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Phonon Polariton Modes in Al doped ZnO Nanoparticles

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Abstract: The several optical properties of crystals are modified due to nonlinearity associated with high intensity of the incident radiation. In the present work, the linear and nonlinear optical characterization of Al-ZnO Nanocomposite materials are discussed in detail. We explore the possibilities of nonlinear effects in the optical parameters. New modes on the polaritonic gap where the propagation of electromagnetic wave is forbidden, are obtained due to nonlinearity. The presence of gap mode shows the propagation of electromagnetic radiation in the polaritonic gap which may be exploited in optical communications.

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Introduction

ZnO is a II-VI semiconductor with a wide and direct band gap energy of 3.3 eV. ZnO films have high electrical conductivity when doped with a group III element such as Al, Ga, or In. Since the band gap energy of ZnO is higher than the visible range, ZnO films are electrically conductive and visibly transparent, which leads to applications for transparent conducting oxides (TCO)^{1,2}. TCO thin films are important for the electrode materials in solar cells and display panel applications. Among TCOs, Al-doped zinc oxide (AZO) is a promising candidate to substitute for indium tin oxide (ITO) because of its low cost and wide availability. However, the electrical conductivity of ZnO thin films is much lower than that of ITO. Therefore, many studies have aimed to increase the electrical conductivity of ZnO thin films, through techniques such as post annealing, doping, and plasma treatment^{3,4}. Taking into account the possibility to use such nanocomposites in various applications, a detailed study of their optical properties will be carried out in this paper.

By studying the optical properties of Nanocomposite, one can easily analyse the behavior of polariton modes in the system⁵. Here, using the plasma frequency of Aluminium(ω_p) the dielectric function of the nanoparticles is determined. Then using Maxwell-Garnet approximation the dielectric permittivity of the entire medium having both Aluminium nanoparticles and ZnO medium is determined for various filling factors. Hence, the value of optical phonon frequencies for different filling factors are observed. It is concluded that the changes observed in the optical phonon frequencies are due to the presence of Aluminium nanoparticles in the medium so that, the behavior of the polariton modes in nanocomposite materials is analysed.

Theory

Composite materials with noble metal nanoparticles are very attractive for a development of new optical devices. Nanoparticles are distributed randomly but homogeneously in dielectric matrix. Let us assume that nano-particles have spherical form with radius about some nanometers, i.e. particle size is much smaller than wavelength and penetration depth of electromagnetic wave in metal. Linear and nonlinear optical properties of these materials are determined by Plasmon resonance of metal nanoparticles and dielectric matrix.

By using Maxwell – Garnett approximation, the dielectric permittivity $\epsilon_{\text{mix}}(\omega)$ can be written as

$$\frac{\epsilon_{\text{mix}}(\omega) - \epsilon_d}{\epsilon_{\text{mix}}(\omega) + 2\epsilon_d} = f \frac{\epsilon_m(\omega) - \epsilon_d}{\epsilon_{\text{mix}}(\omega) + 2\epsilon_d}$$

$$\epsilon_{\text{mix}}(\omega) = \frac{\epsilon_d[\epsilon_m(\omega)(1+2f) - 2\epsilon_d(f-1)]}{\epsilon_m(\omega)(1-f) + \epsilon_d(2+f)} \quad (1)$$

Where ϵ_d is a dielectric constant of transparent matrix, ω is optical frequency⁶. $\epsilon_m(\omega)$ is a dielectric permittivity of nanoparticles material, f is a filling factor of nanoparticles, i.e. their volume fraction. We can find the bulk metallic dielectric permittivity with Drude approximation in the following way:

$$\epsilon_m(\omega) = \epsilon_0 - \frac{\omega_p^2}{\omega(\omega + i\gamma)} \quad (2)$$

where ϵ_0 is a constant ($\epsilon_0 = 1.6$ for Al), ω_p is a plasma frequency ($\omega_p = 3.7$ PHz for Al), γ is a relaxation constant ($\gamma = 144.7$ THz).

For distinctness, further we shall consider silver as a material of nanoparticles. The dielectric constant for ZnO is given by

$$\epsilon(\omega) = \epsilon_\alpha - \frac{(\epsilon_0 - \epsilon_\alpha)\omega_{TO}^2}{\omega_{TO}^2 - \omega^2 - i\omega\gamma} \quad (3)$$

where ω_{TO} is the frequency of the transverse optical phonons⁷, ϵ_α and ϵ_0 are the high frequency and static dielectric constants respectively. Here γ is the damping factor.

Recently it has been shown that for centrosymmetric crystals, the dielectric function, is modified to

$$\epsilon_i(\omega) = \epsilon(\omega) + \frac{3b_{12}^4 g E_0^2(\omega)}{(b_{11} - \omega^2)^4 \epsilon_0} \quad (5)$$

If nonlinear effects are included[9]. In the above equation $b_{12} = b_{21} = \left[\frac{(\epsilon_0 - \epsilon_\infty)}{4\pi} \right]^{1/2} \omega_{TO}$, $b_{11} = -\omega_{TO}^2$ and

$$g = \frac{\omega_{TO}^2}{d^2}$$

d is the lattice parameter⁸. Equation (5) can be used to calculate the dielectric constant for ZnO. The Polariton dispersion relation is given by

$$\frac{v^2 k^2}{\omega^2} = \epsilon_i(\omega) \frac{(2-f)\epsilon_m(\omega) + f\epsilon_i(\omega)}{f\epsilon_m(\omega) + (2-f)\epsilon_i(\omega)} \quad (6)$$

Results and Discussion

The composite dielectric permittivity of the system of metal nanoparticles and dielectric medium is calculated using Eqn. (1),(2)&(3). The Polariton dispersion of the Nanocomposite material can be analysed for both linear and nonlinear cases using Eqn.(4) & (6). Here, the dispersion of the system consisting of ZnO with Aluminium nanoparticles have been studied for various filling factors. When the filling factor is zero, we have obtain the usual polariton dispersion of ZnO. When we make the filling factor as $f=0.1$, the metal behavior of Aluminium places a major role in the polariton dispersion. There are three modes of propagation for the above said configuration Here, the lower mode becomes a straight line and as a constant value i.e., ω_{TO} of ZnO.

We have observed 2 more modes of propagation when the nonlinear effect is included is as shown in Fig. 2. The lower mode of propagation is split up into three other modes. When the filling factor is increases to

0.9 we found that the behavior is exactly similar to the previous case as in Fig.1, except the middle mode. The middle mode tries to become a straight line that is due to the presence of metal nanoparticles.

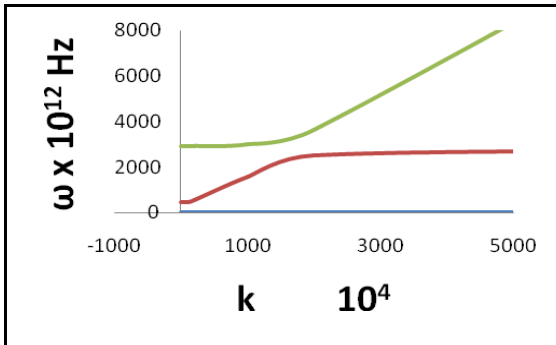


Fig.1 Dispersion curve for f=0.1 for linear

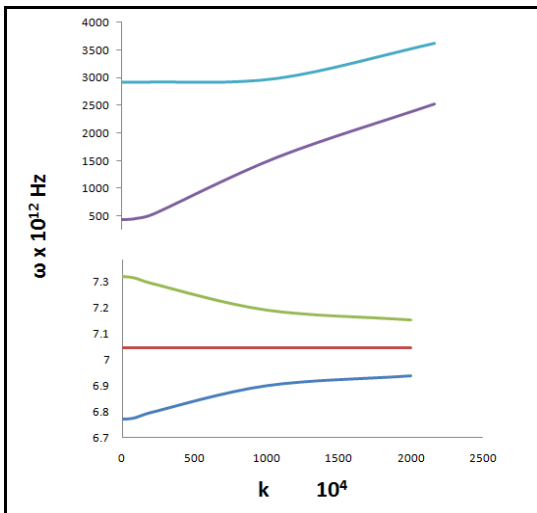


Fig. 2 Dispersion curve for f=0.1 for nonlinear

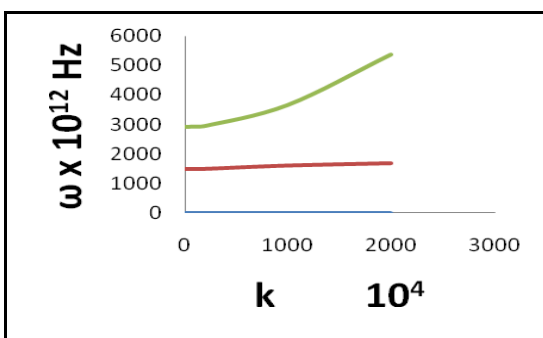


Fig. 3 Dispersion curve for f=0.9 for linear

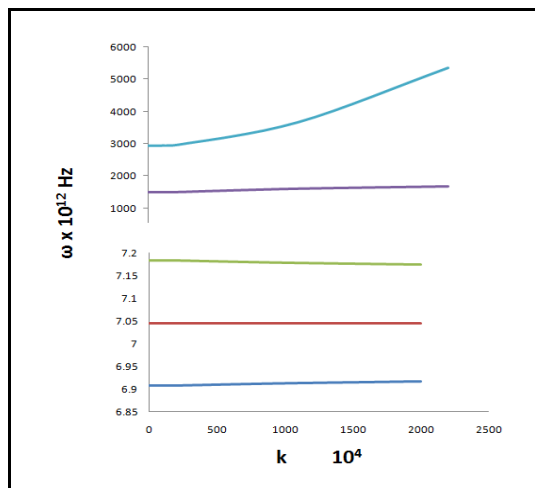


Fig. 4 Dispersion curve for $f=0.9$ for nonlinear

Conclusion

Tuning of the propagation modes occurs in the polariton dispersion is the basic concept of several applications in this era. In this work, propagation modes can be tuned by using Nanocomposite material (Al-ZnO).Polariton dispersion for various filling factor with linear and nonlinear effects are analysed. Additional modes are provided by the effect of nonlinearity. It is concluded that the tuning of propagation modes can be done by increasing the filling factor of metal nanoparticles. As the concentration of metal nanoparticles increases the curve nature of the modes becomes straight line. This may be helpful to improve the available technologies especially in the field of communication and hence to invent the devices that have potential uses across several industrial sectors.

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