Absorption saturation of inter-molecular vibrational bands of molecular crystal observed by intense THz pulse irradiation

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Terahertz induced nonlinear effects have been observed in variety of semiconductors, such as Si [1]. Jewariya *et al*. reported the nonlinear response of amino acid microcrystalline L-arginine using an intense monocycle THz pulse [2]. The nonlinear response of the absorption bands provides us information of the molecular interactions, such as hydrogen-bondings and the unharmonicity of the interaction potential, etc. Here we report another example of saturable absorption of bio-molecular crystal in THz frequency region.

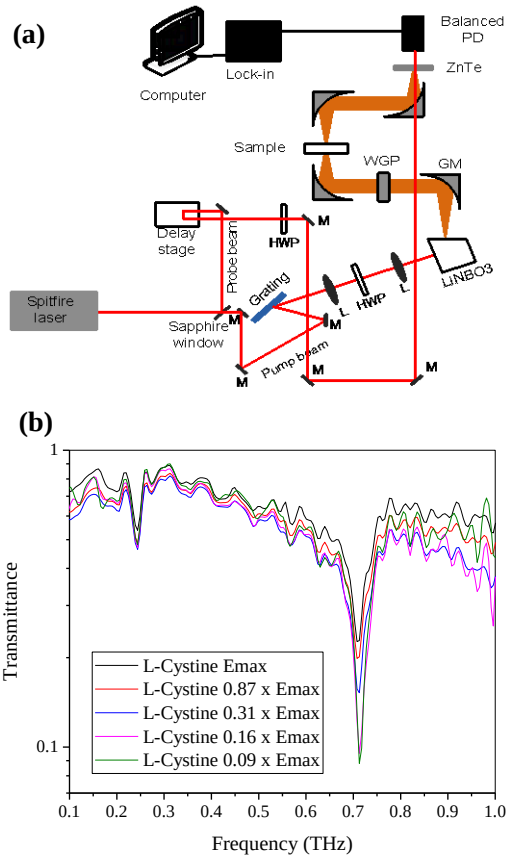

Fig. 1 (a) Experimental setup used in this study, (b) Transmittance spectra of L-cystine sample at various THz electric fields.

The sample used in this study was L-cystine crystal, the oxidized dimer form of the amino acid cysteine. The sample was prepared in the form of a pellet with a thickness of 1.2 mm and a diameter of 13 mm. The THz pulse was generated by using tilted pulsed front scheme with a Mg-doped stoichiometric $LiNbO_3$ crystal as shown in Fig. 1(a). The maximum THz electric field E_{max} at the focal point of THz beam was estimated to be 450 kV/cm. The terahertz electric field was varied by using a pair of wire-grid polarizers. A femtosecond laser system (800 nm, 100 fs, 1 kHz, maximum pulse energy ~ 3 mJ) was used as the optical source for the generation and detection of THz emission. THz radiation is guided with four off-axis parabolic mirrors of different focal lengths to the EO detection optics consisting of ZnTe crystal (t=1 mm) and a balanced photodetector. The time-domain THz waveform was obtained in a standard THz-TDS scheme. The sample was placed at the focal point of THz beam in the four parabolic mirror system. All the measurements were conducted at room temperature at around 20^{O}C. Figure 1(b) shows the transmittance spectra of L-cystine at various THz electric fields. The inter-molecular absorption bands observed at 0.24 THz and 0.71 THz [3] show saturation of absorption with increasing the THz electric field. However, their frequency shifts and broadening of the absorption bands are small, indicating the unharmonicity of these vibration modes are not strong. This result shows THz field in the order of ~100 kV/cm is already strong enough to investigate saturable absorption of small bio-molecules and their unharmonicity in inter-molecular potentials.

References:

1. S. Li, G. Kumar, T.E. Murphy, Optica **2**, 553 (2015).
2. M. Jewariya, M. Nagai, and K. Tanaka, Phys. Rev. Lett. **105**, 203003-1 (2010).
3. N. N. Barndt, *et al*, Vibrational Spectroscopy **47**, 53-58 (2008).