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Two-beam nonlinearity in indium tin oxide in the continuous wave limit

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ABSTRACT

Indium tin oxide (ITO) is an example of a material with a greatly enhanced optical nonlinearity for wavelengths at which the dielectric permittivity is near zero. Its enormous nonlinearity may enable compact photonic devices. All-optical devices involve multiple beams, and it was found recently [Paul *et al.*, Optics Letters **46**, 428–431 (2021)] that two-beam interaction modifies the effective nonlinearity in ITO for co-polarized beams. In that work, results of a degenerate pump-probe experiment were compared to a numerical model of the hot electron nonlinearity in ITO. The numerical model successfully explained the polarization dependent differential transmission and reflection, including the dependence on the chirp of the pulses. Here, we consider a simpler analytical model for the two-beam interaction in the continuous wave limit. We directly test this analytical model using a nondegenerate pump-probe transmission experiment using long pulses and find reasonable agreement.

Keywords: Nonlinear optics, ultrafast optics, transparent conductive oxides

1. INTRODUCTION

The nonlinear optics of transparent conductive oxides (TCO), such as indium tin oxide (ITO) and aluminum zinc oxide is a subject of growing interest.[1–8] At wavelengths where the dielectric function epsilon is near zero (ENZ), the effective optical nonlinearity is greatly enhanced.[1,2,4] The dominant effect in these materials is thought to be electron heating combined with an electron temperature-dependent optical response.[1,9–11] The heating builds up during a pulse, then falls rapidly as the electrons give up their thermal energy to the lattice. To lowest order, the electron temperature change is linear in optical intensity, causing an effective third-order nonlinearity. Both the refractive index and absorption coefficient depend on temperature, so the effective Kerr coefficient n_2 and the two-photon absorption coefficient β are both nonzero.

Many all-optical applications involve the interaction of two or more light beams. The coherent interaction of multiple beams in nonlinear media is complex, as it involves the generation of transient nonlinear gratings that diffract light from one beam into another. Consequently, the effective nonlinear coefficients for multiple beams can differ from one-beam coefficients. For the simplest case of a pure electronic nonlinearity ($\chi^{(3)}$), the nonlinear effect of one beam on another is simply multiplied by two.[12] For more complicated *effective* nonlinearities such as in ITO, other behavior can occur.[13] Depending on the nature of the nonlinearity, these gratings can lead to counterintuitive polarization and wavelength dependence of the two-beam nonlinearity. The effect of the grating depends on the temporal and spectral dependence of the underlying nonlinearity. This *two-beam coupling* (TBC) effect has been studied in many contexts.[13–20] Hot electron materials are different from most of these transparent materials, however, because the refractive and absorptive components of the susceptibility are of similar magnitude.

Previously, a simple theory of two-beam coupling developed for calculations of the two-beam coupling phase shift in transparent media[13] was adapted to model an experiment that used pump and probe beams at near normal incidence.[21] Here, we present results of a nondegenerate pump-probe experiment, which allows direct comparison to the continuous wave (cw) model of the two-beam nonlinearity in ITO.[21]

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2. THEORY

For pulses shorter than the characteristic timescale of the material, numerical modeling is typically required to simulate two-beam nonlinear effects.[13,17] In transparent conductive films, the effective nonlinearity is caused by changes in the electron temperature that accumulate during the pulse and decay over hundreds of femtoseconds. Numerical calculations were done in Ref. 21 to compare to experimental data that used 60 fs pulses, both Fourier transform limited and with chirp. In the limit of long pulses, the dynamics can be neglected and the spectral response can be explained using an equation with a few parameters.[21] Using numerical simulations and the observed cooling time of $\tau \approx 85$ fs, we find that the cw limit should work well for pulses of duration greater than 250 fs.

In deriving the analytical model, we closely follow the approach in Ref. 12, section 7.4, which was based on Refs. 14, 22. The decaying nonlinearity used was developed to model the nonlinearity in a photorefractive material, and it is suitable to model the thermal nonlinearity in ITO and related materials, but we must include an absorptive nonlinearity in addition to the refractive nonlinearity. For this we define a complex nonlinear coefficient $\nu = n_2 + i\beta/(2k)$, where β is the effective two-photon absorption coefficient, n_2 is the effective Kerr coefficient, and $k = \omega/c$. The nonlinear refractive index change versus time obeys[12] $\tau(d\Delta n/dt) + \Delta n = \nu I$. We use the observed electron cooling time $\tau \approx 85$ fs.[21]

We assume two beams with central frequencies ω_1 (pump) and ω_2 (probe) that are sufficiently closely spaced that we can approximate $\omega = (\omega_1 + \omega_2)/2$ in place of ω_1 or ω_2. We are assuming that n_2 and β are the same at ω_1 and ω_2, which holds as long as $\omega_1 - \omega_2$ is relatively small. Defining $\delta = \omega_1 - \omega_2$, the evolution of the complex envelope of beam 2 is

$$\frac{dA_2}{dz} = 2in_0\nu\omega\epsilon_0 \left[\left(|A_1|^2 + |A_2|^2\right) A_2 + \frac{|A_1|^2 A_2}{1 + i\delta\tau} \right]. \tag{1}$$

To derive the gain or loss, we introduce intensities $I_1 = 2n_0\epsilon_0 c A_1 A_1^*$ and $I_2 = 2n_0\epsilon_0 c A_2 A_2^*$, so that

$$\frac{dI_2}{dz} = 2n_0\epsilon_0 c \left(A_2^* \frac{dA_2}{dz} + A_2 \frac{A_2^*}{dz} \right). \tag{2}$$

This leads to

$$\frac{dI_2}{dz} = -\beta I_2^2 - \beta I_1 I_2 - \beta \frac{I_1 I_2}{1 + \delta^2\tau^2} + \frac{2n_2\omega}{c} \frac{\delta\tau}{1 + \delta^2\tau^2} I_1 I_2. \tag{3}$$

The first term corresponds to effective two-photon absorption of the probe beam acting on itself. The second term corresponds to two-photon absorption where one photon comes from the probe and the other comes from the pump. The third term is additional probe absorption caused by two-beam coupling. The fourth term is the conversion of the refractive nonlinearity to absorption through two-beam coupling.

We can define a modified absorption coefficient for the effect of beam one on beam two,

$$\beta' = \beta \left(1 + \frac{1}{1 + \delta^2\tau^2} \right) + n_2 \frac{2\omega}{c} \frac{\delta\tau}{1 + \delta^2\tau^2}. \tag{4}$$

3. EXPERIMENT

We performed a pump-probe experiment to test the analytical theory for long pulses. The light source is an optical parametric amplifier (OPA) pumped by a 1030 nm Yb:KGW laser. A $4f$ pulse shaper is used with a spatial light modulator (SLM) to generate nondegenerate pump and probe beams from the OPA spectrum. A grating spreads the OPA spectrum across the SLM horizontally. The SLM creates blazed gratings that diffract a small bandwidth of the light vertically into two output beams, one of which becomes the pump beam and the other the probe beam. The probe beam is delayed using a translation stage, and both beams are focused on the sample. The probe beam is focused with a 60 mm focal length lens and the pump beam is focused with a 120 mm focal length lens. The relative angle between beams is approximately 10°.

The probe beam differential transmission is measured using a mechanical chopper and a lock-in amplifier as a function of time delay between the pump and probe pulses. The pulse duration of each beam is approximately

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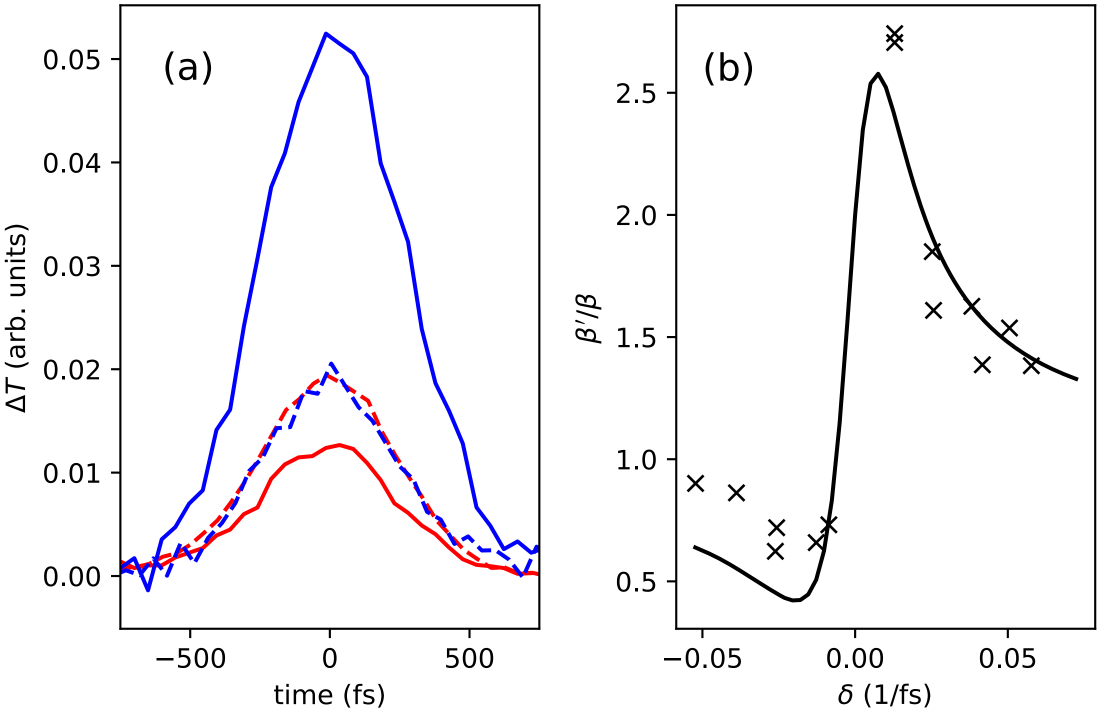

Figure 1. Experimental results. (a) Pump-probe traces for $\lambda_{\text{probe}} = 1216$ nm and two different pump wavelengths: $\lambda_{\text{pump}} = 1206$ nm (blue) and $\lambda_{\text{pump}} = 1226$ nm (red). The solid lines are for parallel polarization and the dashed lines are for perpendicular polarization. (b) Measured normalized two-beam nonlinear coefficient β'/β as a function of detuning parameter $\delta = 2\pi(c/\lambda_{\text{pump}} - c/\lambda_{\text{probe}})$.

400 fs, which is sufficiently long to be in the long pulse limit. Figure 1a shows pump-probe traces for parallel and perpendicular polarization for an ITO sample at near normal incidence. The duration of the transmission signal agrees with a cross correlation measurement using second harmonic generation.

The two-beam coupling contribution is isolated by dividing the peak transmission change for parallel polarization by that for perpendicular polarization.[21] This normalized nonlinear coefficient β'/β is plotted in Fig. 1b for four different sets of pump and probe wavelengths λ_{pump} and λ_{probe} as a function of the detuning parameter $\delta = 2\pi(c/\lambda_{\text{pump}} - c/\lambda_{\text{probe}})$. Since the perpendicular signal is proportional to the one-beam nonlinearity β, the theoretical polarization anisotropy ratio is

$$\frac{\beta'}{\beta} = 1 + \frac{1}{1+\delta^2\tau^2} + \frac{n_2}{\beta}\frac{2\omega}{c}\frac{\delta\tau}{1+\delta^2\tau^2}. \tag{5}$$

We find quite good agreement using the values of β and n_2 found in Ref. 21 if we use the magnitude of n_2 reported in Ref. 21.

4. CONCLUSION

In summary, we have tested an analytical model of the two-beam optical nonlinearity in ITO using a nondegenerate pump-probe experiment using narrow bandwidth pulses. This experimental scheme may prove useful for probing two-beam effects in ENZ films in a way that can be more straighforwardly simulated. Our results indicate that the nonlinear coefficients can indeed be tuned by adjusting the relative wavelength of the two beams, as proposed in Ref. 21.

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