COMPOSITE LINBO$_3$-AG NANOPARTICLES DISPERSION WITH COMPENSATED LOSS

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Abstract

The composite constituted by lithium niobate ($LiNbO_3$) with silver (Ag) nanoparticles (NPs) is theoretically proposed as a candidate of a metamaterial with plausible photonics applications. We numerically evaluate the effective dielectric function (ε_{eff}) of this composite by using Maxwell-Garnett (M-G) effective theory. The dielectric function of Ag NPs is described by Drude theory of free electrons, while $LiNbO_3$ dielectric function is evaluated through the Sellmeier equations which describe their ordinary and extraordinary refraction indexes as a function on the incident light wavelength. Once the ε_{eff} is obtained, we investigate the lossless optical condition, where $\varepsilon''_{eff} = 0$ and $\varepsilon'_{eff} < 0$ at some frequency. We obtained that the above condition depends on the volume fraction and sizes of Ag NPs immersed in the $LiNbO_3$ matrix. The extinction of the composite is also dependent on the radius and the volume fraction of Ag NP embedded in lithium niobate.

Keywords: Composite $LiNbO_3$-Ag nanoparticles, optical metamaterial, Sellmeier equations, Drude theory, Maxwell-Garnett effective theory.

1. INTRODUCTION

An extremely large variety of metal/dielectric combinations (types of materials and configurations) called *metamaterials* are currently the subject of intense studies, revealing new fundamental properties and leading to novel devices with improved performances [1, 2]. Indeed, metamaterials are synthetic structures with electromagnetic properties which are not met in naturally occurring materials, such as artificial magnetism and negative refractive index. The later characteristic is of fundamental importance in small-scale optics, which comes to be known as *nanophotonics* [3].

Ferroelectric materials such as lithium tantalate ($LiTaO_3$) or lithium niobate ($LiNbO_3$) has been the subject of a considerable amount of study due to its important applications in several fields such as integrated optical technologies or non-linear optics. Recently, Yraola et al. [4, 5] have proved that Nd^{3+}-doped periodically poled $LiNbO_3$ shows a spontaneous emission and nonlinear response enhancement by Ag NPs inclusion. The above authors claim that this composite could be a plausible metamaterial with a plenty of applications in non-linear optics among others. On the other hand, Yannopapas and Paspalakis [6] have designed a multilayered metamaterial consisting of alternating planes of the ferroelectric lithium tantalate and n-type germanium (Ge) spheres in air. This metamaterial has a negative refractive index and, at the same time, the electromagnetic radiation propagates with a group velocity that is of the order of 10^5 slower than the vacuum one.

The aim of this work is to investigate if $LiNbO_3$-Ag NPs composite really behaves as a metamaterial and fulfills the optical lossless conditions. For this, we evaluate the effective dielectric function of the composite using the Maxwell-Garnett effective theory. Such model is appropriate when the NP´s concentration is small enough so that the composite may be considered as a dilute environment [7]. From the composite´s effective dielectric function, we study the compositional and geometrical parameters which satisfy the metamaterial condition.

2. THEORETICAL MODEL

The system under investigation is a composite constituted by the ferroelectric material $LiNbO_3$, considered as an embedding medium, with inclusions of Ag NPs of nanometric size. To calculate its effective dielectric function (ε_{eff}), we proceed first by defining the Ag NPs and $LiNbO_3$ dielectric functions, respectively. For the Ag NPs, we use the Drude theory of free electrons, neglecting the interband transitions and considering the NP size effect in the plasmon damping term [8]; i.e.,

$$\varepsilon_{Ag} = \left(1 - \frac{\omega_p^2}{\omega^2 + i\,\omega\Gamma}\right) \quad (1)$$

where ω_p and Γ are, respectively, the Ag plasmon frequency (ω_p = 4.60×10^5 cm^{-1}) and its damping dependent sized term defined by $\Gamma = \gamma + A\, v_F/R$, being γ the bulk dumping term (γ = 10^4 cm^{-1}), A = 1, v_F, the Ag Fermi velocity (v_F = 1.39×10^8 cm/s) and R the radius of the Ag NP.

For the biaxial ferroelectric $LiNbO_3$, considered as the embedding medium of the composite and as a first approach, we use for the average dielectric function the relationship $\varepsilon_e = n_{av}^2$, where

$$n_{av} = \frac{2n_0 + n_e}{3} \quad (2)$$

being n_0 and n_e the ordinary and extraordinary refraction indexes, respectively, which are defined by means of the well known Sellmeier equations as functions of the wavelength of the incident light [9]. In this work, we fix the wavelength to a value of 450 nm yielding an n_{av} around 5.5.

Once the dielectric functions of the embedding medium and the Ag NPs are defined, we can use the Maxwell-Garnett effective theory to describe the ε_{eff} of the composite, assuming that the volume fraction of Ag NPs, f, is very small [7]; i.e.,

$$\frac{\varepsilon_{eff} - \varepsilon_e}{\varepsilon_{eff} + 2\varepsilon_e} = f\,\frac{\varepsilon_{Ag} - \varepsilon_e}{\varepsilon_{Ag} + 2\varepsilon_e} = f\,Q. \quad (3)$$

By solving the above equation, we obtain the following composite´s dielectric function, that is;

$$\varepsilon_{eff} = \frac{\varepsilon_e\left(1 + 2\,f\,Q\right)}{1 - f\,Q}. \quad (4)$$

Now, from the real and imaginary parts of ε_{eff}, the extinction coefficient is given by [10]

$$\alpha\left(cm^{-1}\right) = \frac{8.88\times10^7}{\lambda(nm)}\sqrt{-\varepsilon'_{eff} + \sqrt{\left(\varepsilon''_{eff}\right)^2 + \left(\varepsilon'_{eff}\right)^2}}\,. \quad (5)$$

Finally, we obtain the optical metamaterial condition by imposing that $\varepsilon''_{eff} = 0$ and $\varepsilon'_{eff} < 0$ at some frequency. The above requirements are reached when $Q'' = 0$ and $Q' > 1/f$ or $Q' < -1/(2\,f)$, being Q' and Q'' the real and imaginary parts of Q defined previously in eq. (3). Therefore, we can infer that the compensated optical loss condition depends on volume fraction, sizes of Ag NPs and frequency of incident light through the dependence on frequency in the expressions of ε_{Ag} and Q. By developing all the terms in $Q'' = 0$ and $Q'' < -1/(2\,f)$ as a function of the frequency, we obtain eight-order polynomials in frequency whose constant coefficients are related to f, Γ, ω_p and ε_e. Only two solutions with physical meaning are obtained for these polynomials, both fulfilling the second set of conditions, $Q'' = 0$ and $Q' < -1/(2\,f)$. The values of the frequencies range from 9×10^4 to 1.6×10^5 cm^{-1} ≈ 10 - 20 eV. However, with the second set of conditions, that is: $Q'' = 0$ and $Q' > 1/f$, no solution with physical meaning is obtained. To proceed with, we also calculate the Fröhlich frequency, which is obtained by imposing $\varepsilon_{Ag} = -2\,\varepsilon_e$. The expansion of the Fröhlich condition in terms of ω leads to a four-order polynomial with constant coefficients related to Γ, ω_p and ε_e. Only one physical solution, around 16 eV, is obtained for this four-order polynomial.

3. RESULTS AND DISCUSSION

We will show the extinction´s dependences on the volume fraction and size of Ag NPs immersed in the lithium niobate. Extinction is defined as the extinction coefficient multiplied by the NP size. Next, we analyze the optical lossless condition by discussing the criteria that must be satisfied for geometrical and compositional parameters.

Figure 1 shows the composite extinction for three radii of Ag NPs with equal value of f = 0.1. We obtain an asymmetric band peaking nearly at 15.6 eV (R = 10 nm), 15.7 eV (R = 20 nm) and 15.8 eV (R = 40 nm). This band can be ascribed to the Fröhlich frequency, whose value is around 16 eV. It seems that the peak position is slightly size-dependent. A good fitting with two lorentzians are obtained for the three sizes investigated. Also, the extinction´s intensity along with its corresponding Full Width at Height Maximum (FWHM) increase with the radius. We obtain values of FWHM around 2.7 eV (R = 10 nm), 2.8 eV (R = 20 nm) and 3.1 eV (R = 40 nm), respectively.

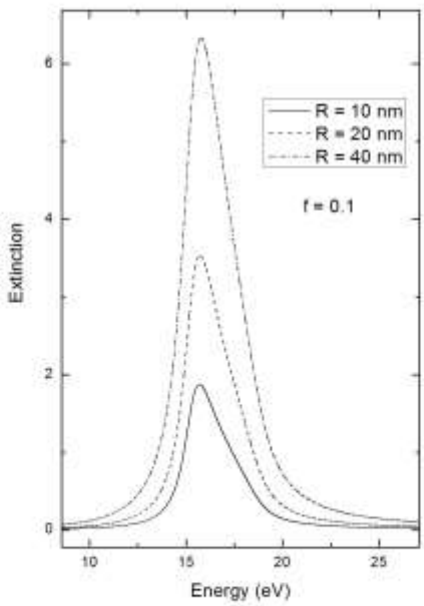

Fig 1. Extinction of the composite as a function of energy for three different radii of Ag NPs and a fixed value of f = 0.1.

In figure 2, we show the composite extinction for three volume fractions with equal radius R = 20 nm. Clearly, the Fröhlich resonances appear at 16.2 eV (f = 0.05), 15.7 eV (f = 0.1) and 14.7 eV (f = 0.2). We observe that the resonance is red-shifted with the increase of the Ag NPs concentration. This phenomenon is similar to that previously reported in semiconductor core-shells for the excitonic peak, which is also red-shifted with the increase of volume fraction [11]. Besides, the intensity and the FWHM increase with the concentration of Ag NPs in the composite. This feature is also previously obtained in semiconductor core-shells [11]. The extinction spectra become asymmetrical, particularly at higher concentration of NPs (f = 0.2).

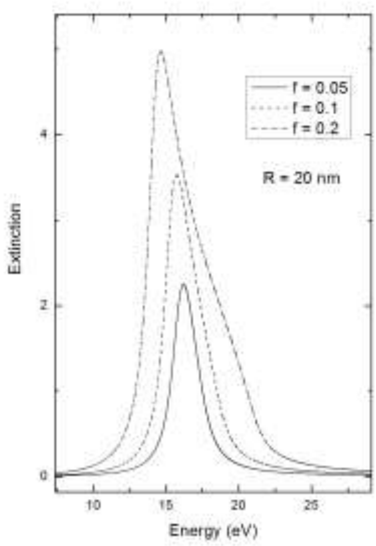

Fig2. Extinction of the composite as a function of energy for three different volume fractions and a fixed value of R = 20 nm.

As an example, we plot in figure 3 a relationship between the frequency and R for f = 0.1 and 0.2, under the condition of optical metamaterial; i.e., when Q´´ = 0 and Q´< -1/(2 f). This frequency is one of the two solutions of the eight-order polynomial with physical meaning. We obtain for both values of f a nearly power-law decay of the frequency´s dependence on the radius. Although it is not shown here, similar frequencies´ dependence is obtained for the other physical solution of the eight-order polynomial.

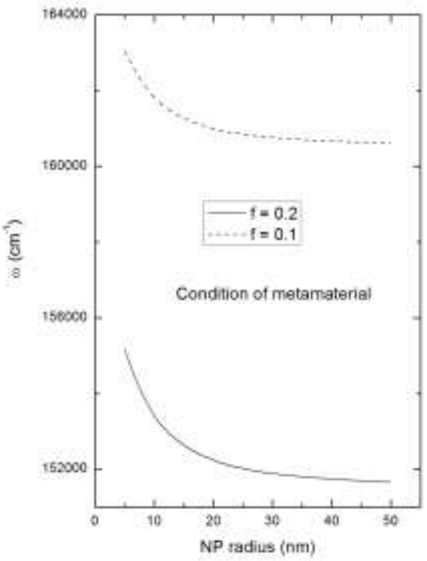

Fig3. Frequency versus Ag NPs radius for f = 0.1 and f = 0.2. This frequency is one of the solutions of the eight-order polynomials which verify the optical lossless condition.

Once the optical compensated loss condition is analyzed, we show in figure 4 the composite dielectric function for R = 20 nm and f = 0.2, where we plot its real and imaginary parts as a function of energy. We clearly observe that around the energy of 20 eV, $\varepsilon_{eff}'' \approx 0$ and $\varepsilon_{eff}' < 0$. On the other hand, at approximately 15 eV, nearly the Fröhlich frequency, ε_{eff}'' has a maximum value, while ε_{eff}' reaches its minimum value, which is negative. Although it is not shown here, we have investigated the optical compensated loss for other combination of parameters; for example: f = 0.2 and R = 10 nm and f = 0.2 and R = 50 nm, where $\varepsilon_{eff}'' \approx 0$ and $\varepsilon_{eff}' < 0$ is verified, obtaining similar behavior for ε_{eff}'' and ε_{eff}', respectively.

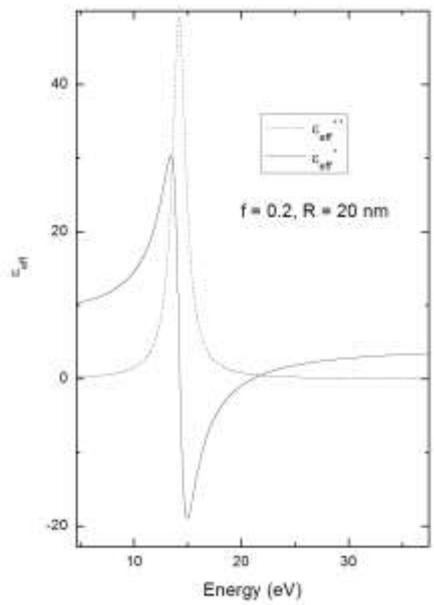

Fig 4. Real and imaginary parts of composite´s dielectric function versus energy for f = 0.2 and R = 20 nm. This parameters yields the metamaterial condition.

4. CONCLUSION

We obtained a plausible candidate of negative dielectric function optical material by including Ag NPs in the biaxial ferroelectric lithium niobate as an embedding material. We evaluated the effective dielectric function of this composite by the M-G effective theory. To describe the Ag NPs´ dielectric function, we use the Drude theory for free electrons and to describe the $LiNbO_3$ matrix one, we use the Sellmeier equations which define their ordinary and extraordinary refraction indexes as a function on the incident light wavelength. The extinction of the composite depends on the radius and volume fraction of Ag NPs embedded in lithium niobate. Finally, we obtained that the optical compensated loss condition also depends on the volume fraction and sizes of Ag NPs immersed in the $LiNbO_3$ matrix. Therefore, this composite could be proposed as an optical metamaterial with photonics applications.

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