PLASMONIC AG-LINBO$_3$ METAMATERIAL: A STUDY OF DIELECTRIC AND OPTICAL PROPERTIES

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Abstract

We report the optical and dielectric properties of a multilayer system constituted by ITO, an square-arrangement of Ag nanoparticles (NPs) on the uniaxial $LiNbO_3$, the polymer PMMA and the substrate SiO_2. We investigate the compositional and geometrical parameters dependences on the optical absorption of the stacked layers. The effective dielectric function of the ensemble aggregates of Ag NPs and uniaxial $LiNbO_3$ crystal is treated through the extended Maxwell-Garnett approximation. The multilayer absorbance is determined by means of the Transfer Matrix Method. We demonstrate that an adequate choice of structural parameters such as NPs sizes, interparticle distances and layers thicknesses can produce an enhancement of optical absorption in the multilayer. In addition, we evaluate the dielectric modulus parameter of the effective medium layer in the perpendicular and parallel directions of the Ag NPs plane. For both directions, when the filling factor is big enough, we find the negative epsilon (NE) condition, typical characteristic observed in metamaterials.

Keywords: Optical properties, Dielectric properties, Multilayer with Ag NPs on $LiNbO_3$; Negative epsilon condition

1. INTRODUCTION

Noble metal nanostructures exhibit the capability to couple light with the collective oscillations of their conduction-band electrons, known as surface plasmon resonance (SPR), which can result into a strong confinement of the electromagnetic field in the vecinity of the metallic physical boundaries. This characteristic can be used in many applications such as light-emitting systems, biosensors or solar cells [1]. Sun et al. [2] reported the photodetector ability of $LiNbO_3$ on insulator with encapsulated plasmonic nanoparticles and doped silver ions. On the other hand, we previously investigated the anisotropy dielectric tensor of the rectangular arrangement of Ag nanoparticles (NPs) on $LiNbO_3$ matrix as a composite system [3]. From the array's effective dielectric function, we studied the compositional and geometrical parameters which could satisfy the negative epsilon (NE) condition, typical in metamaterials, but we did not investigate their optical absorbance nor their dielectric properties.

Therefore, in the present paper, the optical absorption of a multilayer with a square array of Ag NPs on $LiNbO_3$ matrix is evaluated by using an extended Maxwell-Garnett (M-G) theory [4] and the Transfer Matrix Method (TMM) [5]. We study the compositional and geometrical parameters which influence the optical absorption of the multilayer. In addition, from the dielectric modulus parameter, we investigate the NE condition and the involved resonances for the Ag NPs arrangement in $LiNbO_3$ crystal in the perpendicular and parallel directions to the Ag NPs plane.

2. INVESTIGATED SYSTEM AND THEORETICAL MODEL

Figure 1 depicts the scheme of the multilayer constituted by (1) a semi-infinite air space; (2) a layer of ITO, transparent in the VIS range, with dielectric function ε_2 and thickness d_2; (3) the effective medium composed by a square arrangement of Ag NPs embedded in $LiNbO_3$, with thickness d_3 and dielectric function described later; the layer (4) of transparent thermoplastic PMMA, with dielectric function ε_4 and thickness d_4; and (5) the substrate of SiO_2 with dielectric function ε_5 and fixed thickness d_5 = 2000 nm. The light impinges from the air with dielectric function $\varepsilon_1 = 1$.

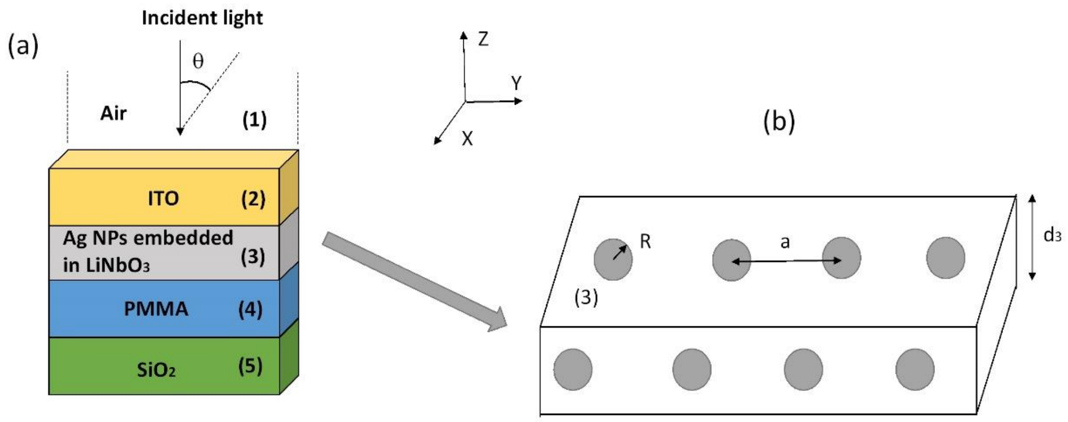

Figure 1 Scheme of the investigated multilayer (figure 1a). Figure 1b is the zoom of the effective medium layer

The dielectric function of ITO is described by Drude model [6], while the refractive indexes of PMMA, SiO_2 and $LiNbO_3$ are taken form reference [7]. The Ag NPs are located at xy plane (square lattice) and they are separated by interparticle distances (a). The array is characterized by spheres with radii R and the interparticle distance (a) is chosen such that R/a = 0.25. Due to the 2D arrangement of Ag NPs, and accounting on interparticle interaction in dipolar approximation; the sum of dipolar electric fields does not cancel out [4]. Accordingly, one expects dissimilar contributions to the local electric field when the electric field is applied perpendicular or parallel to the NPs in-plane. That dissimilar responses results in an anisotropic dielectric function for the layer 3. Following the same treatment than in ref. [3], we deduce for square lattice the corresponding effective dielectric functions: $\varepsilon_3^{\perp}(\omega)$ and $\varepsilon_3^{\parallel}(\omega)$, extensively described in ref. [3].

With the knowledge of the dielectric permitivity of each layer, we use the TMM for our system, similar to that reported in ref. [8]. Using the expressions for the reflectance, R, and transmittance, T, one calculate the absorbance (A) of light in such stacked layers by means of the standard relation: A = 1 – R – T.

Focusing on the dielectric properties of the effective medium layer in the perpendicular and parallel directions, we evaluate the real and imaginary parts of the dielectric modulus, M, defined in ref. [9].

3. RESULTS AND DISCUSSION

To clarify the presentation of our results, we subdivide the discussion in the following two subsections.

3.1 Optical absorbance of the multilayer system

In order to know the influence of the Ag NPs in the absorbance of the investigated system, we show in **Figure 1** the multilayer absorbance with and without Ag NPs at normal incidence with λ = 450 nm. The presence of Ag NPs benefit the excitation of plasmonic modes, which entails an absorption enhancement of 25%. He et al.

[10] proved both experimentally and theoretically the Ag NPs induced surface plasmonic modes centered at 459 nm. On the other hand, Sun et al. [2] showed an enhancement of the absorption in the VIS range with the inclusion of Ag NPs in $LiNbO_3$ crystal. For both cases, with and without Ag NPs, the absorbance values increase for greater wavelength until a saturation value around 0.4 at 1100 nm.

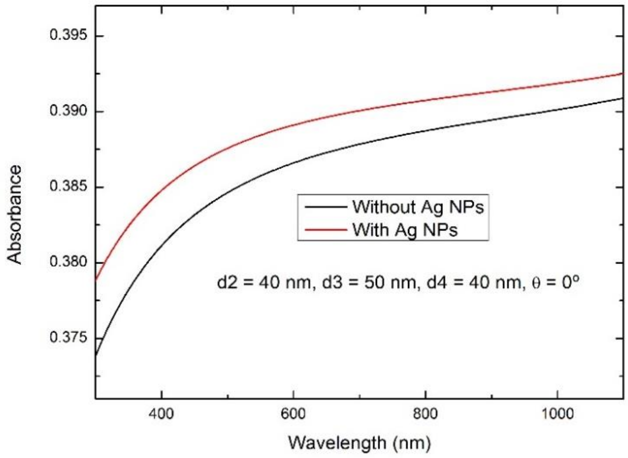

Figure 2 Absorbance of the stacked layers with and without Ag NPs arrangement in the layer 3.

In order to investigate the thicknesses effect on the stacked layers absorbance, we evaluate their dependences modifying the thicknesses of ITO, PMMA and the effective medium layers. **Figure 3a** depicts the stacked layers absorbance for different ITO thicknesses (d_2 = 20 nm, 40 nm and 60 nm), fixing d_3 = 50 nm and d_4 = 40 nm. For higher ITO thickness, we obtain a greater absorbance for λ > 600 nm, while for shorter λ, the greater absorbance relates to the smallest ITO thickness. We identified λ = 700 nm, where the three absorption spectra have a crossing point. We show in **Figure 3b** the absorbance dependence on the PMMA thickness

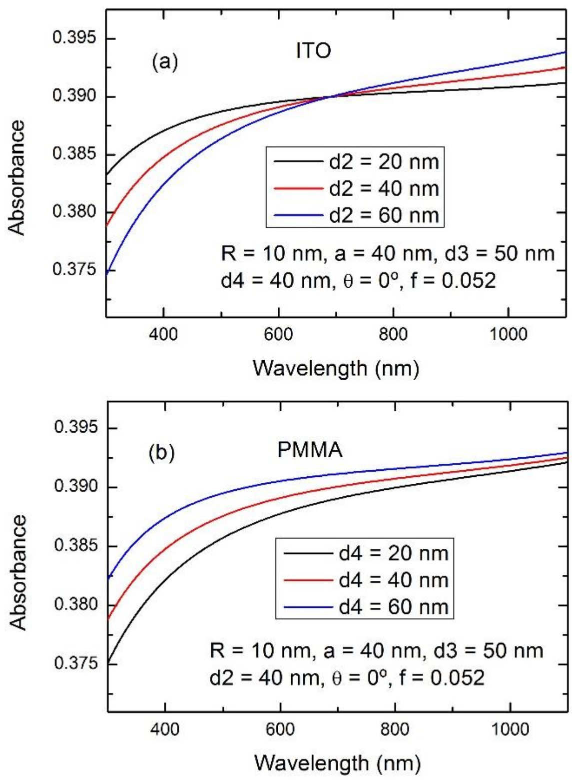

Figure 3 Absorbance of the multilayer for different thicknesses of ITO layer, d_2 (a) and PMMA layer, d_4 (b).

(d_4 = 20 nm, 40 nm and 60 nm), fixing d_2 = 40 nm and d_3 = 50 nm. On the contrary to the dependence of the ITO thickness, we obtain greater absorbance values for PMMA thicker layers in all wavelength range. Although it is not shown here, for greater thickness of PMMA layer (d_4 = 100 nm, 200 nm, 400 nm), the behavior of absorbance with the thickness is analogous to the case of ITO; i.e., there are crossing points in the optical spectra. Attending to the effective medium thcikness dependence, **Figure 4a** shows the absorbance for d_3 = 25 nm, 50 nm and 75 nm with fixed R = 10 nm, a = 40 nm, d_2 = 40 nm and d_4 = 40 nm. A different d_3 thickness imply a different filling factor, f. For higher f, the absorbance values increase and then, a greater concentration of Ag NPs entails a greater optical absorption of the multilayer. We observe a saturation value around 0.4 for λ > 800 nm. In addition, we investigate the Ag NPs geometrical parameter size effects on the multilayer absorbance. **Figure 4b** depits the absorbance for radii R = 10 nm, 20 nm and 40 nm, where R/a = 0.25, fixing d_3 = 100 nm, d_2 = 40 nm and d_4 = 40 nm. We obtain greater absorbance values for higher NPs density (f = 0.105). We previously discussed that higher concentration of Ag NPs benefit the absorption of the multilayer. The effective medium (layer 3) constitutes the active layer in this stacked system.

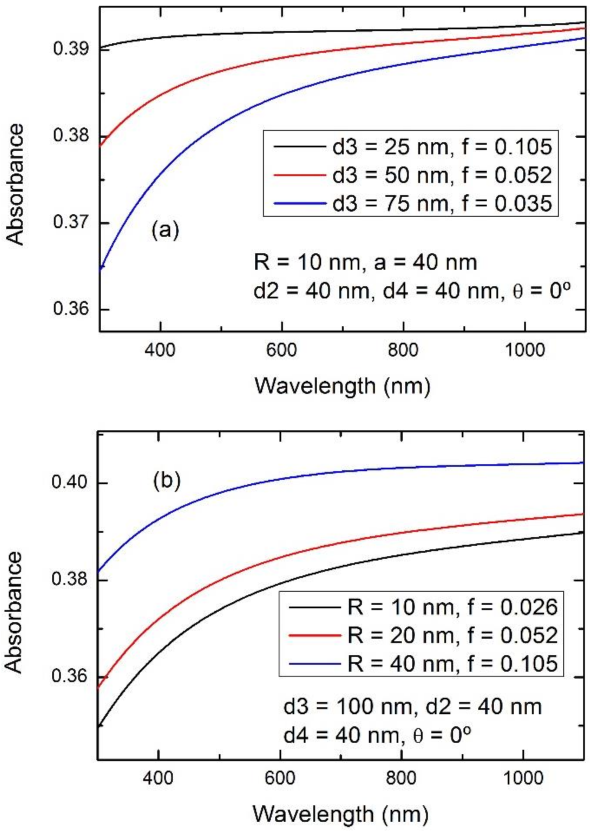

Figure 4 Absorbance of the multilayer for different thickness of effective medium layer, d_3 (a) and for different radii and interparticle distance of Ag NPs lattice (b), which implies different filling factor, f.

In summary, we identify a direct relationship between absorbance intensity and filling factor values.

3.2 Dielectric properties of the active layer

Attending to the dielectric properties of the effective medium layer, we show in **Figure 5** the dielectric modulus (real and imaginary parts) in the parallel direction of Ag NPs plane (xy plane) with different filling factors. For both real and imaginary parts, we observe a resonance around 5.6x10^{14} Hz, which is blue shifted for increasing f. For f = 0.052, M‘ becomes negative in a frequency interval centered at 5.6x10^{14} Hz; then, the NE condition (ε_{eff}‘ < 0) is obtained for specific values of radii or filling factor. This feature was previously reported in refs. [3,9]. This kind of negative dielectric behavior is observed in metameterials due to the plasmonic resonance as suggested by Drude model [11]. For NE condition, the displacement of carriers is opposite to the direction of electric field, **E**. Consequently, the **D** vector is aligned opposite to **E**. In fact, this produces the negative polarization leading to the negative epsilon or dielectric constant value.

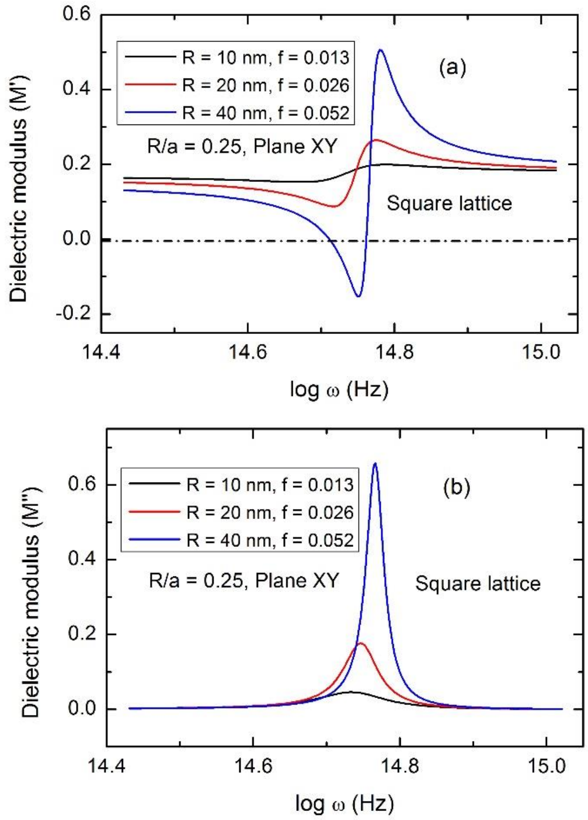

Figure 5 Real- (a) and imaginary- (b) parts of the dielectric modulus dependences on log ω in the xy plane direction. The horizontal dashed line in fig 5a remarks the NE condition.

Although it is not shown here, we obtain a resonance around 7.9×10^{14} Hz for the dielectric modulus (real and imaginary parts) in the perpendicular direction of the Ag NPs plane (z axis). For f = 0.052, the NE condition is reached in a frequency interval centered at 7.9×10^{14} Hz.

In summary, the NE condition, typical in metamaterials, is obtained in the ordinary as well as in extraordinary directions of the Ag NPs plane for filling factor big enough (f = 0.052).

4. CONCLUSION

We have evaluated the optical absorption of a multilayer system composed by ITO, a square-lattice of Ag NPs in uniaxial $LiNbO_3$ crystal, PMMA and a substrate of SiO_2. We investigated the geometrical parameters dependences on the multilayer optical absorption. The stacked layers absorption is simulated by TMM using an extended M-G model to define the dielectric function of the effective medium. We observed that an adequate choice of geometrical parameters such as NPs sizes and layers thicknesses can produces an enhancement of optical absorption of the multilayer. Indeed, when the filling factor is higher, the multilayer absorbance is greater. In addition, we evaluated the dielectric modulus parameter of the effective medium layer (active layer) in the parallel and perpendicular directions of the Ag NPs plane. For both directions, when f = 0.052, we found the NE condition, typical feature observed in metamaterials.

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