Ultrafast Optical Tuning of Epsilon-Near-Zero Thin Films

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Abstract: Large and ultra-fast transient reflectivity and transmissivity are recorded via pump (λ=325nm) and probe (λ=1300nm) experiments on aluminum-doped zinc oxide thin films engineered to possess ultra-fast electron-hole recombination and epsilon-near-zero behavior in the NIR.

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1. Introduction

Epsilon-Near-Zero (ENZ) materials are classified by a permittivity which is approximately zero, and for materials with Drude-Lorenz dispersion, the ENZ regime will occur near the bulk plasmon frequency. This phenomenon is well known; however, in the last few years, the study of ENZ materials has generated a prolific number of applications including electromagnetic wavefront engineering, supercoupling, and subwavelength confinement [1]. Consequently, large efforts are underway to synthesize or engineer new materials with ENZ properties occurring across all of the electromagnetic spectrum, particularly in the near-infrared (NIR) and visible. One such class of naturally occurring ENZ materials are transparent conducting oxides (TCOs). With an electron density that can be set as high as 10^{21} cm^{-3} and low electron effective mass, TCOs are characterized by a bulk plasma frequency and hence an ENZ regime at NIR wavelengths [2].
Here we report on the ultrafast modulation of aluminum-doped zinc oxide's (AZO) optical properties at ENZ frequencies using pump-probe spectroscopy. We find that our films exhibit sub-picosecond recombination times and modulation amplitudes exceeding 30% for pump energies above AZO's bandgap and well below the damage threshold, indicating that the induced refractive index change is dominated by photo-induced conduction electrons. Our numerical models allow us to extract the induced carrier concentration and recombination times for our AZO thin films.

2. Results

Our AZO films were deposited by pulsed laser deposition (PVD Products Inc.) using a 2wt% doped AZO target from the Kurt J. Lesker Corp. with a purity of 99.99% or higher. Such a growth technique allows for tuning of the film's plasma frequency not only through aluminum doping, but also through oxygen pressure: low oxygen concentrations during deposition promote the formation of oxygen vacancies which contribute additional free-carrier to the conduction band and allow for sub-picosecond recombination times [3]. Post annealing in an oxygen environment can also modify the concentration of these oxygen-defects. We can therefore fabricate AZO films with a wide range of NIR plasma frequencies, making AZO a versatile material for ENZ applications. Using a pump-probe technique we investigate the temporal response of highly doped as-grown AZO films. To study the electron interband recombination, we set the pump and probe wavelength to be 325nm and 1300nm, respectively. Ellipsometry and linear optical characterization were used to retrieve both the bandgap and the complex dielectric permittivity of our thin films; we find the bandgap to be at 350nm and the ENZ wavelength range (i.e. $|\varepsilon| < 0$) to be 1100-1500nm [3]. The normalized transient reflectivity and transmissivity of our 350nm-thick AZO films as a function of the pump-probe pulse delay are shown in Figure 1-a and 1-b, respectively, for several incident pump fluence levels. A numerical model was created to fit the experimental results (Fig. 2-a and 2-b) allowing the extraction of physical parameters such as the excess carrier density and recombination time. For the highest fluence of 3.9 mJ/cm^2 an average carrier density of 0.7×10^{20} cm^{-3} was found along with a recombination time of 88 fs. This corresponds to a shift in the refractive index of $\Delta n = -0.17+i0.25$ [3]. Similar ultrafast recombination times are reported in studies of low-temperature grown GaAs films, where high densities of mid-bandgap defect states open additional carrier recombination pathways [4]; however, our materials show a dynamic change in their optical properties which is orders of magnitude higher.

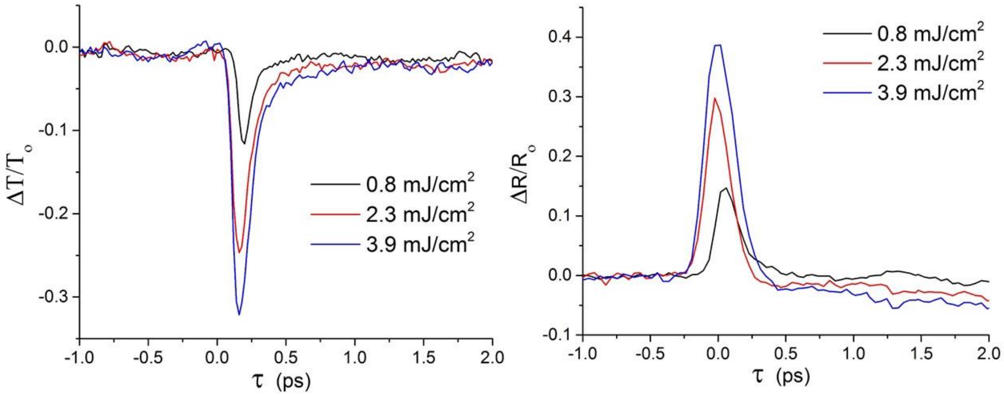

Fig. 1 a) Normalized change in the transmission and b) reflection power for a pump wavelength of 325nm and probe wavelength of 1300nm.

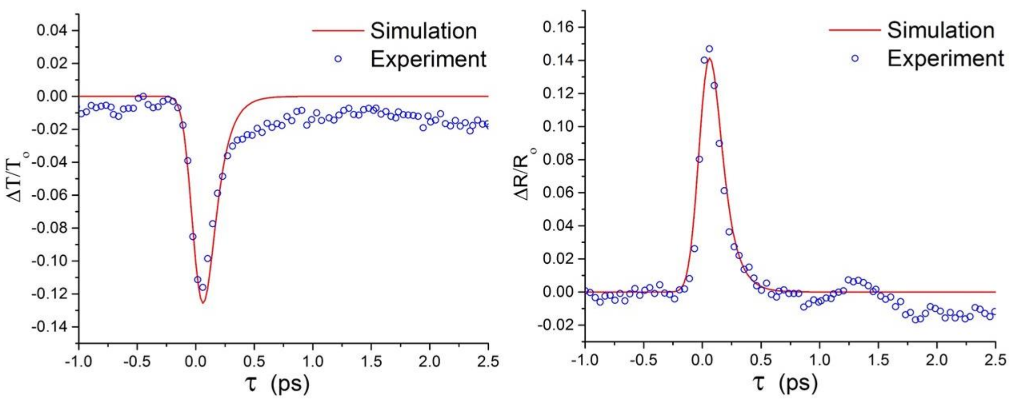

Fig. 2 Numerical fits for normalized a) transmission and b) reflection for the 0.8 mJ/cm^2 pump fluence.

3. Conclusion

In this work we demonstrate ultrafast tuning of engineered ENZ aluminum-doped zinc oxide thin films. Based on our analysis for above-bandgap pumping (325nm), we extracted a recombination rate of 88fs and an induced carrier concentration of 0.7×10^{20} cm^{-3}. The strong and ultrafast modulation that can be obtained across a broad range of excitation frequencies opens up new avenues towards ultra-fast tunable nano-photonic devices based on active AZO.

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