

## ULTRASHORT LASER INDUCED OPTICAL SWITCHING EFFECTS OF Ga NANOPARTICLE FILMS

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The laser induced phase transformations in Ga nanoparticles deposited on silica substrate have been investigated theoretically. A reversible and controllable structural phase transformation take place at the Ga-silica interface. The temperature and intensity dependent changes in the optical properties of the two phases coexisting at the Ga-silica interface results in the formation of a metastable phase. The reflectivity changes occurring during the process have been analysed.

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### 1. Introduction

The growth of research in nonlinear optics is closely linked to the rapid technological advances that have occurred in related fields, such as ultra-fast phenomena, fiber optics, and optical communications. It includes the studies of the interaction between matter and radiation to the development of devices, components, and systems of tremendous commercial interest for widespread applications in optical telecommunications, medicine, and biotechnology [1].

The search for materials with large optical nonlinearities, as required for applications such as all-optical switching, has concentrated on media whose optical electrons exhibit a highly anharmonic response, most notably semiconductors, which exploit free excitonic and near-band-gap effects, and organic materials with weakly bound electrons [2]. Following the discovery of an optical nonlinearity based on a reversible light-induced phase transformation in nanoscale layers of material at gallium-dielectric interfaces and demonstrations of its application to cross-wavelength optical switching, photoelectric light detection, and passive *q*-switching, a similar transition-based nonlinearity has been observed in gallium nanoparticles, indicating their potential for use as active elements in low energy consumption nanophotonic and plasmonic devices [3].

The gallium nonlinearity is associated with a light-induced structural phase transition in the common form of  $\alpha$ -gallium in solid phase. It is believed that in the nanosecond-microsecond regime of optical excitation, the mechanism of the phase transition is predominantly non-thermal. Optical excitation is highly localized and destabilizes covalent bonding within the crystalline structure of alpha-gallium thus provoking a surface assisted transition to a more reflective, more metallic metastable phase. The structural phase transition drives a considerable change in the electronic, and in particular, in the optical properties of the material, also across a very broad spectral range [4].

The strength of the phase coexistence concept is supported by calculations of the optical properties of gallium nanoparticle films on a dielectric substrate performed using effective medium theory [5]. We shall present the calculations of the variation of effective dielectric constant of nanoparticles as a function of shell thickness and geometrical factor. Then, the gallium film's reflectivity and its dependence on temperature has been studied theoretically and compared with

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the available experimental data. Besides, considering the transient state of gallium as a homogenous mixture of two phases, i.e. the  $\alpha$ -Ga and liquid gallium, it is found that the reflectivity exhibits hysteresis behavior with temperature.

## 2. Theoretical Analysis

To study the variation in optical properties of gallium due to the light induced structural phase transformations, a laser beam of wavelength 1550nm is considered to be incident at the Ga-Si interface. The film is considered to be composed of spherical or nearly spherical nanoparticles (Fig.1).

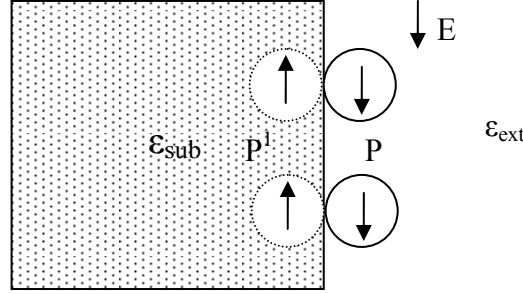


Fig. 1 The dipole moment of spherical particle lying on a silica substrate and its mirror image

The application of an external field causes the metallic inclusions to become electrically polarized and so to generate their own local fields. The dielectric constant  $\epsilon_{eff}$  of a plane-parallel film equivalent to a monolayer of small particles with the same shape, size and orientation is given by[6]

$$(\epsilon_{eff} - \epsilon_{ext})E = q \frac{(\epsilon_{int} - \epsilon_{ext})}{1 + f(\epsilon_{int} - \epsilon_{ext})/\epsilon_{ext}} E_{loc} \quad (1)$$

where  $\epsilon_{int}$  is the dielectric constant of the particles,  $E$  is the microscopic electric field inside the film,  $q$  is the volume filling factor ( $=V/l^2h$ ) and  $f$  is the geometrical (depolarizing) factor of the particles. This can be put in a better form using the relations,

$$E_{loc} = E_{ext}/(1 + \epsilon_0 \epsilon_{ext} \alpha \beta) \quad (2)$$

and

$$\alpha = (\epsilon_{int} - \epsilon_{ext})/(\epsilon_{ext} + f(\epsilon_{int} - \epsilon_{ext})) \quad (3)$$

so as to obtain

$$(\epsilon_{eff} - \epsilon_{ext})E = q \frac{(\epsilon_{int} - \epsilon_{ext})}{1 + F((\epsilon_{int} - \epsilon_{ext})/\epsilon_{ext})} E_{ext} \quad (4)$$

where

$$F = n_{ii} - 0.016\gamma^2 - \frac{0.302}{\gamma} q^{\frac{3}{2}} \quad (5)$$

is the effective geometrical factor of the particles, the quantity  $\gamma$  is called the geometrical aspect ratio and is given by the relation  $\gamma = 2r/h$ . Here  $n_{ii}$  is a tensor ( $f$ ) consisting of three depolarizing factors  $n^{(x)}$ ,  $n^{(y)}$  &  $n^{(z)}$ , which depends only on the shape of nanoparticle. Applying the boundary

condition at the interface, we can rewrite eq.(4) as the expression for dielectric constant of the monolayer [7]:

$$\epsilon_{eff} = \epsilon_{ext} \left( q \frac{(\epsilon_{int} - \epsilon_{ext})}{(\epsilon_{ext} + F(\epsilon_{int} - \epsilon_{ext}))} + 1 \right) \quad (6)$$

The interface's optical properties change on illumination because of a thin wetting layer of a highly reflective metallic phase formed between the glass and the  $\alpha$ -gallium. The change in metallic layer's thickness  $D$  with temperature  $T$  is given by[8 ]:

$$D = d_0 + \Delta \exp[-\mu(T_m - T)] \quad (7)$$

Where  $\Delta$  and  $\mu$  are fitting parameters and  $T_m$  is the melting temperature of gallium  $\sim 30^\circ\text{C}$ . Considering the transient state of gallium as a homogenous mixture of two phases i.e the  $\alpha$ -Ga and liquid gallium, if  $\epsilon_{\alpha-Ga}$  is the dielectric function of  $\alpha$ -Ga and  $\epsilon_{liq-Ga}$  is the dielectric function of liquid-Ga, then the transient dielectric function is given by[9.]:

$$\epsilon_{trans} = \epsilon_{\alpha-Ga} \left[ 1 + \frac{3C(\epsilon_{liq-Ga} - \epsilon_{\alpha-Ga})}{\epsilon_{liq-Ga} + 2\epsilon_{\alpha-Ga} - C(\epsilon_{liq-Ga} - \epsilon_{\alpha-Ga})} \right] \quad (8)$$

Where,  $C$  is the volume fraction of the liquid phase formed in crystalline  $\alpha$ -Gallium.

### 3. Numerical results and discussion

The variation of  $\epsilon_{eff}$  with  $\epsilon_{int}$  is shown in fig.2. This shows that effective dielectric constant of monolayer varies with the dielectric constant of the particles. Numerical simulations of the effective dielectric properties of gallium nanoparticles on a silica substrate indicate that phase transitions in gallium nanoparticle film just a few nanometers thick can markedly change the dielectric coefficients. As shown in fig 3., it is clear that the reflectivity of the gallium nanoparticle film increases as soon as it approaches the binary nanoshell structure [6]. However, it is observed that for shell thickness around 10nm, the response of the input signal changes markedly and the material switches to a state in which all the particles attain the limiting shell thickness for a nonlinear response. This results in the loss of reversibility of light induced transition as most of the particles are converted to new phase and the signal falls.

Fig.4 represents the dependence of the exponential factor  $\mu$  on light intensity. The graph has been plotted using the light induced metallization model which show dependence of the metallic layer's thickness through the exponential parameter  $\mu$  as a function of laser intensity. Fig.5 represents the variation of dielectric function of the transient phase with volume fraction  $C$  of liquid gallium. It is found that initially the dielectric function  $\epsilon_{trans}$  varies smoothly with  $C$  but at  $C \sim 4.5$  there is a sudden fall in the curve and then it acquires its original shape but with some decrease in value of dielectric function of the transient phase. This shows that the light-induced surface metallization effect in  $\alpha$ -gallium gives rise to a new phase (metastable phase) which exhibits entirely different dielectric properties.

Fig.6 represents the dependence of interface reflectivity on temperature and incident light intensity for  $\lambda = 1310 \text{ nm}$  &  $\lambda = 1550 \text{ nm}$ . The theoretical measurements are based on thin film formulas for reflectivity. In the linear regime a significant and abrupt reflectivity change is seen at the melting and solidification points. Supercooling is clearly seen, resulting in a well-defined hysteresis curve. On heating, a small reflectivity increase can be seen just below the bulk melting point indicating the presence of pre-melting at the interface. The dielectric coefficients of both the phases are taken as  $\epsilon_\alpha = -1.29 - i21.55$  and  $\epsilon_m = -12.18 - i38.11$  respectively. Using  $N_\alpha^2 = \epsilon_\alpha$  where  $N_\alpha = n_\alpha - ik_\alpha$  is the complex refractive index of the material we find  $n_\alpha = 3.186$  and

$k_\alpha = 3.382$ . Similar calculations for the metastable phase has been performed and the values obtained are  $n_m = 3.73$  and  $k_m = 5.108$  respectively. It is observed that the reflectivity of  $\alpha$ -gallium is  $\sim 0.55$  for  $\lambda = 1310$  nm & 1550 nm and it is found to be  $\sim 0.7$  for metastable phase. At a certain temperature  $T_a$  below the phase transition point  $T_1$  the influence of light on the surface layer's thickness becomes apparent as optical excitation changes the reflectivity of the film. With increasing temperature or level of optical excitation, the surface layer's thickness increases until the transformation of the core to the "surface" phase is completed. When at  $T_1$ , the core of the particle is fully consumed by the new phase, the nanoparticle becomes stable against return to the old phase. This process is reversible for a particular temperature range between  $T_a$  and  $T_1$ . Theoretically this range lies between 275°C and 302°C. Experimental results show the range to be between 250°C and 350°C [11]. The theoretical model presents correctly positioned peaks for reflectivity of film and the discrepancies between the theoretical and experimental results may be due to defect or inclusion in the new phase that change the optical properties of the host phase at temperatures far below its transition point.

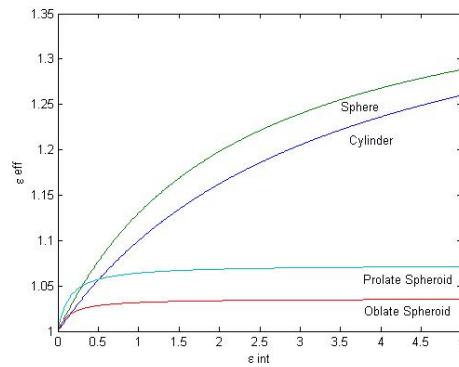


Fig. 2 The variation of  $(\epsilon_{eff})$  with dielectric constant of the particles  $(\epsilon_{int})$ .

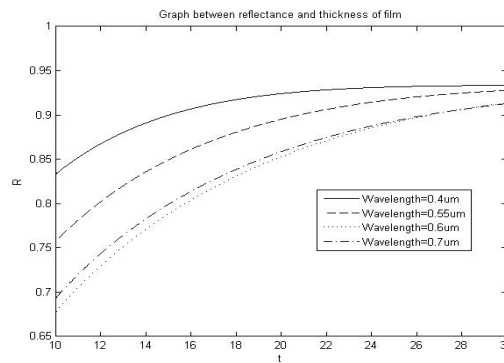


Fig.3 The Reflectance variation with film thickness.

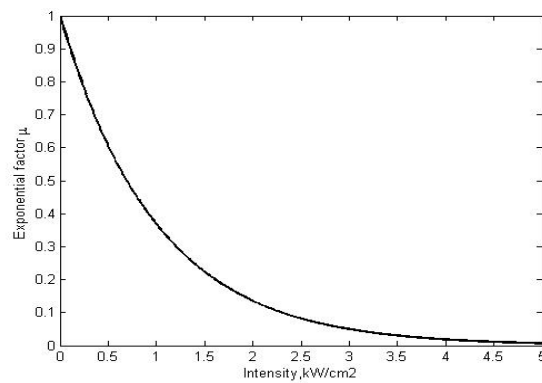


Fig.4. Variation of exponential factor with intensity of light for fitting parameter  $\Delta=18.4$  nm

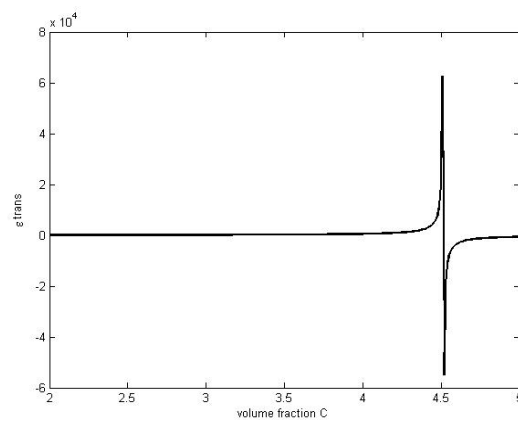


Fig. 5. Variation of effective dielectric constant of the transient phase with volume fraction  $C$

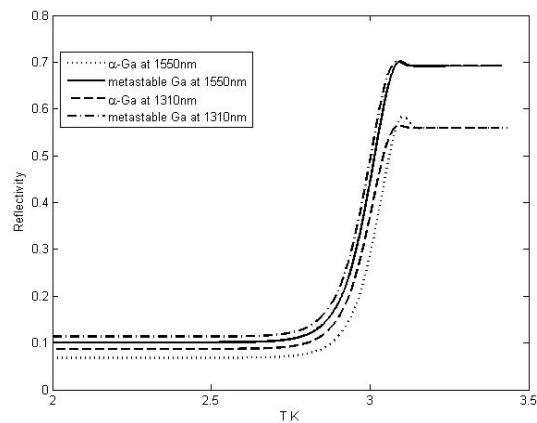


Fig. 6. Variation of reflectivity of film with temperature  $T$ .

#### 4. Conclusions

1. The optical properties of ga results from the change in the dielectric constant as a result of the geometrical factor.
2. Light can stimulate reversible structural transitions in gallium nanoparticles that belong to a novel class of surface-driven excitation-induced phase transitions.

3. The light induced conversion of  $\alpha$ -gallium to a new, more metallic, more reflective phase give rise to a big nonlinearity which can be applied to perform passive Q-switching of fiber-lasers.

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