Optical time reversal from time-dependent Epsilon-Near-Zero media

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Abstract: We provide an efficient surface time-reversal of the incident electric field in an ENZ material producing both phase-conjugated and negative refracted beams. The results obtained exploiting degenerate four-wave mixing show an efficiency conversion over 200%.
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1. Introduction

The time response of materials to an optical perturbation is fundamental in many physical phenomena and is involved in a wide range of applications ranging from magnetic field-free optical isolators to fundamental studies of quantum field theories. Recently, a step forward in the study of materials with a spatially uniform but temporally varying optical response was made by proposing four-wave mixing (FWM) experiments in a deeply subwavelength medium (see J. Pendry [1]). Here the third order susceptibility (χ_3), excited by a pumping optical beam, induces a polarisation wave that oscillates at 2ω. The parametric oscillation will modulate a probe photon ω at the same frequency 2ω as it propagates through the medium and will induce a transition from a point (k,ω) to a point $(k,-\omega)$ on the dispersion plane. This corresponds to the time-reversal of a monochromatic field and leads to the emission of a backward propagating phase conjugate (PC) wave and a forward propagating negative refracted (NR) wave [1]. It is important to underline that this configuration of the FWM is different with respect to standard FWM in bulk material. In this configuration, conservation of the component of the momentum normal to the surface is relaxed, due to the fact that the nonlinear medium is much thinner than the effective wavelength, and only the component of the in-plane momentum needs to be conserved giving rise to both PC and NR beams with comparable efficiencies. These emitted beams can be considered as a clear indication of a time-dependent surface oscillating at 2ω.

The high nonlinearity required and the fact that the efficiency of this process scales quadratically with the interaction length are the major limitation in this time reversed media, resulting in low efficiency conversion in sub-wavelength materials (at best 0.1%)[2,3].

In this work we experimentally show how a thick film (500 nm) of Aluminium Zinc Oxide (AZO) can act as an extremely efficient time-reversing medium. It has been recently demonstrated that Transparent Conductive Oxides exhibit large nonlinearities in terms of Kerr coefficient when operating in the epsilon-near-zero (ENZ) region [4,5]. Within the ENZ region, the refractive index (n) of the material is also very small, and therefore, radiation inside the material will exhibit an effective wavelength that tends to infinity as n tends to zero. This fact implies that, in this spectral region a relatively thick sample of ENZ media will behave essentially as a deeply subwavelength thin film.

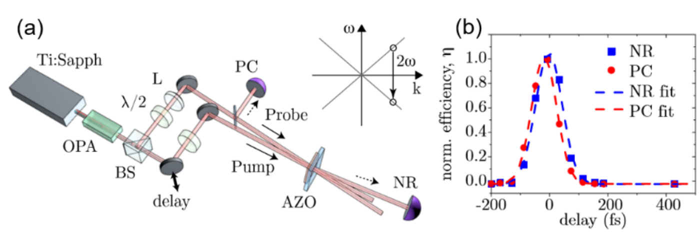

Fig.1. Schematics of the setup for degenerate FWM on AZO. The inset shows a sketch of the time-reversal process in the (k,ω) space. (b) Normalized PC and NR signals as function of the temporal delay between pump and probe beams.

2. Experimental Results

A schematic of the experimental setup is illustrated in Fig. 1(a).

We performed degenerate FWM, spectral range λ = 1140 to 1500 nm, with the pump beam at normal incidence and the probe beam at a small angle (~6°). Both beams are vertically polarised. In contrast to typical FWM experiments, we observe both PC and NR signals using only a single pump beam. Therefore, the only possibility is that the sub-wavelength phase-matching condition is satisfied with our ENZ film and is, therefore, acting as a time-dependent optical surface. This departs from the standard phase conjugating experiment configuration in bulk materials, where the conservation of total momentum is naturally obtained with two counter-propagating pump beams.

In Fig. 1(b) we report the PC and NR as a function of the delay between pump and idler. As can be seen from the figure, the two processes are simultaneous and nearly instantaneous (FWHM is ~ 110 fs, very close to the pulse duration), as expected from a FWM interaction.

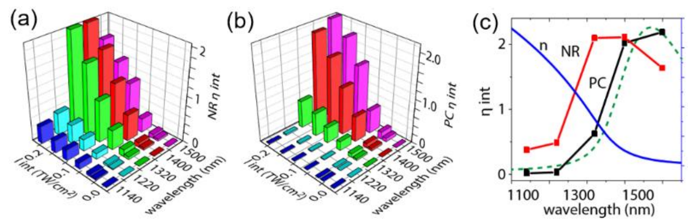

Fig. 2. (a,b) Internal conversion efficiency η_{int} of NR and PC as function of the pump intensity inside the sample I_{int} and wavelength. (c) Efficiency of NR and PC as function of wavelength at fixed $I_{int} = 0.22\ (TW/cm^2)$. The green dashed curve shows the internal efficiency predicted from Eq. 1 with parameters used in the experiments. The refractive index *n* measured by ellipsometry is plotted as a solid blue line.

We investigate the efficiency of the two nonlinear processes for different wavelengths, normalized with to respect pump intensities inside the material (Fig. 2a and 2b). The efficiency of the time reversed PC beam goes from 2% at 1140nm to approximately 200% at 1500 nm at $I_{int} = 0.22\ (TW/cm^2)$. At the same pump power, the NR efficiency shows the same trend with a maximum value at 1400 nm (~210%). As shown in Fig. 2(c), the efficiency greatly increases above the ENZ point, around 1350 nm, for both NR and PC.

The near-zero index of refraction (blue line in Fig.2(c)) plays two important roles on the high efficiency of the two processes. First, it guarantees the sub-wavelength film phase-match conditions, since the effective wavelength inside the material is $\lambda_{eff} = \lambda_0/n$. Second, it provides an enhancement of the nonlinearity. Following Ref. 6, the formula for the internal efficiency, valid in the case of perfect phase-match and for low gains and/or short propagation lengths L, in absence of losses can be written as,

$$\eta_{int} = {P_{out}}/{P_{in}} \cong \frac{9}{4} I_{int}^2 L^2 \frac{\omega^2 |\chi_3|^2}{n^4 \varepsilon_0^2 c^4} \qquad (1)$$

It is possible to see from the eq. 1 that the efficiency of the process scales with $1/n^4$ and will be strongly enhanced in the ENZ region. The green dashed line in Fig 2(c) is the theoretical prediction of the efficiency of the process using a $\chi_3 = 1.3\ 10^{-19}\, m^2/V^2$ (as found in Ref. 4), which is in good agreement with experimental results. From an application point-of-view, it is more useful to evaluate the efficiency with respect to the incident pump intensity. In this case, the PC beam exhibits an external efficiency of 34%, which is still two order-of-magnitude greater than Ref. 2 for the time reversed media.

In conclusion, we have demonstrated that an optically thick film of AZO behaves like an ideal time-dependent surface in the ENZ and low-index spectral region. Due to the enhancement of the nonlinearity and the high damage thresholds, this near-zero refractive index material realizes a time reversing surface, which emits both a phase conjugate and a negative refraction signal with a high efficiency (200%). This high efficiency obtained with homogeneous ENZ material presents an advantage for nanoscale device and could pave the way for exciting applications to obtain subwavelength resolution.

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