ANALYSIS OF THIRD ORDER NONLINEAR SUSCEPTIBILITY OF METAL NANOCOMPOSITE SYSTEMS

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Abstract: We investigate the properties of third order nonlinear susceptibility in copper nanoparticles embedded in $LiNbO_3$ (metal/dielectric composite materials). The dielectric permittivity of the nanocomposite system is calculated by incorporating the temperature dependent plasma frequency of copper nanoparticles. The increase in temperature leads to decrease in the value of susceptibility. These nanostructures can find application in ultrafast photonic switches where light guiding is combined with the optical nonlinearity of third order susceptibility.

Keywords : Nanoparticle, Plasma frequency, Optical & dielectric properties
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Introduction: Photonic crystals are composite materials with periodically varying dielectric permittivity and maximally high localization of light, so that optical integrated circuits might possibly be created. Due to the introduction of Yablonovitch[1] ideas, photonic crystals control the optical properties of materials and those of John[2] concerning the effect of random or periodic variation of the refractive index on the localization of photons. In recent decade the technology is in terahertz range has been intensively developed for scientific and technical applications such as spectroscopy, data transmission, information processing and medical and security scans [3]

Materials with dielectric permittivity at frequencies below their plasma frequencies have a significant impact for the design and fabrication of novel hybrid materials. The linear and nonlinear optical properties of composite materials are determined by Plasmon resonance of metal nanoparticles in transparent matrix. It was shown that optical resonance takes place in transparent matrix with metal nanoparticles. The temperature dependence of the Surface Plasmon Resonance is important in most of the recent applications [4] of noble metal nanoparticles such as thermally assisted magnetic recording, computer chips etc. In this paper, we analyze the temperature dependence of dielectric permittivity by embedding Copper nanoparticles in the silica matrix. The modulation of the dielectric permittivity of the transparent matrix is achieved by incorporating the temperature dependence of the plasma frequency which depends upon the volume expansion coefficient of the nanoparticles. This modulation is also achieved by changing the filling factor and for different radii of the nanoparticle.

Theory: Lithium Niobate is a very important electro-optic material. Indeed it has a very attractive combination of electro-optic, piezoelectric and other optical properties. When the temperature of the crystal is increased the values of the optical parameters are altered [5].

The dielectric constant as a function of temperature can be expressed as [6]

$$\varepsilon_{\infty(T)} = \varepsilon_{\infty}(1+2\alpha_1 T) \quad \text{--- (1)} \qquad \text{and}$$

$$\varepsilon_{0(T)} = \varepsilon_0(1+\beta_1 T) \quad \text{----(2)}$$

where $\varepsilon_{\infty(T)}$ & $\varepsilon_{0(T)}$ are the high frequency dielectric constant and static dielectric constant of the material respectively. $\alpha_1 = \frac{dn_0}{dT}$ and $\beta_1 = \frac{dk_1^T}{k_1^T dT}$ are the temperature coefficients.

The values of α_1 and β_1 are given by α_1 = 3.3 x 10^{-5}/°c and β_1 =3.82 x 10^{-4}/°c[6]. The changes in transverse optical phonon frequency with temperature is given by [6]

$$\omega_{TO(T)} = X(T-T_c)^{1/2} \quad \text{---------(3).}$$

Where X is temperature co-efficient, T_C is Critical Temperature and T is the temperature of the nanocomposite system. The corresponding longitudinal optical phonon frequency at different temperatures can be determined using Lydanne-Sachs-Teller (LST) relation. The dependence of the frequency ω on the wave vector k of an electromagnetic wave in a crystal with a dielectric function $\varepsilon_1(\omega)$ is determined by [7]

$$\varepsilon_1(\omega) = \varepsilon_{\infty(T)} + \frac{(\varepsilon_{0(T)} - \varepsilon_{\infty(T)})\omega_{TO(T)}{}^2}{\omega_{TO(T)}{}^2 - \omega^2}$$

The dielectric constant of the nanoparticles is calculated using the Drude model [8]

$$\varepsilon_m(\omega) = \varepsilon_0 - \frac{\omega_P^2}{\omega(\omega+i\gamma)} \quad (5)$$

where ε_0 is a constant (ε_0 =1 for copper [9]), ω_p is a plasma frequency (ω_p=7.6ev [10]), γ is a damping constant of plasma oscillations. The spherical copper nanoparticles are distributed randomly and homogeneously in transparent matrix.

The volume expansion coefficient depends on temperature according to [11] as

$$\beta(\mathrm{T}) = \frac{192 \in K_b}{r'\alpha(16 \in -7TK_b)^2} \quad (6)$$

where K_b is the Boltzmann constant and α , $\in$, r' are the parameters of Morse potential used to describe the potential of interatomic interaction in copper [11]

The expression for temperature dependent Plasmon frequency is modified as [11]

$$\omega_p(T) = \frac{\omega_{po}}{\sqrt{1+\beta(\mathrm{T})\Delta\mathrm{T}}} \quad (7)$$

Where ω_{p0} = 8.9854 eV at room temperature.

With increase of temperature, the plasma frequency is decreased due to volume expansion coefficient.

Nonlinear susceptibility of Nanocomposites: The optical susceptibility of the composite material is related to the intrinsic NLO third-order susceptibility $\chi_m^{(3)}$ of metal crystal [12]

$$\chi^{(3)} = ff_1^2|f_1^2|\chi_m^{(3)} \quad (8)$$

Where f is the volume fraction of metal nanocrystal, f_1 is the local field effect, and $\varepsilon_1(\omega)$ and ε_m are the dielectric constants of the matrix and metal respectively.

Results and Discussion: The various optical properties like Damping constant (γ), Volume expansion coefficient (β), Susceptibility (χ) and Dielectric constant (ε_A) of the metal nanocomposite system for various temperature are studied. Hence the location and width of the photonic Band Gap are determined. Also, the tuning of PBG with filling factor is analysed.

Variation of Damping Constant (γ) With Temperature: The values of damping constant for various temperature are calculated. Fig. 1 (a) shows the variation of damping constant with temperature for various size of metal nanoparticles. From the graph we found that the values of damping constant increases with temperature irrespective of size of the metal nanoparticles.

Fig. 1 (b) explains the variation of Volume expansion coefficient (β) with temperature. The value of β linearly increases with temperature.

Variation of susceptibility (χ) with Temperature: Fig. 2 explain the variation of susceptibility for various filling factor at 393 K. Radius of the nanoparticles is taken as R = 2 nm. When the concentration of metal nanoparticles increases the value of susceptibility also increases. When the temperature of the system increases the susceptibility χ decreases and the resonance occurs at lower values.

Conclusions: The optical parameters such as dielectric permittivity, volume expansion coefficient, susceptibility, damping constant of the Photonic crystals are influenced by the temperature, size and filling factor of the metal nanoparticles. The metal nanoparticles enhance the value of third order nonlinear susceptibility in the composite medium. This can be used to tune the width and the location of the Photonic band gaps of nanocomposite systems. Thus the new optical properties can be used in manufacturing of novel optical devices.

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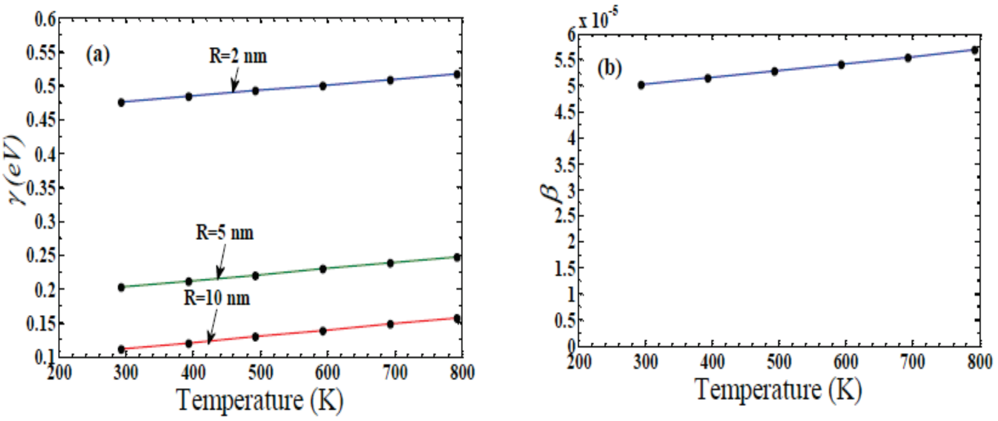

Fig. 1 (a) &1 (b) Variation of damping constant γ and volume expansion coefficient β of nanoparticle with Temperature

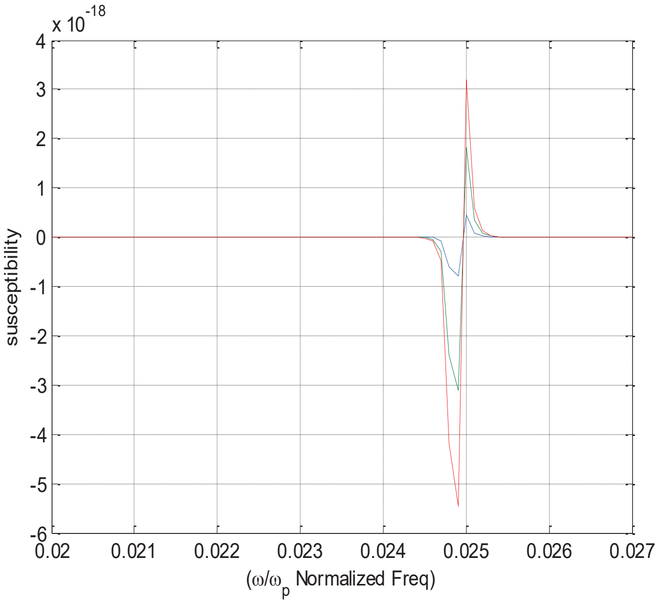

Fig. 3 Variation of susceptibility for various filling factor at 393 K. Radius of the nanoparticles is taken as R = 2nm

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