Optical Depolarization in Liquids and Second Harmonic Generation from the Surface Induced by Intense THz Pulses

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***Abstract*— The terahertz Kerr effect and terahertz induced optical second harmonic generation from the surface of different materials were investigated using intense THz pulses. Terahertz Kerr nonlinear coefficients and relaxation times for several liquids were measured. The significant increase of the optical second harmonic signal when strong THz field was applied were demonstrated and discussed theoretically.**

I. INTRODUCTION

PROGRESS made in the last decade in the development of intense table-top terahertz sources has stimulated nonlinear studies in the terahertz range. Here we address two nonlinear effects: the terahertz Kerr effect and the terahertz-induced optical second harmonic (SH) generation from the surface.

The terahertz Kerr effect, where a high THz electric field induces depolarization of probe laser radiation in liquids, has been demonstrated recently [1]. In our experiment, in addition to low absorbing liquids studied in Ref. [1] (C_6H_6, $CHCl_3$, CCl_4), we investigated liquids with strong THz absorption (acetone, water). A more elaborate theoretical model (which takes into account THz pulse dispersion, absorption and diffraction) was developed to determine nonlinear constants.

Optical SH generation from the surface is widely used for surface studies. The surface breaks the symmetry allowing SH generation even in isotropic media. Application of external electric field can also cause SH generation, that was used in Ref. [2] to measure a weak THz electric field. Here we investigated SH generation induced by intense THz radiation from dielectric, metal and semiconductor surfaces. An elegant theory of SH generation from a metal surface in the presence of a strong THz field was developed.

II. RESULTS

A Ti:Sapphire laser system (5 mJ, 10 Hz, 70 fs) was used in our experiments. The optical beam was divided into two parts. The pump pulse generated THz radiation in a $LiNbO_3$ crystal by the tilted-front technique [3,4]. The terahertz pulse (energy 1-2 μJ and duration ~1 ps) was focused on the sample (liquid filled cuvette, semiconductor and metal surfaces) to produce 200-300 kV/cm THz electric field. The probe (weak) optical pulse passed a delay line and was superimposed with the THz beam on the sample.

For investigating the Kerr effect the optical probe pulse was polarized 45° relative to the THz polarization. The THz and optical pulses passed through a quartz cuvette (with 4 mm transmitted path length) filled with the liquid (acetone, water, chloroform, CCl_4, and benzene). The THz field E_{THz} induced a transient birefringence Δn inside the sample resulting in phase retardation $\Delta\varphi$ between the parallel and perpendicular (relative to the THz field) components of the probe pulse. This phase retardation was measured by cross-polarization technique. Figure 1 shows $\Delta\varphi$ as a function of time delay τ between optical and THz pulses for some liquids (acetone and benzene). At $0 < \tau < 6$ ps for all liquids we observed approximately constant nonzero value of $\Delta\varphi$ (see Fig. 1(a)) due to the THz induced depolarization in an entrance wall of the quartz cuvette (of 4.2 mm long). At a later time, $\Delta\varphi(\tau)$ showed a fast electronic part ($6.5 < \tau < 7.5$ ps) and slow decays ($\tau > 7.5$ ps) for some liquids with nonsymmetrical molecular (acetone, benzene) due to the orientational contributions from the molecules. Minimum THz induced Kerr signal we observed in liquids with nonzero dipole molecular moment $d \neq 0$ (acetone, chloroform) and maximum for liquids with $d = 0$ (benzene, CCl_4). Indeed, acetone and chloroform have strong THz absorption that significantly limits interaction length between THz and optical pulses resulting in low $\Delta\varphi$. In benzene and CCl_4 the terahertz radiation have low absorption and good velocity matching with the optical probe pulse that gives high $\Delta\varphi$. Note, that we did not succeed in observation of optical depolarization in water. This is explained by low nonlinear coefficient and strong THz absorption in water.

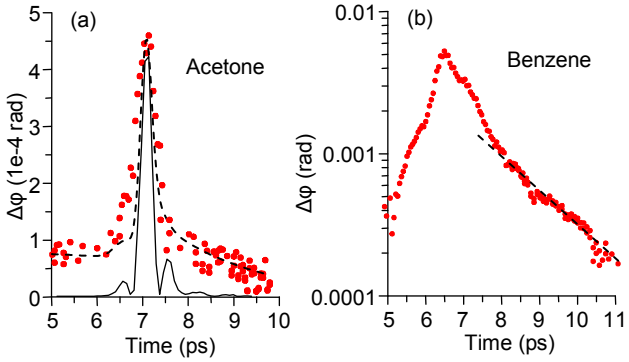

Fig. 1. THz induced phase retardation in (a) acetone and (b) benzene as a function of time delay. (a) Solid curve – squared THz electric field (peak THz field 300 kV/cm), dashed line – theoretical calculation. (b) Dashed line – exponential fit to orientational contribution.

For estimation of fast electronic and slow orientation contributions to the nonlinear response we developed a theory for calculation of $\Delta\varphi(\tau)$. In the theory, we took into account diffraction, dispersion and absorption of the THz pulses. Under assumption of short probe pulse duration (compare to the duration of the THz pulse) the phase retardation reads as $\Delta\varphi(\tau) = \frac{2\pi}{\lambda_{opt}} \int_0^L \Delta n(\tau + z/V_g, z)dz$, where V_g – group velocity of the probe pulse, λ_{opt} – optical wavelength, L – length of the

sample, $\Delta n = \Delta n^e + \Delta n^o$ with orientational $\Delta n^o = n_2^o \tau_{NL}^{-1} \int_0^\infty I^{THz}(t-t',z) \exp\left(-t'/\tau_{NL}\right) dt'$ and electronic $\Delta n^e = n_2^e I^{THz}(t,z)$ parts, where intensity of THz radiation $I_{THz}(t,z) = c/(4\pi) E_{THz}^2(t,z)$, n_2^e and n_2^o electronic and orientational nonlinear coefficients, respectively, τ_{NL} – relaxation time. Values n_2^e, n_2^o and τ_{NL} were found by fitting experimental data according to theoretical model (Table I). For water we can only estimate an upper limit of electronic nonlinear coefficient $<5\cdot10^{-16}$ cm^2/W that gives the absence of the Kerr signal with experimental accuracy.

Table I. Electronic and orientational nonlinear coefficients

Material	n_2^e, 10^{-16} cm^2/W	n_2^o, 10^{-16} cm^2/W	τ_{NL}, ps	n_2, 10^{-16} cm^2/W from Ref. [1]
Fused quartz	3.5	–	–	no
Acetone	25	85	1	no
Chloroform	$n_2 = n_2^e + n_2^o$ =15		<0.3	10
CCl_4	10	–	–	27
Benzene	14	40	2	56

We also investigated generation of the second optic harmonic induced by a high-intensity THz radiation. Different materials including semiconductors (silicon, GaAs), metals (Au, Cu) and quartz at different relative polarizations of THz, optical and SH fields were investigated. The THz-induced SH signal was measured using a PMT (Hamamatsu R4220) in reflection geometry for nontransparent materials with incident angle of optical and THz pulses ~ 35°. Transmission geometry was used for optically transparent materials.

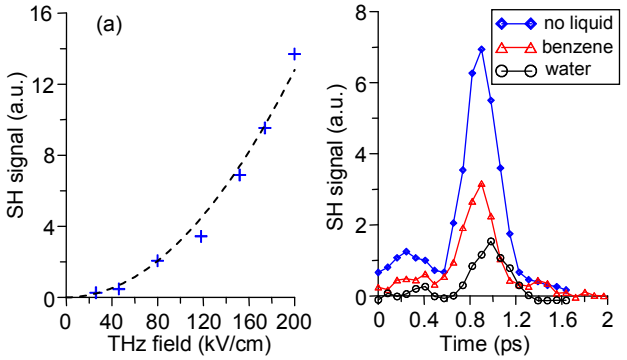

Fig. 2. (a) SH signal from Au as a function of THz field for s-polarization of SH, probe and THz, dashed line shows quadratic fit. (b) SH signal from the inner surface of quartz cuvette filled with different liquids as a function of time delay (peak THz field 200 kV/cm).

We observed a significant increase of the SH signal from the metal and silicon when a high THz field was applied (Fig. 2(a)). Polarization properties of the THz–induced SH was found to be in agreement with the symmetry of the electric field induced SH generation process. A theory describing SH generation from the metal in the presence of a strong THz field was developed. It was observed that the SH from the inner surface of quartz cuvette depend on the liquid containing inside (Fig. 2(b)) and decreased when the cuvette was filled up.

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