Ultrashort laser pulse induced soliton propagation in Ga nanomaterial: a phase plane analysis

S. BISSA, P. NARUKA, A. BHARGAVA

Department of Physics, Engineering College, Bikaner-334005, India

*Nanophysics Laboratory, Department of Physics, Government Dungar College, Bikaner-334001, India

Using the well known phase plane analysis, a mechanism for achieving a large optical nonlinearity via ultrashort laser pulse induced phase transformation in Ga-Si nanostructures has been developed. It is shown that the excellent phase transformation properties of gallium may be used in optical applications. The soliton solutions of nonlinear Schrodinger equation for Ga-Si switch on the tip of an optical fiber are obtained. It is shown theoretically that the laser pulse induced excitations are solitonic in nature and lead to focusing or defocusing of the pulse. Variations in soliton shape and size at the interface for different phases has been calculated. The switch on time of gallium at the tip of an optical fiber is seen to be of order of few ps and thus can be used for the nonlinear optical miniature devices in which the medium properties may be controlled by light or laser. The critical power for the solitonic propagation of the pulse is calculated numerically.

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1. Introduction

Today gallium has become a subject of special interest as the light induced structural phase transition recently observed in gallium films has allowed for the demonstration of all optical switching devices that operate at low laser power. A large reflective nonlinearity can be achieved through optically induced phase transitions in nanoscale layers of gallium at an interface with glass [1,2]. In nanoparticles, phase transitions occur through a surface-driven dynamic coexistence of structural forms i.e. the nanophase appears first at the surface and grows inwards with increasing intensity of ultrashort laser pulse [3]. Once the core of the particle is completely converted to the new phase, the nanoparticle becomes stable against a return to the old phase because that would require the creation of a nucleation center. However, in the range where both phases are present, the balance between them can be reversibly controlled by external optical, or indeed electron beam excitation[4,5]. In the present work, the properties of Ga and light-induced phase transition mechanism in Ga-Si interface is elaborated. The optical nonlinearity produced at this interface has been analysed. The solitonic solutions of nonlinear Schrödinger Equation for Ga-Si switch on the tip of an optical fiber are obtained using phase plane analysis. The nonlinearity due to structural phase transitions in Ga-Si interface has brought us to an ideal optical data processing device.

Gallium has some useful properties which may be applied to the nanoscale nonlinear optics [6-9]. It has polymorphism forms called Ga-I (α-gallium) with metastable phases β, δ, ε, γ; Ga-II; Ga-III and Liquid Gallium, which have been seen in X-ray experiments. The ‘metallic’ behavior of Ga-II and Ga-III, similar to those of liquid gallium, is little deviated from free electron model but their optical and electronic properties are very close to an ideal free electron model. However α-Ga is special metal and it has a unique structure in which molecular and metallic properties coexist. Some inter-atomic bonds are strong covalent bonds, forming Ga2 dimers (molecules). Such type of covalent bonding leads to a dip in the density of states at the Fermi level and the rest are metallic bonds. Due to this metallic plane a strong optical absorption peak centred at 2.3 eV and spreading from approximately 0.68 eV (~1820 nm) to about 4 eV (~310 nm) is obtained.

2. Theoretical Analysis

2.1 Phase-plane analysis of Ga-Si interface

Consider a laser pulse propagating in an optical medium having a nanoscale film of gallium particles. The general discrete nonlinear Schrodinger equation for such a film can be written as [10]

\[ i \frac{d A_j}{dt} + \gamma \sum_k m_{jk} A_k + \Gamma |A_j|^2 A_j = 0 \]  

(1)

where the pulse envelope \( A=A(x,t) \) is a function of axial space coordinate and time. Here, \( \gamma \) is the coupling constant which may be considered negligible in this discussion, \( m_{jk} \) stands for the matrix element, \( \Gamma \) is a constant given by \( \Gamma = \sqrt{2 \omega_0 \varepsilon n_1 n_2} \) and the relaxation time nonlinearity is given by [1]

\[ n = n_o + \frac{n_2}{\tau} \int_0^t |A(r')|^2 \exp \left[ -(t-t')/\tau \right] dt' \]  

(2)

where \( n \) the the pulse intensity dependent index of refraction and \( n_2 \) is nonlinearity coefficient of the medium which is considered cubic here.
The susceptibility \( \chi^{(3)} \) of the medium depends upon the permittivity of the medium \( \text{Im}(\varepsilon) \), the thickness \( a \) of nanoparticle film in nm and the relaxation time \( \tau \) of the excitation in ns and the Planck’s constant \( h \) in the following manner [11]

\[
\chi^{(3)} = \frac{\text{Im}(\varepsilon)^2 a^2 \tau}{h}
\]

The propagation constant \( \beta \) and velocity of incident light depend upon the angle of incidence as:

\[
\beta = 2 n_0 \cos \psi \quad \nu = 4 n_0 \sin \psi
\]

since the pulse duration of laser beam is ultrashort, in the continuum limit the nonlinear Schrödinger equation for the optical medium under discussion can be approximated by the corresponding continuous equation. Let the set of variables \((x,t)\) change to \((x, \xi)\) for a moving frame \(gv_{xt} = \xi\). By changing over from \(x,t\) coordinated to \(x,\xi\) coordinates, we obtain the equation

\[
0 = \frac{\partial^2 A}{\xi^2} + \frac{\partial^2 |A|^2}{\xi^2} A = 0
\]

Let us assume the solution of this equation is of the form

\[
A(x, \xi) = A_\xi(\xi) \exp(-i \phi(x))
\]

Substituting the above solution in Eq. (5) and solving, we get the following equation.

\[
c(\phi_{0\xi})^2 + b \phi_0^3 + \phi_0^3 = 0
\]

Here \( \phi_{0\xi} = d^2 \phi_0 / d \xi^2 \). Again, multiplying Eq. (7) by \( 2 \phi_0 \xi \) and integrating we get

\[
c(\phi_0) = B - b \phi_0 - \sqrt{2} \phi_0^4
\]

where \( B \) is the constant of integration. As the nonlinear coefficient \( \alpha \) depends on the dielectric coefficient of various phases formed at the interface, we can obtain phase-plane curves of Eq. (8) for various phases of gallium.

2.2 Soliton Characteristics at Ga-Si interface

To study soliton propagation and characteristics at the Ga-Si interface, we consider the evolution of a laser pulse through a medium in which a nanoscale thin film of Gallium nanoparticles is deposited on the tip of a silica substrate (fig.3). In such geometry a natural decoupling of the Cartesian field components occurs. These field components satisfy Maxwell’s equations into separate sets describing transverse electric (TE) and transverse magnetic (TM) waves. Here the equation for the envelope of a TE wave is:

\[
\vec{E} = \gamma \{ f (x, z) \exp\left[ i (\beta c^2 z - \omega t)\right] \} + (*)
\]

It is assumed that the wave is polarized in the y-direction, and is incident normally at the interface while propagating along z-direction and confined in the x-direction. Thus, the envelope of wave is propagating in a nonlinear dielectric medium with refractive index \( n^2 (x, |f|^2) \). The nonlinear Schrödinger equation for such a medium is given by Eq. (1).
Eq. (1) has a general solitonic solution of the form:

\[ F(x, y) = 2\eta \sec h 2\eta (z)(x - x_0) \exp \left( \frac{ivx}{2} + 2i\sigma \right) \]  

Where,

\[ \frac{d\sigma}{dz} = -\frac{v^2}{8} + 2\eta^2 \]  

The plot of Eq.11 for gallium nanoparticle film and the silica substrate is shown in fig.4 and fig.5 [12]. The variation of nonlinearity of gallium with the effective geometrical factor of nanoparticles is shown in fig.6. For the film of nanoparticles Eq. 10 takes the form:

\[ |f(x, z)| = \sqrt{\frac{2(\beta^2 - n_0^2)}{\alpha_0}} \sec h \sqrt{\beta^2 - n_0^2} (x - x_0) \]  

Whereas, for the silica substrate we have solution of the form:

\[ |f(x, z)| = \sqrt{\frac{2(\beta^2 - n_i^2)}{\alpha_i}} \sec h \sqrt{\beta^2 - n_i^2} (x - x_i) \]  

On applying the boundary conditions at the interface, we have for the gallium nanoparticle film:

\[ x_0 = \frac{1}{2(\beta^2 - n_0^2)} \ln \left( \frac{1 + \sqrt{1 - \mu^2}}{1 - \sqrt{1 - \mu^2}} \right) \]  

Where,

\[ \mu = \frac{\beta^2 - n_0^2}{\beta^2 - n_i^2} \]  

And, in silica substrate;

\[ x_1 = \frac{1}{2\sqrt{\beta^2 - n_i^2}} \ln \left( \frac{1 + \sqrt{1 - \mu^2}}{1 - \sqrt{1 - \mu^2}} \right) \]
2.3 Dependence of soliton shape on various phases formed at the Ga-Si interface

The soliton shape characteristics at the interface depend strongly on the angle of incidence of the incident beam, the dielectric coefficients of various phases formed at the interface, the nonlinear refractive index at the interface for various phases and the nonlinear susceptibility. Measurements of the dielectric coefficient for $\alpha$-Ga for the different crystalline axes of the metal are given in [13]. Based on this data, we estimate that the dielectric coefficient of $\alpha$-Ga is $\varepsilon_1 = -25.9 - i39.7$ at 1310 nm. This is based on an assumption of equal contributions from the $a$ and $c$ axes of the Ga crystal, which is consistent with the broadband reflectivity characteristics of $\alpha$-Ga for polarized light [14]. Consequently using $N_\alpha^2 = \varepsilon_\alpha$ where $N_\alpha = n_\alpha - ik_\alpha$ is the complex refractive index of the material, we find that $n_\alpha = 2.934$ and $k_\alpha = 3.76$. From this the reflectivity $R_\alpha$ at a gallium glass interface can be calculated respectively from:

$$ R = \frac{(n_0 - n_\alpha)^2 + k_0^2}{(n_0 + n_\alpha)^2 + k_1^2} $$

(18)

Thus, using the above assumptions the optical constants of various phases of gallium can be obtained as follows;

<table>
<thead>
<tr>
<th>Phase</th>
<th>$\alpha$-Ga</th>
<th>$\gamma$-Ga</th>
<th>Liquid Ga</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n$</td>
<td>2.934</td>
<td>3.186</td>
<td>4.259</td>
</tr>
<tr>
<td>$k$</td>
<td>3.76</td>
<td>6.386</td>
<td>8.55</td>
</tr>
<tr>
<td>$\chi_0$ (m$^2$/V$^2$)</td>
<td>$1.185 \times 10^{-18}$</td>
<td>$6.539 \times 10^{-18}$</td>
<td>$8.15 \times 10^{-18}$</td>
</tr>
<tr>
<td>$R$</td>
<td>0.489</td>
<td>0.7028</td>
<td>0.7664</td>
</tr>
<tr>
<td>$\beta$ at 45°</td>
<td>4.1</td>
<td>4.505</td>
<td>6.095</td>
</tr>
</tbody>
</table>

Soliton propagation for various phases of gallium is shown in following fig. 7 and fig.8.

![Fig. 7: The solitonic pulse for different phases in Gallium Nanoparticle film](image)

![Fig. 8: Solitonic pulse in silica substrate for different phases of gallium](image)

The pulse power for soliton propagation is determined by applying paraxial ray approximation for the confined beam for the self-channeling optical medium that we have considered here

$$ P_{cr} = A_0^2 \frac{\sigma}{\tau'} \exp \left( -\frac{1}{n} \frac{dn}{dT} \right) $$

(19)

$\sigma$ is a function of reflectivity of the nanoparticle film and the duration of the pulse and $\tau'$ is the recovery time for the light induced reflectivity change for the confined gallium. Alternatively, it can be given as:

$$ P^\pm = \int_{-\infty}^{\infty} \left| F^\pm(x) \right|^2 dx $$

(20)

![Fig. 9: The pulse power as a function of propagation constant.](image)
3. Results and discussion

From the phase plane analysis of the discrete nonlinear Schrödinger equation we obtain fig. 1. It shows that the laser pulse induced excitation is solitonic in nature and it can lead to focusing or defocusing of the pulse depending upon the relaxation time of the nonlinearity. As the nonlinear coefficient \( \Gamma \) in Eq. (6) depends strongly on the effective dielectric constant \( \varepsilon_{eff} \) and nonlinear refractive index \( n_2 \) of the phase formed at the Ga-Si interface we can obtain the phase-plane curves for different phases as shown in fig. 1. It is clear from the figure that as soon as a new phase is formed at the interface the solitonic nature of the pulse changes. These phase curves are symmetrical about axes (eq. (9)). For the molecular \( \alpha \)-phase of nanoparticles we have \( b = 1.49. \) As soon as the nanoparticles undergo laser induced structural phase transition, a more metallic phase (liquid-Ga) is formed at the interface for which the constant \( b \) is obtained by using the nonlinear refractive index given in [15]. Thus, the constant \( b = 0.3239 \) for liquid-Ga is obtained. The curves are seen to have centers at \( \phi_0 = \pm |p|^{1/2}. \) For \( B = -\frac{1}{2}|p|^2, \) the whole phase curve shrinks to a point at one of these centers. The small ellipses around \( \phi_0 = |p|^{1/2}, \) depict small amplitudes. As the value of constant \( b \) obtained for the liquid phase is less than that obtained for \( \alpha \)-phase, it is found that as we move out from the center, the contours get larger for \( \alpha \)-phase and less like ellipses for the liquid phase, making the corresponding waves becoming more and more nonlinear. Moreover, the solitonic nature of the signal is effective as long as the relaxation time is within some optimum range, which is \( \sim \) ns here. This result is consistent with the experimentally observed results [7].

From fig. 2 it is clear that the nonlinearity depends upon the size of gallium nanoparticles and also on the relaxation time \( \tau (\text{ns}) \). The dependencies of nonlinearity on the relaxation time \( \tau (\text{ns}) \) can, however, be modelled accurately by assuming that a thin wetting layer of a highly reflective metallic phase forms between the glass and the \( \alpha \)-gallium via the non-thermal metallization mechanism. As soon as the layer’s thickness increases, the size of nanoparticles increases and thus the sample’s nonlinearity, increases with applied light intensity. It should be noted that even without optical stimulation, gallium (like ice at a temperature just below its melting point) develops a thin \((\sim \text{ few nm})\) film of another phase at the interface. This layer propagates into the \( \alpha \)-gallium bulk with increasing light intensity. The working hypothesis is that when the interface is exposed to light and inclusions of the new phase are created, the presence of these nuclei of the new metallic phase in the \( \alpha \)-gallium bulk shifts the delicate energy balance at the interface, leading to an increase in the thickness of the metastable layer. The energy needed to switch gallium’s nonlinearity from the \( \alpha \)-gallium level to the metallic level is about 10 mJ cm\(^{-2}\). The low energy required to achieve this kind of transition based nonlinearity in nanoparticles suggests that they could be used as active elements in nanophotonic devices operating at extremely low power levels. The non-thermal light-induced phase transformation in gallium dominates its response in the cw regime and for excitation with pulses longer than a few tens of nanoseconds. Conversion of \( \alpha \)-gallium into the metallic phase changes the nonlinearity until the molten layer’s thickness exceeds the optical skin depth. This result is consistent with the results obtained for pump-probe experiment performed with 3 ns optical pulses in the visible part of the spectrum is reported by Parravicini [16].

Fig. 4 and fig. 5 represents the solitonic solutions of Eq. (1) for the gallium nanoparticle film and interface respectively. It is clear that a measurable loss in amplitude of incident soliton is observed when it passes through the interface. It becomes sharper after change in nonlinear medium. The necessary condition for the existence of these solutions is that \( \beta > n_2 \). Also, \( \mu > 1 \) given by Eq. (16) should be less than 1 in order to have real solutions. The condition \( \mu > 1 \) implies that the propagation constant \( \beta > n_2 \). Thus, we can say that the solutions expressed in eq. (13) and eq. (14) correspond to wave packet whose envelope has a peak at \( x > 0 \). Due to laser enhanced structural phase transformation a new phase is formed at the interface and the constant \( \alpha \) change from \( \alpha = 1.185 \times 10^{-18} \) \((\alpha \text{-Ga})\) to \( \alpha = 8.15 \times 10^{-18} \) \((\text{liquid-Ga})\). Consequently, the parameters \( \Delta, n_0, \) and \( n_1 \) change giving rise to change in soliton characteristics at the interface. Thus, we conclude that stable solutions occur only when the peak is in the medium with the higher nonlinear refractive index \( \alpha_1 \).

Fig. 6 show that nonlinearity of the film increases with increase in value of effective geometrical factor significantly. Here we have used a theoretical model recently developed to describe the optical properties of a closely packed film of non-spherical, two-phase nanoparticles [17] and the predictions of this theory (CPNF model) are found to be consistent with experimental measurements of the nonlinear response of a gallium nanoparticle film. The CPNF model provides the exact expression for the electric field and effective geometrical factor of nanoparticles to account for the interaction between a given particle and its 24 nearest neighbours, their mirror images in the substrate and its own image. The effect due to the rest of the monolayer/substrate system is calculated using the dipole field approximation as in the Yamaguchi, Yoshida and Kinbara (YYK) model [18]. Using the CPNF model the particles are considered to be spheres with a radius of 50 nm distributed on the surface of a silica substrate with a mean separation of 100 nm (an approximation to the sort of film typically produced by light-assisted self assembly of gallium nanoparticles [15]). The solid core of the particles was assumed to have optical properties intermediate between those of the liquid and \( \alpha \) phases, and the shell was taken to be in the liquid state. Thus, based on
new phase for which the nonlinearity changes from enhanced structural phase transformation giving rise to a interface. This may be considered due to the laser amplitude of incident soliton when it passes through the substrate shows that there is a measurable loss in soliton formation in gallium nanoparticle film and silica nanoparticles and the relaxation time influence of an optical excitation. The nonlinear response transforms to a more metallic metastable phase under the properties of substrate show that there is a vast change in the optical curves for the upon the relaxation time of nonlinearity. The phase plane can lead to focusing or defocusing of the pulse depending laser pulse induced excitations are solitonic in nature and plane curves obtained at the Ga-Si interface depict that the solitonic solutions of the nonlinear Schrodinger equation. The phase of phase formed at the interface can be explained with the structural phase transformation at the Ga-Si interface and equation at the Ga-Si interface. It has been shown that the limitation of the Plank’s constant.

4. Conclusions

The method of phase plane analysis is used to solve the NLSE at the Ga-Si interface. It has been shown that the method of phase-plane analysis can be used for obtaining the solitonic solutions of the nonlinear Schrodinger equation at the Ga-Si interface. α-gallium undergoes structural phase transformation at the Ga-Si interface and transforms to a more metallic metastable phase under the influence of an optical excitation. The nonlinear response of phase formed at the interface can be explained with the help of the nonlinear Schrodinger equation. The phase plane curves obtained at the Ga-Si interface depict that the laser pulse induced excitations are solitonic in nature and can lead to focusing or defocusing of the pulse depending upon the relaxation time of nonlinearity. The phase plane curves for the α and liquid phase of gallium and for the substrate show that there is a vast change in the optical properties of α-gallium as it transforms to the more metallic liquid phase at the interface. This gives rise to a huge nonlinearity which depends on the size of gallium nanoparticles and the relaxation time τ(ns). The plot for soliton formation in gallium nanoparticle film and silica substrate shows that there is a measurable loss in amplitude of incident soliton when it passes through the interface. This may be considered due to the laser enhanced structural phase transformation giving rise to a new phase for which the nonlinearity changes from \( \alpha_0 = 1.185 \times 10^{-18} \) to \( \alpha_1 = 8.15 \times 10^{-18} \). Beside this the nonlinearity of film also depends on the effective geometrical factor of nanoparticles. This dependence can be studied with the help of an effective geometrical factor constructed with the help of effective medium theory used in the closely packed nanoparticle films model (CPNF). It can be further implemented for the study of solitonic character of the pulse for different phases of gallium. The critical power for solitonic propagation of pulse can also be calculated and its variation with propagation constant \( \beta \) can be studied. Thus, the nonlinearity at a gallium silica interface offers potential for the development of a wide range of practical nonlinear optical devices. The dynamic, reversible light induced phase transition mechanism can be potentially used in power limiting applications in optoelectronic applications.

References


Corresponding author: shiwangi_bissa2005@yahoo.co.in