Giant nonlinear frequency shift in epsilon-near-zero aluminum zinc oxide thin films

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Abstract: Degenerate pump/probe experiments have been performed on aluminum zinc oxide thin films at their epsilon-near-zero wavelength. A remarkable spectral shift of the reflected beam is reported to be approximately twice the pulse bandwidth.

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1. Introduction

The concept of time varying metasurfaces promises to enable ultra-fast control over the propagating electromagnetic wavefront by inducing a time phase gradient across an optical interface. In particular, the temporal variation of the refractive index n in time (dn/dt) leads to a time phase gradient of the incident radiation and to a consequent frequency shift according to $\Delta\omega = -d\varphi/dt = -k_0 L(dn/dt)$ [1]. Clearly, such effect is maximized when large nonlinearities are associated to ultra-fast dynamics, a condition which is rarely met in common materials. Due to these limitations, few experiments with time varying surfaces have been carried out, for instance by exploiting four-wave mixing (FWM) processes in graphene sheets [2]. However, in general a very low efficiency of the nonlinear effects has prevented the observation of a remarkable frequency shift.

Recently, it has been shown that Transparent Conductive Oxides (TCOs) exhibit large and ultrafast nonlinear optical response [3,4]. In particular, such nonlinearities are enhanced at Epsilon-Near-Zero (ENZ) wavelengths, where the near-zero value of the linear refractive index leads to the enhancement of the nonlinear optical properties in terms of the Kerr coefficient n_2 [3]. It has also been shown that an optically thin film of Aluminium-doped Zinc Oxide (AZO) behaves like an ideal time-varying medium at ENZ wavelengths, with an efficiency of the time reversed phase conjugate beam of the order of the unity [5].

In this work, using a pump and probe system in its degenerate configuration, we demonstrate how we can further boost the nonlinearities of AZO thin films by combining the enhancement attained by working in the ENZ spectral region with another raising from a higher third order nonlinear susceptibility $\chi^{(3)}$. As a consequence of this nonlinear enhancement, we recorded a very large frequency shift for the reflected beam carrier approaching 50nm (approximately twice the probe bandwidth). This attainment is a direct consequence of the optically induce temporal phase gradient. The enhancement of the $\chi^{(3)}$ is directly linked to the degenerate configuration and can be predicted by the empirical model by Wang et al. [6].

2. Results

We carried out the nonlinear characterization of the AZO by measuring the variation in both transmission and reflection in a pump and probe experiment (Fig.1a) in its degenerate configuration, where the operational wavelength has been tuned between 1100 nm and 1500 nm. We retrieved the material $\chi^{(3)}$ and the change in the refractive index Δn by means of the inverse transfer matrix method [3]. We observe that the $\chi^{(3)}$ is enhanced at longer wavelengths following the semi-empirical model by Wang and co-authors [6]. This is reported in Fig.1b where a 5-fold enhancement of the third order susceptibility is observed around $\lambda \approx 1550nm$ in comparison with the non-degenerate case reported in [3]. From the same set of experimental data, we also evaluated the optically induced change in the real refractive index and the correspondent nonlinear Kerr coefficient n_2 which is 22 times the value reported in [3]. It is worth noting that, this initial characterization of the material nonlinearities has been carried out at relatively low pump intensity (<90GW/cm^2).

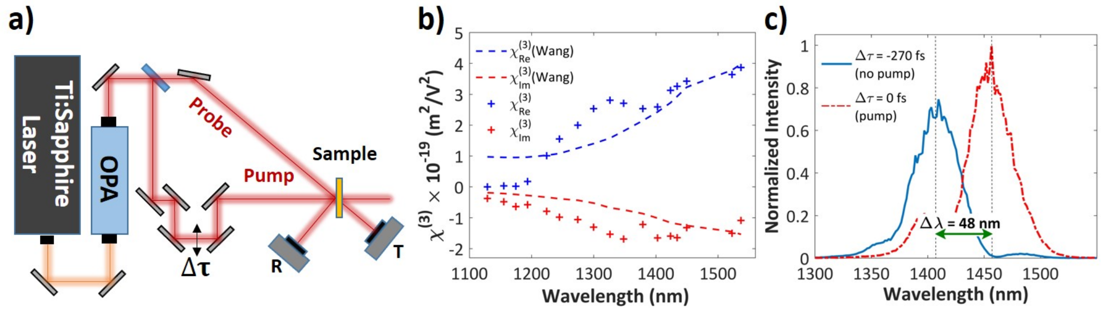

Figure 1: a) Sketch of the degenerate pump and probe setup. An Optical Parametric Amplifier (OPA), pumped by a Ti:Sapphire laser, provides optical pulses of about 100fs with a repletion rate of 100Hz with a central wavelength tuneable in the range between 1100nm and 1550nm. The pump and the probe are attained by splitting the OPA output while a delay line induces a temporal shift $\Delta\tau$ between the two pulse trains. Both reflected (R) and transmitted (T) beams are recorded as a function of the time delay. b) Real and imaginary part of the third order nonlinear susceptibility $\chi^{(3)}$as a function of the operational wavelength. Experimental data (+) are plotted together with the general trend as set by the application of the Wang rule [6]. c) Spectra of the reflected probe beam at 2 different time delays $\Delta\tau$ between the pump and the probe pulses. The blue solid line represents the probe spectra in the condition of no temporal superposition between pump and probe. The red dashed curve instead is recorded when both signals are overlapped in time. Pump power 690GW/cm^2.

As previously stated, the time variation of the refractive index is expected to produce a remarkable wavelength shift which we recorded by collecting the spectra of the reflected probe for different temporal delays $\Delta\tau$ between pump and probe. For 690 GW/cm^2 of incident pump intensity at 1400nm, where the AZO exhibits the maximum absolute variation of the real refractive index, we observed a remarkable wavelength shift as large as 48nm (see Fig. 1c). The measured frequency shift of the reflected probe is about twice the entire bandwidth of the incident pulses and in first approximation can be justified by cross-phase modulation [7].

3. Conclusion

In conclusion, we showed how a degenerate configuration enables full exploitation of the nonlinearities of ENZ AZO thin films. Under these conditions, the ultra-fast optically induced refractive index change is so large that the correspondent wavelength shift, induced by cross-phase modulation on the reflected radiation, is measured to be as large as twice the bandwidth of the incident pulse. Our results could be of key importance not only for telecom applications (e.g. ultra-fast all-optical routers) but also for designing novel flat components where a time phase gradient is used instead of the more typical space phase gradient.

4. Acknowledgements

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