applied THz field.

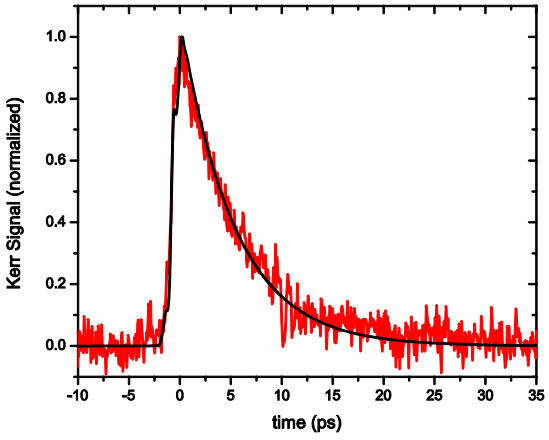

Fig.3 Normalized THz Kerr effect signal in KTN (red). The signal was fit to a convolution of the square of the THz field measured by electro-optic sampling in ZnTe with a single exponential decay of 5.2 *ps*. Note that measurement artifacts occur due to a reflection of the THz pulse in the KTN crystal at 10 *ps*.

The Kerr signal for CS_2 shows a characteristic decay slower than the timescale of the THz field. The time constant obtained from a fit to the slow decay is 1.7 *ps*, which agrees with previous measurements using all-optical Kerr effect methods [13]. A similar value of 2.1 *ps* was observed for benzene. For diiodomethane (CH_2I_2), a fit to the slow component of the signal gives a time constant of 13 *ps*. This is two times slower than what was previously reported [15].

4. Conclusions

THz Kerr effect measurements were performed on a variety of molecular liquids and the relaxor ferroelectric crystal, KTN. A fast electronic response was seen to follow the square of the THz field, and a slower orientational response was observed in several liquids. In KTN, a slow decay was observed whose origins are under further investigation. This work was supported in part by ONR Grant No. N00014-06-1-0459.

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