Nondegenerate, Transient Nonlinear Refraction of Indium Tin Oxide Excited at Epsilon-Near-Zero

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Abstract: We measure nondegenerate nonlinear refraction of ITO as we tune the excitation wavelength through Epsilon-Near-Zero (ENZ) for a fixed probing wavelength. Unlike the degenerate case, there is no observed peak in the nonlinearity at ENZ.

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1. Introduction

Materials at wavelengths where the real part of the permittivity is near zero, so-called "Epsilon-Near-Zero" (ENZ), have received significant attention due to their unique nonlinear optical features such as enhanced second and third harmonic generation [1,2], enhancement in nonlinear refraction [3], and applications including all-optical switching [4]. Recent studies on Indium Tin Oxide (ITO) thin films have shown strong enhancement in NLR in the ENZ regime [3]. For any given change in permittivity, the change in index of refraction, $\Delta n \propto \Delta\varepsilon/\sqrt{\varepsilon}$ will be large for materials showing near zero real part of the permittivity. $\Delta\varepsilon$ can be dependent on both linear and nonlinear properties of the material. In the case of bound electronic nonlinearities, this change depends upon the third-order susceptibility and linear index of refraction for non-degenerate nonlinearities, as follows [5]:

$$\Delta n(\omega_p) = n_2 I_e = \frac{1}{2n_p}\left(\frac{3}{2}Re\{\chi^{(3)}(\omega_p;\omega_e,-\omega_e,\omega_e)\}|E_e|^2\right), \quad (1)$$

where the index change is observed at ω_p, the optical frequency of the probe wave due to an excitation of irradiance I_e at frequency ω_e. Because $I_e = 1/2\, n_e c\varepsilon_0 |E|^2$, we find that $n_2 = 3Re\{\chi^{(3)}(\omega_p;\omega_e,-\omega_e,\omega_e)\}/(2n_p n_e \epsilon_0 c)$ in which case the n_2 can be enhanced when either excitation or probe wavelengths are at ENZ for bound-electronic nonlinearities. However, in the case of carrier-redistribution type nonlinearities of the type seen in ITO, the effect depends on the rate of excitation of carriers by the pump and not directly on the magnitude of the electric field in the excitation beam. In this case only the probe wavelength at ENZ will show an enhancement. An additional factor in these types of experiments is that a large discontinuity of the normal electric field component can be obtained for TM polarization at oblique incidence [1,2], which has shown further enhancement in NLR at ENZ [3].

Here, we apply the recently developed Beam-Deflection (BD) technique [6] to directly measure the time-resolved NLR of a thin layer of ITO (~310 nm) deposited on a float glass substrate, while we vary the wavelength of the excitation as well as the relative polarization between the two beams. We keep the probe at 1050 nm, reasonably far from ENZ, to reveal the separate effect of tuning the excitation wavelength through ENZ. In addition, the ability to study the dependence on relative polarization of excite and probe waves allows for separation of the ultrafast bound-electronic nonlinear refractive index from other non-instantaneous mechanisms such as carrier nonlinearities [6,7]. This is in contrast to other methods such as Z-Scan [3] which only measure the magnitude and sign of the temporally averaged nonlinearities.

2. Experiment and results

For our Beam-Deflection measurements [6], a Ti:sapphire amplified laser system (Coherent Legend Elite Duo HE +) with pulses at 800 nm, 1 kHz repetition rate, and a pulsewidth of ~100 fs is used. A portion of the output of the laser is focused on a 5 mm thick sapphire plate to generate a white-light-continuum probe and then filtered by a bandpass filter at 1050 nm. The excitation beam is from the output of an optical parametric amplifier (Light Conversion, TOPAS-800) whose output is filtered by a bandpass filter at the desired wavelength. The probe wavelength is chosen to be nondegenerate to the excitation in order to avoid artifacts in degenerate experiments and to remain within the spectral range of the silicon detector. This also allows us to separate the effects of the excitation being near ENZ from effects of the probe near ENZ. The polarization angle of the excitation and probe beams are independently controlled by half-wave plates. The strong excitation pulse induces an index gradient inside the sample that follows the Gaussian irradiance profile of the beam. By focusing the weak probe beam to a spot size smaller than that of the excitation beam and displacing it to the wings of the excitation beam, the index gradient deflects the probe by a small angle [6]. This deflected signal is recorded using a quad-segmented silicon detector. The excitation beam is modulated with a mechanical chopper at 285 Hz that was synchronized with the output from the laser. The BD signal is taken to be the normalized difference of the energy falling on the two sides of the detector in the far-field,

$\Delta E/E$ versus the delay between excitation and probe pulses. The signal is proportional to the phase change accumulated by the weak probe beam due to the strong excitation pulse and provides the transient change in NLR including sign and absolute magnitude of each mechanism, their rise and fall time constants, and polarization dependencies.

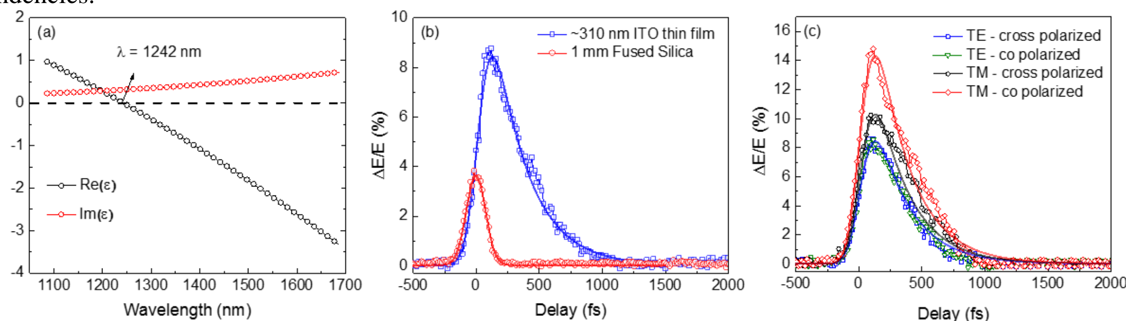

Fig. 1. (a) Real and imaginary part of the permittivity of a thin ITO film obtained from spectroscopic ellipsometry measurement and analyzed by Drude oscillator model, (b) BD signal (nonlinear refraction) for pulse energies of 10 µJ for ~310 nm ITO thin film and 1 mm thick fused silica with an excitation beam at 1242 nm, (c) BD measurement of the ITO thin film at 60° angle of incidence for TM and TE polarized light in parallel and perpendicular polarization combinations between excitation and probe beams.

The electric permittivity of the ITO thin film is experimentally measured using variable angle spectroscopic ellipsometry, and the result is fit using the Drude model to find the ENZ point as shown in Fig. 1(a). The ENZ condition occurs at λ_0= 1242 nm at the plasma frequency and the electric permittivity remains between -1 and 1 for the spectral range between 1100 nm and 1400 nm. We performed BD measurements on a 1 mm thick fused silica and fit the data using a bound-electronic instantaneous third-order response which agrees with literature values [6]. The maximum signal happens where the delay between excitation and probe pulses is zero. This establishes the zero time-delay position. Similar measurements are also performed on ITO at normal incidence for different excitation wavelengths and polarization combinations with a probe wavelength centered at 1050 nm. At normal incidence, the NLR does not show any significant difference for excitation wavelengths centered at 1200, 1242, and 1300 nm, indicating that the ENZ condition does not enhance the effect of the excitation beam. This is as expected for a carrier-redistribution effect, as the fractional energy absorption, $(1-R)(1-\exp(-2\,\kappa\omega d/c))$, with R the reflectance , κ the imaginary index and d the film thickness, is approximately the same for all three wavelengths. By varying the angle between the polarization direction of excitation and probe, we obtain the polarization dependence of NLR which usually helps to distinguish the bound-electronic instantaneous nonlinearities from non-instantaneous contributions [7]. The vanishing polarization dependence at normal incidence and delayed accumulated rise compared to the instantaneous response of fused silica indicates that the carrier redistribution mechanism contribution to NLR completely dominates the bound-electronic n_2 in ITO . The measurements shown in Fig. 1(b) are fit using a non-instantaneous response function corresponding to hot electron thermalization which is modeled by exponential rise (τ_r) and decay (τ_f) constants and a magnitude corresponding to the phase change. τ_r is taken to be much shorter than the pulsewidth. The fall time associated with the electron scattering mechanism is not dependent on the excitation wavelength and is ~220 fs. The BD measurements were also performed on the 1.1 mm thick float glass substrate under identical experimental conditions separately, and the contribution from the substrate is subtracted. The fit magnitude of the effective n_2 of the ITO film is 0.55×10^{-16} m^2/W which is ~ 1000 × larger than that of fused silica and ~ 5 × less than the previously reported values for normal-incidence, degenerate experiments at ENZ [3].

Similar measurements were performed on the ITO film at 60° incident angle. Four different combinations of TE and TM polarization of the excitation beam and the probe with parallel and perpendicular polarization are shown in Fig. 1(c). For TE polarized light, the parallel and perpendicular signals are similar to that for normal incidence, however, for TM polarized light the nonlinear refraction is enhanced by ~20% for perpendicular and ~70% for parallel polarization between excitation and probe beams. Additional measurements at different excitation wavelengths close to the ENZ point at oblique incidence are required to investigate this enhancement further. Experiments with probe beam near the ENZ wavelength are currently being performed using an InGaAs quadrant detector.

3. References

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