Designing thin-film metamaterials by tuning effective ENZ behavior

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***Abstract*—Using an effective refractive index calculation, we demonstrate the ability to efficiently tune epsilon-near-zero (ENZ) behavior within compact thin-film stacks. Results show the desired effect of broadening the reflectivity profile and improving absorption for an all-oxide ENZ metamaterial.**

Keywords—plasmonic, metamaterials, near-zero index, effective index, thin film, perfect absorption, ENZ tuning

I. INTRODUCTION

Materials with a permittivity (ENZ) or refractive index near zero (NZI) are widely sought after given their ability to impedance match structures radiatively with free space. This characteristic enables metamaterials inclusive of ENZ or NZI media to exhibit extremely high absorption in otherwise radiatively lossy materials [1]. They are also able to induce slow or stopped light, which can lead to field enhancements that in turn yield strong nonlinear responses such as soliton generation. Plasmonic modes excited within the light cone such as Ferrell-Berreman (FB) or ENZ modes have even demonstrated perfect antireflection (PAR) and absorption (PA) [2]. These modes frequently display narrow absorption bands ranging over a few to tens of nanometers. Useful if one wishes to apply such technology to optical or magnetic sensing devices. Recently, efforts have been focused on tuning ENZ resonances by adjusting the fabrication method, i.e. optimizing the annealing process or carrier doping [3]. By focusing on design rather than fabrication methods, we aim to construct compact and robust metamaterials that can tune and broaden the reflectivity and absorption profiles such that ultra-low reflection (< 1%) may be achieved over the widest range of wavelengths possible. Such designs can significantly help to maximize the efficiency within energy conversion devices. We show an efficient method of doing this by using an established approximation to the refractive index of composite structures that convey an effective material response [4-6]. We apply the effective index calculation towards developing an all-oxide metamaterial and show that it is able to predict tuning to ENZ regions, and therefore the changes seen to reflectivity and absorption.

II. METHODS

ENZ metamaterials are frequently formed from symmetrically layered films in which the effective material responses are dependent on the number of layers deposited. In order to maintain compactness of many devices, our study includes asymmetrically layered materials and their ability to induce ENZ behavior. Of note, the terminology 'NZI' is typically reserved for materials exhibiting both, near-zero permeability and permittivity. The investigation herein focuses on non-magnetic materials and therefore the convention 'ENZ' is used to describe the low index behavior.

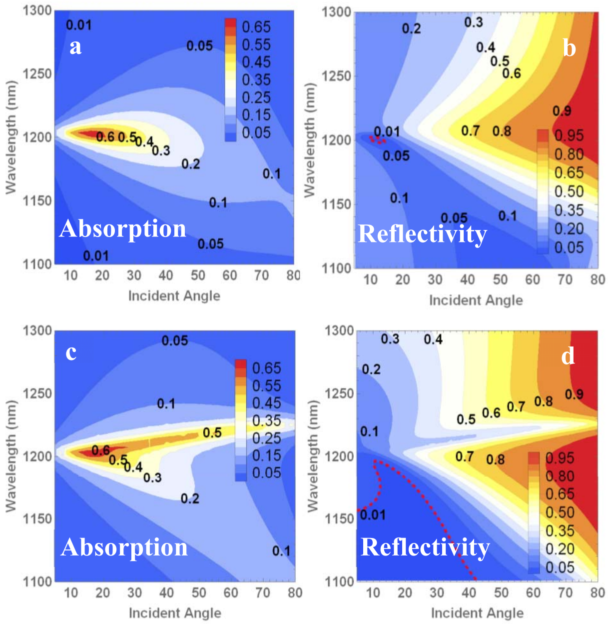

Fig 1: Absorption (left) and reflectivity (right) profiles for a bilayer structure of (a-b) 100 nm ITO on 150 nm SiO_2 and a 3-layer system of (c-d) 100 nm ITO - 50 nm SiO_2 -100 nm ITO. Contours of 0.01 are highlighted (red-dashed line, ×100 = %).

We begin by taking advantage of materials known to have ENZ resonances in the near infrared such as Indium Tin Oxide (ITO). The transfer matrix of the system is then calculated which yields the reflectivity and transmission coefficients [2]. The absorption and reflectivity profiles of Fig. 1 are extracted from this analysis that is quantified in (1). The electric field is represented by TM-polarized light where the + (-) sign represents the transmitted (reflected) wave at the boundary interface of layers *i* and *i+1* with thicknesses *d*. The transfer matrix ($M_{i,i+1}$) between successive layers is defined by the 2×2 matrix whose elements consist of the reflectivity (*r*) and transmission (*t*) coefficients.

$$\begin{pmatrix} E_i^+ e^{ik_{z_i}d_i} \\ E_i^- e^{-ik_{z_i}d_i} \end{pmatrix} = \begin{pmatrix} 1/t_{i,i+1} & r_{i,i+1}/t_{i,i+1} \\ r_{i,i+1}/t_{i,i+1} & 1/t_{i,i+1} \end{pmatrix} \begin{pmatrix} E_{i+1}^+ e^{ik_{z_{i+1}}d_i} \\ E_{i+1}^- e^{-ik_{z_{i+1}}d_i} \end{pmatrix} \tag{1}$$

The transfer matrix of the composite structure is given by $\mathrm{T} = \prod_i \mathrm{M}_{i,i+1}$. The wavevector normal to the incident plane is defined as $k_{z_i} = \sqrt{\varepsilon_i \omega^2/c^2 - k_x^2}$, with $k_x = k_{in}\sqrt{\varepsilon_1}\sin\varphi_1$ conserved at each boundary. The permittivity of ITO is defined within the Drude free-electron model, $\varepsilon_{ITO} = \varepsilon_\infty - \omega_p^2/(\omega^2 + i\omega\gamma)$, with values for the plasma frequency (ω_p =3.13 rad/fs), scattering rate (γ=1.07×10^{-2} rad/fs), and background permittivity (ε_∞ = 4.0). The reflectivity profiles are equivalent to the ratio of reflected and incident field amplitudes at the superstrate-metamaterial

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interface $|E_1^-/E_1^+|$. The absorption data is given by $\alpha = 1 - R_{k,\Phi} - T_{k,\Phi}$ where $R_{k,\Phi}$ and $T_{k,\Phi}$ refer to the spectral and directional reflectivity and transmission powers.

Fig. 1 shows the absorption and reflectivity profiles for two cases including ITO on SiO_2 (bilayer) structure and a 3-layer design comprised of two ITO layers separated by a thin-film of SiO_2. The super/substrate is considered to have a refractive index of free space. Determination of the material set and film size was aided by an effective index calculation that makes use of the coefficients derived from the transfer matrix analysis. This method had previously been applied to non-continuous metamaterials under normal incidence such as u-shaped resonators as well as periodically layered designs [6]. In this work, we successfully extend its treatment to nanoparticle arrays and asymmetrically layered media at various angles of incidence. We find it is able to approximate the tuning of ENZ resonances in the layered metamaterials and accurately predict changes seen in the absorption and reflectivity profiles. Results shown in Fig. 2 correspond to the real and imaginary parts of the effective index calculated from (2) that was originally derived in [4] assuming a material of finite thickness and arbitrary material response, $n_{eff}^2 = \varepsilon_r \mu_r$.

$$\varepsilon_r \mu_r = -\{\ln(T_{22})/kd\}^2. \tag{2}$$

Here ε_r and μ_r refer to the relative permittivity and permeability of the composite metamaterial with d corresponding to the full thickness of layered thin films. The transfer matrix element T_{22} corresponds to the inverse of the transmission coefficient. Dealing with passive materials only, we take the positive root of the imaginary part of the effective refractive index. Fig. 2a and 2b show the real and imaginary parts, respectively, of the effective refractive index components for the ITO-SiO_2 bilayer structure whose absorption and reflectivity profiles are shown in Fig. 1a and 1b. Contours of 0.01 are highlighted for reference. The results agree well with a known ENZ and FB resonance circa 1200 nm for ITO using roughly bulk plasma frequencies in the Drude model [1,2]. The resonance overlays with values of ultra-low reflection (< 1%) and increased absorption for incident angles under 30 degrees while favoring reduced wavelengths. It is important to note that ENZ behavior requires both the real and imaginary parts of the refractive index to be well below 1.0, and frequently below 0.1. Additionally, this behavior forecasts the relatively high reflection closer to normal incidence.

The ultra-low reflection demonstrated in the bilayer was found to exist over a small range of wavelengths (≈ 10 nm) and incident angles. By analyzing the effective index of a few structures, we were able to infer an all-oxide 3-layer design that is able to significantly broaden the regions of reduced reflectivity while increasing absorption. This is done by examining the red/blue shifting of the effective ENZ behavior. Fig. 2c and 2d correspond to the real and imaginary parts, respectively, for the 3-layer structure. One can see a noticeable reduction in the imaginary component of the effective index for the 3-layer structure in Fig. 2d, compared to that of the bilayer in Fig. 2b, in particular for shorter wavelengths. A lesser change occurs in the real components which remain close to 0.1 in this region. The reduction of the imaginary component is enough to reduce reflectivity from roughly 5-10% to below 1% going from the bilayer structure to the 3-layer structure in the region stretching from 1100 - 1200 nm in Fig. 1. Regions of PAR or PA are a function of layer thickness and improvements in our results may be seen if one adjusts the film thicknesses accordingly depending on the preferred input wavelength and incidence angle [1,2].

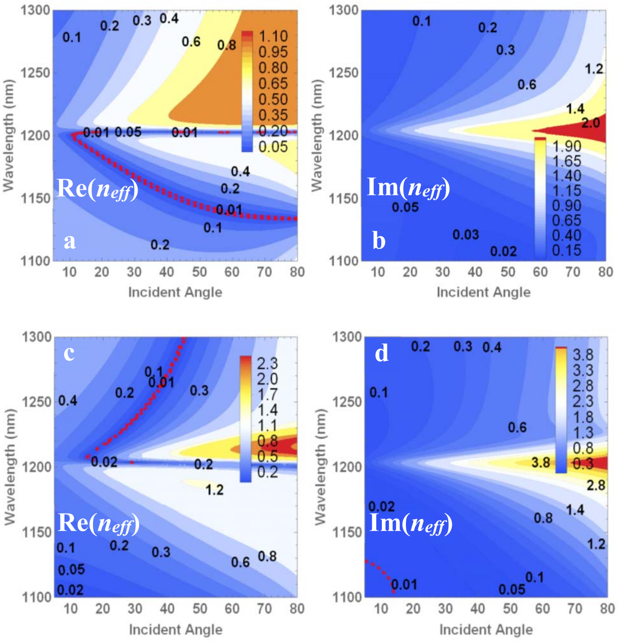

Fig. 2 The absolute value of the real and imaginary components of the effective refractive index approximated from (2). Fig. 2a and 2b correspond to a single layer of ITO on SiO_2 while 2c and 2d correspond to two layers of ITO interspaced with SiO_2.

To conclude, we utilize an effective index method to accurately predict the changes to reflectivity and absorption profiles by examining the ENZ tuning behavior in a number of metamaterial designs. The applied method is demonstrated to work on structures that may be asymmetric and whose resonances are directionally dependent on incidence light. Furthermore, beyond all-oxide structures, the method has revealed asymmetric 3-layer designs of Ag nanoparticle/ITO films for additional tuning of antireflective behavior towards the visible light regime. Altogether, these compact, non-periodic metamaterials show potential as sources for slow light with improved energy efficiency.

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