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

E. G. Carnemolla¹, V. Bruno¹, L. Caspani², M. Clerici³, S. Vezzoli¹, T. Roger¹, C. DeVault⁴, J. Kim⁵, A. Shaltout⁶, V. Shalaev⁷, A. Boltasseva⁷, D. Faccio⁸ & M. Ferrera¹

1 Institute of Photonics and Quantum Sciences, Heriot-Watt University, SUPA, Edinburgh, Scotland, EH14 4AS, UK
2 Institute of Photonics, Department of Physics, University of Strathclyde, Glasgow G1 1RD, UK
3 School of Engineering, University of Glasgow, Glasgow, G12 8LT, UK
4 Dept. of Physics & Astronomy and Birck Nanotechnology Center, Purdue University, West Lafayette, IN, 47907, USA
5 Institute for Research in Electronics and Applied Physics, University of Maryland, College Park, Maryland 20742, USA
6 Gehalle Laboratory for Advanced Materials, Stanford University, Stanford, California 94305, USA. (A.S.)
7 School of Electrical and Computer Engineering, Purdue University, West Lafayette, IN 47907, USA

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. © 2018 The Author(s)

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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 \( \Delta \omega = -d\varphi/dt = -k_0L(dn/dt) \) [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 \( \Delta n \) 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/cm²).
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 \( \Delta t \) 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 nonlinearity 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.

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5. References