Time-Resolved Nonlinear Refraction of Indium Tin Oxide at Epsilon Near Zero

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Abstract: Using Beam-Deflection measurements we directly measure the temporal dynamics of nonlinear refraction and transmittance of an ITO thin film in the $\mathcal{E}$-near-zero regime, and find a nonlinear index 5000 times that of glass.

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1. Introduction

Indium tin oxide (ITO) is a widely used transparent conductive oxide with multiple applications in photonic devices due to its ease of fabrication and tailoring its properties by engineering the doping carrier concentrations [1,2]. Nonlinear photonic devices need large ultrafast nonlinearities. Recently, large nonlinear optical responses for the epsilon-near-zero (ENZ) regime have been reported [3,4]. This enhancement has been attributed to the low dielectric constant, i.e. small linear index of refraction, in the ENZ regime [3]. Materials showing a near zero real part of the permittivity show an enhanced nonlinear refraction which provides larger changes in the refraction for the same change in permittivity. Knowledge of the ultrafast dynamics and the magnitude of nonlinear refraction near the ENZ wavelength helps to realize the physical origin of the fast and large nonlinearities happening in this region.

In this paper, we present direct measurements of the time-resolved nonlinear refraction of a thin layer of ITO (~ 310 nm) with a sheet resistance of 5 Ω/sq (Präzisions Glas & Optik GmbH), deposited on a float glass substrate of thickness 1.1 mm. Using our Beam-Deflection (BD) technique [5], we quantify the ultrafast temporal response near its ENZ wavelength.

2. Experimental Results and Discussion

Beam-Deflection [5] is an extension of the excite-probe technique capable of measuring the temporal dynamics of the change in the refractive index of the material induced by a strong excitation pulse. Here, we use a Ti:sapphire laser system (Coherent Legend Elite Duo HE +) at a repetition rate of 1 kHz with ~40 fs, 800 nm pulses to pump an optical parametric amplifier (Light Conversion, TOPAS-HE) whose output is filtered by a bandpass interference filter centered at 1242 nm. A portion of the excitation beam is used to generate a white-light-continuum by focusing the beam into a 5 mm thick sapphire plate and then filtering by a bandpass filter at 1050 nm to use as the probe to avoid coherent artifacts observed in degenerate experiments [6]. In this method, a ~ 3-5 × smaller probe beam spot size (w_p = 90 μm) is focused on the wings of the Gaussian index gradient induced by the temporally delayed excitation beam in the sample with a larger spot size (w_e = 380 μm). The induced phase gradient deflects the probe beam as measured using a position sensitive quad-segmented silicon detector. To detect the time dynamics signal we modulate the 1 kHz repetition rate excitation beam at 285 Hz for lock-in detection. The normalized difference of the energy on the left and right sides of the detector in the far-field, $\Delta E/E$, and normalized change in the transmission signal, $\Delta T/T$ versus delay provide the transient change in nonlinear refraction (NLR) and absorption (NLA) respectively.

The linear refractive index of the ITO thin film was determined using spectroscopic ellipsometry and analyzed by the Drude oscillator model to find the ENZ point as shown in Fig. 1(a). We perform time-resolved BD measurements at normal incidence on the ITO thin film as well as the bare substrate with parallel and perpendicular polarization at two different excitation pulse energies to measure the temporal dynamics of the nonlinear absorption, Fig. 1(b), and refraction, Fig. 1(c). The difference between parallel and perpendicular polarizations vanishes which indicates that the mechanism responsible for the reported dynamics is out of equilibrium electron scattering in the conduction band and is not a bound electronic response. We may also not be in the regime of exact third-order nonlinear response since the signals do not scale linearly with excitation energy as seen in Fig. 1(b,c) and the scaling of NLR is slightly different from the scaling of NLA. This apparent saturation is currently under study. An exponential decay time (τ_f) of ~ 220 fs for transmission and ~ 320 fs for refraction is observed, which is likely due to different hot carrier scattering mechanisms.
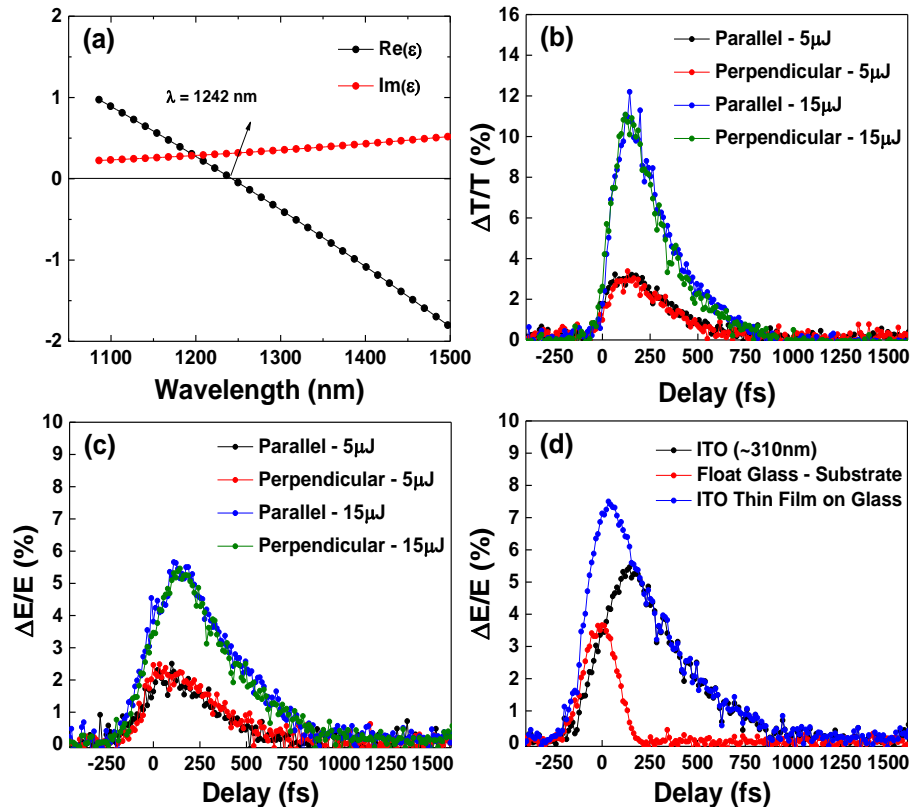
Fig. 1. (a) Real and imaginary parts of the permittivity of a thin ITO film obtained from spectroscopic ellipsometry measurement and analyzed by Drude oscillator model, the ENZ condition happens at λ_0 = 1242 nm (b, c) Change in the transmission (NLA) and BD signal (NLR) for pulse energies of 5 µJ and 15 µJ for parallel and perpendicular polarization of excite and probe (d) BD measurement of the ITO sample deposited on a glass substrate, signal from the glass, and subtracted signal, all for parallel polarizations.

The magnitude of the measured effective nonlinear refraction is ~ 5000 × larger than that of glass and on the order of 10^{-3} cm²/GW as shown in Fig. 1(d). The peak index change averaged through the sample is as large as $\Delta n \approx 0.1$. Further enhancement is obtained at different angles of incidence which will be presented. We observed similar temporal responses by changing the probe to a shorter wavelength. However, probing at longer wavelengths, closer to the ENZ wavelength, shows larger enhancement. We note that this nonlinear refraction measurement technique makes the result independent of nonlinear absorption. The observed nonlinear transmission sign change is consistent with the change in the index of refraction; however, experiments measuring both reflection and transmission are needed to fully determine the contribution due to nonlinear absorption, and these are in progress. This enhancement in nonlinear refraction is due to the ultrafast dynamics of the hot carriers excited by the excitation beam into the conduction band, and it opens a possibility for this material to be utilized in different applications requiring a large nonlinear refractive index such as all-optical switching photonic devices.

3. Acknowledgment

4. References

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