THz Field Induced Second Harmonic Generation in Epsilon Near Zero Indium Tin Oxide Thin Films

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Abstract: Epsilon-near-zero (ENZ) materials boost strong nonlinear optical effects. In this study THz pulses break the symmetry enabling THz-field-induced second-harmonic generation in ENZ thin films. A four-wave-mixing model matches experiments, revealing strongly coupled linear–nonlinear dynamics in the NIR–THz regime.

Epsilon-near-zero (ENZ) materials have recently emerged as promising platforms for enhanced and tunable light–matter interaction. In the field of nonlinear optics such materials demonstrated their capability to boost nonlinear optical processes even at relatively low optical fluences, thanks to the substantial field enhancements realized [1]. Indium tin oxide (ITO), featuring ENZ properties in the telecom wavelength range, is one of the most broadly explored conducting oxides thanks to the easy fabrication. Yet, being centrosymmetric, ITO thin films are commonly applied for the enhancement of odd nonlinear processes, such as third harmonic generation [2]. In this work, we employ a ITO thin film to study THz-field-induced second harmonic generation (SHG), highlighting a THz-controlled χ^2 response in a nominally centrosymmetric ENZ material. This phenomenon provides a new route for integrating ultrafast, field-programmable nonlinear functions into THz-driven photonic systems.
Using high-field, single-cycle THz pulses synchronized with a near-infrared pump (here named probe), we demonstrate that an intense THz bias is capable of dynamically breaking the inversion symmetry in ITO via strong perturbation of conduction-band electrons (see Figure 1a). The induced asymmetry enables SHG that is absent under equilibrium conditions. As expected, the generated SHG signal scales quadratically with the THz field amplitude (i.e. linearly with the THz field intensity) and is maximized when the NIR probe is tuned near/above the ENZ wavelength (i.e. > 1250 nm), where the internal field enhancement boosts the effective nonlinear susceptibility [3] (see Figure 1b). Time-resolved measurements show sub-picosecond gating of the nonlinear response, directly following the THz waveform and enabling precise temporal control of the frequency-conversion process. A nonlinear Drude-type model incorporating field-dependent carrier dynamics reproduces the observed behavior and confirms that the dominant mechanism originates from strong-field electronic distortion rather than structural effects.

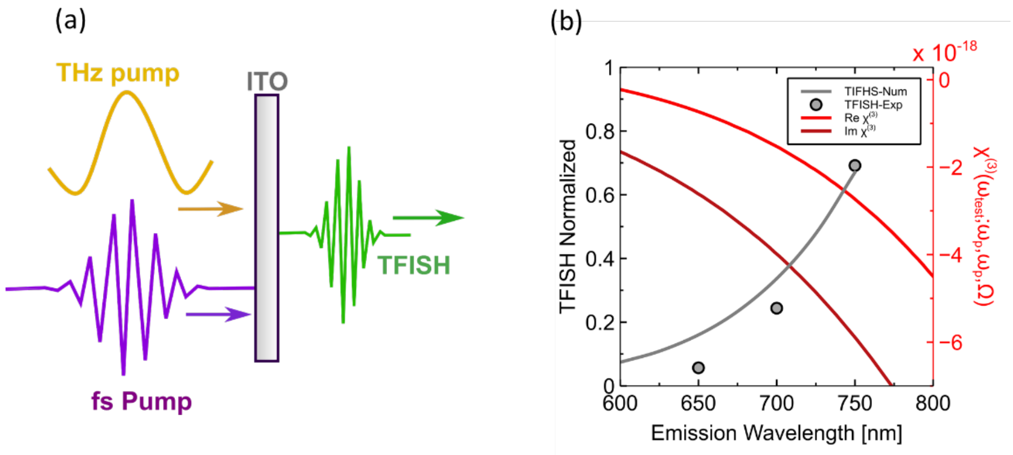

Figure 1 (a) Schematic diagram of the TFISH four wave mixing process in centrosymmetric ITO. The spatial and temporal overlap of a NIR fs pulse and THz pulse results in the emission of a TFISH pulse at double the frequency of the fs pump pulse. (b) The calculated and measured TFISH signal as a function of emission wavelength which scales directly with the third order nonlinear susceptibility of the ITO thin film.

These results establish ITO at its ENZ condition as a versatile platform for THz-programmable nonlinear optics and demonstrate a practical pathway for achieving dynamic χ^2 functionalities in compact thin-film devices. The approach opens new opportunities for THz detection, ultrafast signal modulation, and active integration of ENZ materials into emerging THz photonic architectures.

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