Field Enhancement of Epsilon-near-Zero Modes in Atomic-Layer-Deposited ZnO:Al Nanolayers

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***Abstract*—We derive an expression for electric-field intensity enhancement due to the epsilon-near-zero modes in ultra-thin conducting layers. We show that absorptance and field enhancement in aluminum-doped zinc oxide nanolayers grown by atomic layer deposition are controlled by nanolayer thickness and optical losses.**

Keywords—plasmonics, atomic layer deposition, epsilon-near-zero, nanophotonics

I. Introduction

Electric-field intensity enhancement (FIE) is one of the key factors enabling successful applications of plasmonic and nanophotonic devices in surface-enhanced spectroscopies, tailoring radiation patterns, enhancing light emission and nonlinear optics. Besides the traditional plasmonic materials, conductive materials with electric permittivity or ε (epsilon)-near-zero (ENZ) and metamaterials show promise in the field enhancement in subwavelength dimensions [1, 2]. In this work we discuss FIE in a series of Al-doped zinc oxide (AZO) nanolayers of varying thickness with ENZ wavelengths between 1540 and 1600 nm. We show how the FIE can be straightforwardly obtained from the measured absorptance of the AZO nanolayers and show FIE dependence on nanolayer thickness and optical losses.

II. Resilts and Discussions

A. Atomic Layer Deposition of AZO Nanolayers

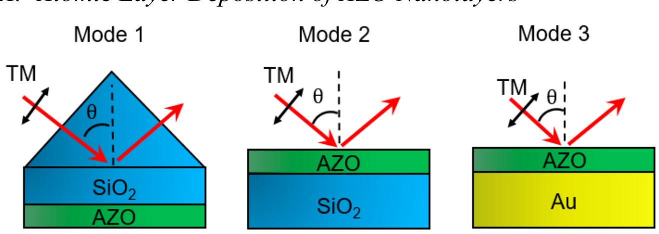

Fig. 1. Schematics of three excitation configurations of TM-polarized ENZ modes.

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The AZO nanolayers of thicknesses between 20 nm and 160 nm were grown by atomic layer deposition (ALD) on silica substrates [3]. The optical properties in a range of wavelengths between 400 and 1700 nm as well as the thickness of the nanolayers were obtained through the use of spectroscopic ellipsometry. The thickness was verified with complementary profilometry measurements and the measurements of AZO nanolayers grown on silicon substrates under the same deposition conditions.

B. Absorptance Measurements

Ultra-thin ENZ films support plasmon-polariton modes [4]. Here, we discuss three different ENZ modes excited in three different configurations shown schematically in Fig. 1. Excitation of ENZ modes lead to an enhanced local density of optical states and, at certain conditions, to the perfect absorption [5]. The enhanced absorptance due to the excitation of ENZ mode 1 in a series of AZO nanolayers was confirmed by measuring their specular reflectivity spectra in the Kretschmann configuration. Figure 2a shows ratios of TM and TE reflectivities measured from the AZO nanolayers at an angle of incidence of 42.3°. Using the optical properties of the AZO nanolayers obtained from ellipsometry measurements, we calculated absorptance spectra as a function of the angle of incidence. We observe a good agreement between measured (light color curves) and calculated (dark color curves) results (Fig. 2a). The maximum absorptance achievable in AZO nanolayers due to the ENZ modes is shown in Fig. 2b. For very thin ENZ films the perfect absorption is observed when the thickness satisfies the critical coupling conditions [5]:

$$\frac{2\pi t}{\lambda} \sim \frac{\varepsilon''}{n_i^3} \frac{1}{\sin(\theta)\tan(\theta)} \; (|\varepsilon'| \ll \varepsilon''). \qquad (1)$$

Here t is the nanolayer thickness, λ - the wavelength, ε' and ε'' - the real and imaginary part of the AZO permittivity, n_i is the refractive index of the incidence medium, θ is the angle of incidence. It has to be noticed that the angle θ and wavelength λ in (1) are coupled by the dispersion relationship of the ENZ mode [5]. Because AZO nanolayers have optical losses of $\varepsilon'' \sim 0.9$ at ENZ wavelength, the perfect absorption is achievable in AZO nanolayers with the thickness $t > 78$ nm (or 30 macro cycles) in the mode 1 configuration (Fig. 1).

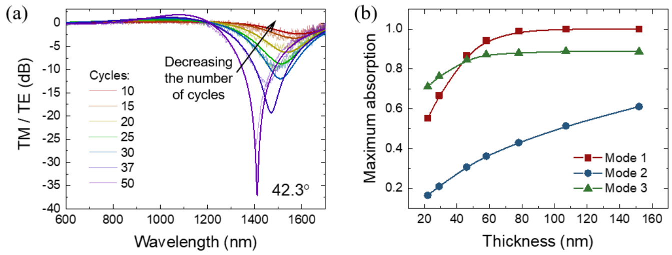

Fig. 2. (a) An example of the measured (light color) and simulated (dark color) ratios of the TM- and TE-polarized reflectivities in the Kretschmann configuration (mode 1, see Fig. 1) for a series of the AZO nanolayers with increasing number of deposition cycles (thicknesses) at a fixed angle of incidence of 42.3°.The dip in the reflectivity ratio is due to the excitation of the ENZ polariton mode. Perfect absorption is observed in the thickest AZO nanolayer (50 cycles). (b) The maximum absorptance calculated from the ellipsometric and reflectivity measurements as a function of the AZO nanolayer thickness for the three modes.

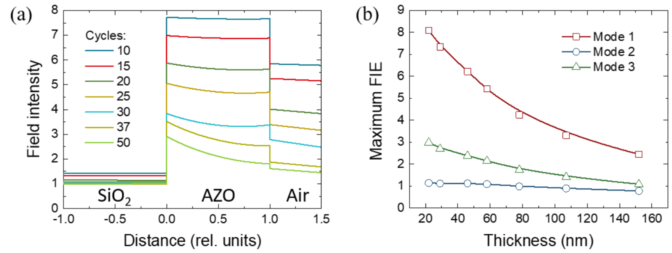

Fig. 3. (a) Transfer matrix calculated TM-component of electric-field intensity distribution normal to the nanolayer surface for the ENZ mode 1. The field intensity calculated at the maximum absorptance (Fig. 2b). It increases as the number of the deposition cycles (nanolayer thickness) decreases. (b) The thickness dependence of the maximum FIE calculated from (2) for the three ENZ modes.

C. *Field Intensity Enhancement*

ENZ modes are characterized by enhanced electric filed inside the AZO nanolayers. FIE can be calculated from the absorptance using the following equation:

$$FIE = \left|\frac{E}{E_i}\right|^2 = \frac{A(\theta,\lambda)\cdot n_i \cdot \cos(\theta)}{\varepsilon''} \frac{\lambda}{2\pi t}. \qquad (2)$$

Here E is the average field inside the AZO nanolayer, E_i is the incident field, $A(\theta, \lambda)$ is the absorptance. Figure 3a shows TM-component of electric-field intensity distribution normal to the nanolayer surface for the ENZ mode 1, which is calculated by the transfer matrix method. The field intensity profiles are calculated at the maximum absorptance (Fig. 2b). The field inside the AZO nanolayer increases as the number of the deposition cycles (nanolayer thickness) decreases. Figure 3b shows the maximum FIE achievable in the AZO nanolayers as a function of the AZO nanolayer thickness, which is obtained from (2). Contrary to the published reports [2, 4], FIE in real ENZ materials with unavoidable optical losses increases weaker than $1 / t^2$ as the nanolayer thickness decreases (Fig. 3b). The weak increase of FIE as the AZO thickness decreases is attributed to a considerable imaginary part of permittivity at the ENZ wavelength, which changes slightly with the AZO thickness. The FIE for the radiative modes 2 and 3 is smaller than for the bound mode 1 because of low refractive index of the incidence medium and low absorptance. It is noteworthy that FIE for the mode 1 has the maximum at the critical angle.

III. Conclusions

We investigated field intensity enhancement of ENZ modes in the ultra-thin ZnO:Al layers grown by ALD and derived a simple analytical expression to calculate FIE from the measured absorptance. We showed that the dependence between FIE and ENZ nanolayer thickness is weaker than predicted in the literature due to inevitable optical losses of materials. Our study is useful to understand and enhance light-matter interaction in ENZ media such as enhanced quantum light emission and ENZ nonlinearity.

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