INTERFACIAL LAYER EFFECTS ON THE ENHANCEMENT FACTOR AND OPTICAL INDUCED BISTABILITY IN SMALL SPHERICAL METAL/DIELECTRIC COMPOSITES

By

Sori Abdissa Segni

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The undersigned hereby certify that they have read and recommend to the School of Graduate Studies for acceptance a thesis entitled "**Interfacial Layer Effects on the Enhancement Factor and Optical Induced Bistability in small Spherical Metal/Dielectric Composites**" by **Sori Abdissa Segni** as a requirement for the degree of **Master of Science in Physics(Condensed Matter Physics)**.

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Supervisor: ______________________
Dr. Sisay Shewamare

Readers: ______________________

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Author: **Sori Abdissa Segni**

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Table of Contents

List of Tables

List of Figures

Abstract

In this thesis, we studied the enhancement factor of local field for pure spherical metal composite, small spherical metal/dielectric composite and optical induced bistability of metal/dielectric composites with interfacial layer in linear host matrices which has not been studded in the literature so far. For relatively small intensity of electromagnetic radiation the local field inside the small particles can be enhanced up to the inner atomic fields. This requires taking into account of the nonlinear part of the dielectric function, which in majority cases is proportional to the square amplitude of the incident electric field $|E|^2$. For such cases the local field inside the particle is determined from the cubic equation with respect to $|E|^2$. The single value of $|E_h|^2$ is able to activate three different values of the local field $|E|^2$.

Using the calculated enhancement factor of local field and the cubic equation of the optical induced bistability of the composite material, the parameters of the interfacial layer are calculated. We take positive, zero, and negative values which represents dielectric like, no interfacial, and metal like, respectively for the pure metal case. In the case of metal/dielectric for which we consider the interfacial layer particle the above mentioned properties are reversed so that it will be positive to realize the metal like properties of the interfacial layer. The analytical and numerical results show that the enhancement factor of local field is extremely enhanced and the optical induced bistability increased its domain.

Key words: Dielectric Function, Interfacial Layer, Enhancement Factor, Optical Bistabilities.

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Chapter 1

Introduction

The interfacial layer with the linear and nonlinear response of the medium strongly affects on the enhancement factor of local field and Optical Induced Bistability in the optical material and may even result in the permanent modification of its physical properties. Here, we analyzed the linear and nonlinear optical features of composite materials with metal nanostructures that are dominated by surface plasma oscillations and the interfacial layer as well as the contribution due to the metallic and dielectric properties of the composite material. It is well known that the properties of the surface plasmon (SP) strongly depends on size, shape, distribution of metal nanoparticles and the surrounding dielectric matrix offers an opportunity for the manufacturing of new promising nonlinear materials, nano-devices and optical elements. Composite materials consisting of small nonlinear metallic particles in the shape of sphere which are randomly embedded in a linear dielectric host, are well known for their complex responses to incident light fields [1, 2].

The combination of metals and dielectrics has two main purposes:

1. The first is to allow light to enter more deeply into metals, and

2. The second is to achieve light localization which in turn leads to an enhanced nonlinear response.

An interesting property of metal nanoparticle is the presence of extinction bands in the visible or infrared that results from the so-called plasmon resonances. These resonances do not exist in bulk metals and can be explained to be a consequence of the confinement of free electrons in a space smaller than one-wavelength of light which can be controlled by changing the shape of the nanoparticle and its orientation with respect to the electric field.

The study of the effective optical properties of composite materials were first concerned with the analysis of linear media [3]. The contribution of the local field of the particle was extended studying the nonlinear properties of composite materials [4, 5] with the contributions being treated as purely real and small perturbations which can be assumed to be resulting in linear behavior [5, 7]. Due to these limitations, in the case of relatively high concentration of the nonlinear components, these approximations are generally unable to properly predict the existence optical properties such as the Optical Bistability associated with the nonlinear optical response of composite materials [2, 4]. In this thesis, we are interested on the nonlinear characteristics of composite materials by considering the interfaces that separates dielectric core from concentric metallic particles embedded in linear dielectric host matrix. It is shown that the interfacial effect plays an important role on the enhancement factor of local field and the optical induced bistability (OIB) of such materials. In particular, we discuss interfacial layer effect on the enhancement factor of local field and optical induced bistability (OIB) of spherical metal/dielectric composite containing interfacial layer embedded in linear dielectric host; by making use of Laplace equation, the Taylor expansion and the classical Drude model. The interfacial factor I is introduced to characterize interfacial layer [7] and has been developed to discuss nonlinear optical response of such types of composite systems [8, 9, 10]. In addition, for numerical calculations the sample with silver/dielectric composite with interfacial layer is utilized.

1.1 Statement of the Problem

In this thesis work, we seek to study the interfacial layer effect on the enhancement factor of local field and the optical induced bistability in small spherical metal/dielectric composite which has not been discussed in the literature so far and to explore the effects metallic and dielectric inclusions.

1.2 Objectives

1.2.1 General Objectives

To study the interfacial layer effects on the enhancement factor of local field and the optical induced bstability in the metal/dielectric composite with spherical shape analytically and numerically.

1.2.2 Specific Objectives

In this study we have two specific objectives to be addressed:

1.To study the interfacial layer effect on the enhancement factor of the spherical nanoparticle for metal/dielectric composite analytically and numerically.

2. To study the interfacial layer effect on the Optical Induced Bistability of the spherical nanoparticle for metal/dielectric composite analytically and numerically.

1.3 Significance of the Study

The interfacial layer effect on the optical properties of different material needs to improve their properties by changing their size and shape. In our study we focussed on the interfacial layer effects on the enhancement factor of local field and the Optical Induced Bistability in small spherical metal/dielectric composite particle in a host matrix which has not been discussed in the literature so far and may have potential application for various optical systems such as optical swiching.

Chapter 2

Literature Review

2.1 Electrodynamical Properties of Composite System

2.1.1 Maxwell Equations and Constitutive Relation

In order to completely describe the electromagnetic properties of materials, we need to have the electric and magnetic constitutive relations, and the set of four equations commonly referred as Maxwell's equations. That is, the constitutive relations are:

$$\vec{D}(r,t) = \vec{E}(r,t) + 4\pi\vec{P}(r,t), \tag{2.1.1}$$

$$\vec{B}(r,t) = \vec{H}(r,t) + 4\pi\vec{M}(r,t) \tag{2.1.2}$$

where $\vec{D}(r,t)$ is electric displacement, $\vec{B}(r,t)$ is the magnetic induction, $\vec{H}(r,t)$ is the magnetic field, $\vec{P}(r,t)$ and $\vec{M}(r,t)$ are the polarization and magnetization of the medium respectively.

$$\nabla \cdot \vec{D}(r,t) = 4\pi\rho(r,t), \tag{2.1.3}$$

$$\nabla \cdot \vec{B}(r,t) = 0. \tag{2.1.4}$$

$$\nabla \times \vec{E}(r,t) = -\frac{1}{c}\frac{\partial \vec{B}(r,t)}{\partial t}, \tag{2.1.5}$$

$$\nabla \times \vec{H}(r,t) = \frac{4\pi}{c}\vec{J}(r,t) + \frac{1}{c}\frac{\partial \vec{D}(r,t)}{\partial t}, \tag{2.1.6}$$

where r is the 3-dimensional coordinate vector and t is time. $\rho(r,t)$ is the charge density, $\vec{E}(r,t)$ is the electric field, $\vec{J}(r,t)$ is the current density.

In the optical frequency ranges most materials are usually non-magnetic, so that the magnetic permeability is practically equal to unity practically and consequently the magnetization can be ignored. Thus, for such cases, the optical response of a medium to an electromagnetic perturbation is completely described solely by the electric constitutive relation, Eq.(2.1.1)

2.2 Electric Susceptibility and Local Field of Dielectric Media

In this section, we define the electric susceptibility (a measure of the electric polarization properties of the material) and derive the most relevant optical constants of interest. In addition, we introduce local field effects in a homogenous media, and discuss the linear and nonlinear optical properties of composite materials with metal nanostructures.

2.2.1 Local Filed and Effective Medium Approximation In Linear Optics

The macroscopic electric fields in a medium alone does not completely describe the response of the medium to an applied external electric field. It is because that, the external field drives the bound charges of the medium apart and induces a collection of dipole moments [11]. In an optically dense medium, the interaction of the induced dipoles in the medium is determined by taking into account of the local field factor. The local field is dependent on the nature the macroscopic properties of the medium. In particular, the linear polarization provides an extensive description of the light-matter interaction when the intensity of the incident radiation is sufficiently small; whereas the nonlinear optical response of a medium depends on the strength of the applied optical field, $\vec{E}(t)$. In nonlinear optics, the polarization $\vec{P}(t)$ of a medium is defined by

$$\vec{P}(t) = \chi^{(1)}\vec{E}(t), \tag{2.2.1}$$

where $\chi^{(1)}$ is known as the linear susceptibility. In nonlinear optics, the optical response can be described by generalizing equation of linear polarization by expressing the polarization $\vec{P}(t)$ as a power series in the field strength $\vec{E}(t)$ as

$$\vec{P}(t) = \chi^{(1)}\vec{E}(t) + \chi^{(2)}\vec{E}^2(t) + \chi^{(3)}\vec{E}^3(t) + ... = \vec{P}^1(t) + \vec{P}^{(2)}(t) + \vec{P}^{(3)}(t) + ... \tag{2.2.2}$$

The quantities $\chi^{(2)}$and $\chi^{(3)}$are known as the second- and third-order nonlinear optical susceptibilities, respectively.

For an optically dense medium, it is essential to know the local field in order to completely describe the response of the medium to electromagnetic fields. It is well-known that the field, also known as the local field, driving an atomic transition in a material medium is in general different from both the external field and the average field inside the medium. The variation of the local field from the average field does not play a significant role when dealing with a low-density media, and hence to describe the optical properties of such systems, one can use the macroscopic field. However, if the atomic density of a system exceeds about $10^{15}cm^3$ [12], the influence of local-field effects becomes significant and must be taken into account when describing their optical properties.

Let us consider a homogeneous dielectric medium with sufficiently large external field applied on it. The local field in such homogeneous medium is related to the macroscopic average field by the following equation:

$$\vec{E}_{Loc} = L\vec{E}, \tag{2.2.3}$$

where L is the local-field correction factor and $\vec{E}$ is the macroscopic average field. In order to find the local field acting on a typical dipole of the medium, assume that the dipole of interest is surrounded with an imaginary spherical cavity of radius much larger than the distance between the dipoles, and much smaller than the wavelength of the applied optical field. It is possible to show that the local electric field can be expressed as [13, 14, 15].

$$\vec{E}_{Loc} = \vec{E} + \frac{4\pi}{3}\vec{P}, \tag{2.2.4}$$

Eq.(2.2.4) is commonly known as the "Lorentz local field"[12].

2.3 Optical Induced Bistability

Next, we will consider the phenomenon known as optical induced bistability (OIB). It refers to the effect that some nonlinear optical systems can produce two different output intensities for a given input intensity [16]. In other words, in such media, a given value of the external electric field applied on the media may produce several values for the local field and polarization. Since its theoretical prediction in 1969 [17] and experimental realization in 1976 [18, 19, 20], OIB has been intensively studied because of its potential applications in the optical frequency ranges [21, 22, 23].

Composite materials, consisting of small, nonlinear metallic particles having a spherical or cylindrical shape randomly embedded in linear dielectric host matrix, are well known for their complex responses to incident light fields [24, 25]. It is shown that both interfacial property and size of metallic particles can affect the optical bistability behavior [26]. Practically, optical induced bistability can be realized in many types of structures. In this thesis we will consider the detailed theoretical and numerical analysis of the local field enhancement and optically induced bistability in small metal and metal covered semiconductor particles in the electrostatic approximation when $a << \lambda$, where a is a typical size of particles and λ is the wavelength of the electromagnetic wave. In addition we assume that the particles size must be large enough that the dielectric function preserve the physical meaning which can be corresponds to the nano-scaled particles. A system consisting of pure metal and metal covered dielectric nano-particles, is important as there will be a considerable enhancement of the local field inside the particle when the frequency of incident electromagnetic radiation is close to the surface plasmon frequency of the metal.

Chapter 3

Materials and Methods

3.1 Materials

The study is devoted to the theoretical study and numerical analysis of the interfacial layer effect of the special composite media with metal covered dielectric inclusions, with their possible combinations. The theory is supposed to be developed in the long wave approximation, which means that the wavelength of radiation is much greater than the typical size of inclusions. Because of the complexity of the equations of the electrodynamics of the composite media even with usage of different approaches such as Maxwell - Garnet formula it would be necessary to employ different mathematical codes such as Matlab. The apparatus that we use to carry out the theoretical part are high capacity computer and software's (MatLab software version 7.10.0.499 (R2010a)) for simulating the dielectric functions of the composite materials.

3.2 Methodology

3.2.1 Analytical Method

In this thesis one of the important methods is solving the problem analytically which is the most important input for the numerical computation.

3.2.2 Numerical Method

For determining the most important parameter for the interfacial layer effect we follow to compute the analytical results with some computational tools in MatLab software version 7.10.0.499 (R2010a) codes.

Chapter 4

Interfacial Layer Effect on Enhancement Factor of Local Field and Optical Induced Bistability in small Spherical Metal/Dielectric Composite

4.1 Analytical Description of Interfacial Layer Effect on the Enhancement Factor

4.1.1 Enhancement Factor for Pure Metal Composite

The distribution of the electric potential in the system is described by the following expressions

$$\begin{aligned} \Phi_m &= -\vec{E}_h A r cos\theta, r \leq r_1, \\ \Phi_h &= -\vec{E}_h (r - B/r^2) cos\theta, r \geq r_1. \end{aligned} \tag{4.1.1}$$

They are the solutions of the Laplace equations of the metal inclusion and the host matrix, respectively. Here $\vec{E}_h$ is the applied field, r and θ are the coordinates of the observation point (the beginning of the coordinate in the center of the inclusion and the z axis is along $\vec{E}_h$). We obtain a system of linear algebraic equations for unknown coefficients A and B from the continuity conditions of the potential and the displacement vector at the boundaries: metal-host matrix.

$$A = \frac{3\varepsilon_h}{2\varepsilon_h + \varepsilon_m} \tag{4.1.2}$$

$$B = \frac{\varepsilon_m - \varepsilon_h}{2\varepsilon_h + \varepsilon_m} r_1^3 \tag{4.1.3}$$

The quantity $|A|^2$, which we call the enhancement factor can be presented as

$$|A|^2 = \frac{9\varepsilon_h^2}{(2\varepsilon_h + \varepsilon'_m)^2 + \varepsilon''^2_m}. \tag{4.1.4}$$

4.1.2 Enhancement Factor for Pure Metal Composite with Interfacial

The distribution of the electric potential in the system is described by the following expressions

$$\begin{aligned}
\Phi_m &= -\vec{E}_h A r cos\theta, r \le r_1, \\
\Phi_s &= -\vec{E}_h (Br - C/r^2) cos\theta, r_1 \le r \le r_1 + t. \\
\Phi_h &= -\vec{E}_h (r - D/r^2) cos\theta, r \ge r_1 + t.
\end{aligned} \tag{4.1.5}$$

They are the solutions of the Laplace equations of the metal inclusion, interfacial layer and the host matrix, respectively. We obtain a system of linear algebraic equations of unknown coefficients A, B, C, and D from the continuity conditions of the potential and the displacement vector at the boundaries: Metal-interfacial,

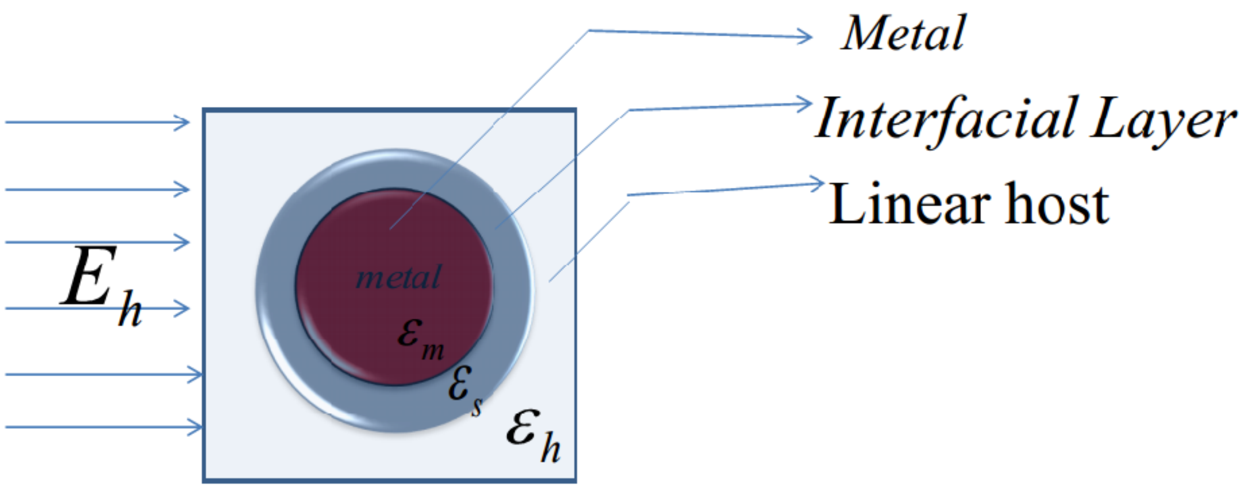

Figure 4.1: Model of the three-component particle composite system. Where ε_m=DF of metal, ε_s=DF of Interfacial Layer, ε_h= DF of Linear Host and and $E_h = AppliedField$

interfacial-host matrix.

$$\begin{aligned} A &= \frac{3\varepsilon_h}{2\varepsilon_h + \varepsilon_m + 2I/r_1} \\ B &= \frac{2\varepsilon_h}{2\varepsilon_h + \varepsilon_m + 2I/r_1} \\ C &= \frac{-\varepsilon_h}{2\varepsilon_h + \varepsilon_m + 2I/r_1} r_1^3 \\ D &= \frac{\varepsilon_m + 2I/r_1 - \varepsilon_h}{2\varepsilon_h + \varepsilon_m + 2I/r_1} r_1^3 \end{aligned} \tag{4.1.6}$$

The quantity $|A|^2$, which we call the enhancement factor can be presented as

$$|A|^2 = \frac{9\varepsilon_h^2}{(2\varepsilon_h + \varepsilon_m' + 2I/r_1)^2 + \varepsilon_m''^2}. \tag{4.1.7}$$

This quantity 4.1.7 is calculated by considering the limiting transition parameters. The limit, $t \rightarrow 0$, while $\varepsilon_s \rightarrow \infty$, the interfacial property is concentrated on a surface of zero thickness and only the quantity $t\varepsilon_s$ is of significance, we take

$$I = \lim_{t\to 0,\varepsilon_s\to\infty} \varepsilon_s t$$

To characterize the interface between pure metal particle and linear host matrix. Here, I is just called the interfacial factor. The interfacial layer is, in fact, the mixture of metal and dielectric; since ε_s is a complex number, I is also a complex quantity. But, the real part of the dielectric function of metallic particle is always a large negative number, whereas the imaginary part is a small, positive one. Thus, for simplicity, in the limit case, we may neglect the imaginary part.

A maximum of the enhancement factor $|A|^2$ in this case is obtained by setting zero the first term in the denominator 4.1.7. It gives the linear equation with respect to ε'_m

$$2\varepsilon_h + \varepsilon'_m + 2I/r_1 = 0 \tag{4.1.8}$$

4.1.3 Enhancement Factor for Spherical Metal/Dielectric composite

The distribution of the electric potential in the system is described by the following expressions

$$\begin{aligned} \Phi_d &= -\vec{E}_h A r cos\theta, r \leq r_1, \\ \Phi_m &= -\vec{E}_h (Br - C/r^2) cos\theta, r_1 \leq r \leq r_2, \\ \Phi_h &= -\vec{E}_h (r - D/r^2) cos\theta, r \geq r_2. \end{aligned} \tag{4.1.9}$$

They are the solutions of the Laplace equations of the dielectric core, metal and the host matrix, respectively. Here $\vec{E}_h$ is the applied field, r and θ are the coordinates of the observation point (the beginning of the coordinate in the center of the inclusion and the z axis is along $\vec{E}_h$). We obtain a system of linear algebraic equations for unknown coefficients A, B ,C, and D are from the continuity conditions of the potential and the displacement vector at the boundaries: dielectric -interfacial, Interfacial-metal, metal-host matrix.

$$\begin{aligned} A &= \frac{9\varepsilon_h\varepsilon_m}{2p\Delta} \\ B &= \frac{3\varepsilon_h(\varepsilon_d + 2\varepsilon_m)}{2p\Delta} \\ C &= \frac{3\varepsilon_h(\varepsilon_d - \varepsilon_m)}{2p\Delta} r_1^3 \\ D &= \{1 - \frac{3\varepsilon_h(\varepsilon_m(3-p) + \varepsilon_d p)}{2p\Delta}\} r_2^3 \end{aligned} \qquad (4.1.10)$$

where

$$\Delta = \varepsilon_m^2 + q\varepsilon_m + \varepsilon_d\varepsilon_h. \qquad (4.1.11)$$

Here

$$q = \varepsilon_d(3/2p - 1) + \varepsilon_h(3/p - 1) \qquad (4.1.12)$$

The local field $\vec{E}$ in the dielectric core can be obtained with the help of relation

$$\vec{E} = |A|\vec{E}_h, \qquad (4.1.13)$$

where $|A|$ is given by Eq.(4.1.10). In general, it is a complex function. Further, it would be convenient to deal with the real quantity $|A|^2$, which we call the enhancement factor. It can be presented as follows

$$|A|^2 = \frac{81}{4p^2} \frac{\varepsilon_h^2(\varepsilon_m'^2 + \varepsilon_m''^2)}{((\varepsilon_m'^2 - \varepsilon_m''^2 + q\varepsilon_m' + \varepsilon_d\varepsilon_h)^2 + \varepsilon_m''^2(q + 2\varepsilon_m')^2)}. \qquad (4.1.14)$$

The dielectric function (DF) of metal ε_m is chosen in the Drude form. Its real ε_m' and imagine ε_m'' parts are

$$\begin{aligned} \varepsilon_m' &= \varepsilon_\infty - \frac{1}{z^2 + \gamma^2}, \\ \varepsilon_m'' &= \frac{\gamma}{z(z^2 + \gamma^2)}. \end{aligned} \qquad (4.1.15)$$

The dielectric function of the dielectric core and the dielectric function for the host are taken to be linear for the sake of simplicity; that means we ignore the imaginary parts of ε_d and ε_h.

A maximum of the enhancement factor $|A|^2$ in this case is obtained by setting zero the first term in the denominator 4.1.13. It gives the quadratic equation with respect to ε'_m

$$\varepsilon'^2_m - \varepsilon''^2_m + q\varepsilon'_m + \varepsilon_d\varepsilon_h = 0 \tag{4.1.16}$$

4.1.4 Enhancement Factor for Spherical Metal/Dielectric Composite with Interfacial

Let us consider an individual composites of small metal/dielectric separated by interfacial layer embedded in linear host matrix. The four-component particles composite system is established: small spherical metallic particle with radius of r_2 and dielectric constant of ε_m is randomly embedded in the linear host with dielectric constant of ε_h, the dielectric core of radius r_1 and dielectric constant of ε_d is interfaced with interfacial layer with dielectric constant of ε_s and radius of $r_1 + t$ in small spherical metallic particle. In the electrostatic approximation; when a wavelength of the incident electromagnetic radiation is much greater than a typical size of the inclusion, the distribution of the electric potential in the system is described by the following expressions

$$\begin{aligned}
\Phi_d &= -\vec{E}_h Arcos\theta, r \leq r_1, \\
\Phi_s &= -\vec{E}_h(Br - C/r^2)cos\theta, r_1 \leq r \leq r_1 + t, \\
\Phi_m &= -\vec{E}_h(Dr - E/r^2)cos\theta, r_1 + t \leq r \leq r_2. \\
\Phi_h &= -\vec{E}_h(r - F/r^2)cos\theta, r \geq r_2.
\end{aligned} \tag{4.1.17}$$

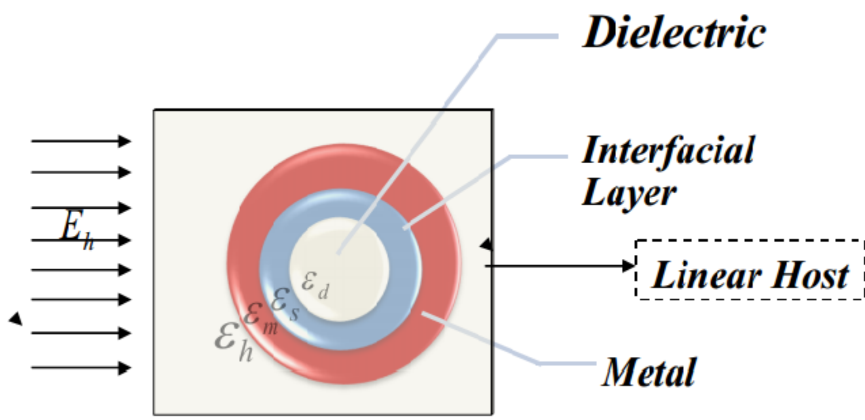

Figure 4.2: Model of the four-component particle composite system. Where ε_d= DF of dielectric core, ε_s= DF of Interfacial Layer, ε_m= DF of metal, ε_h= DF of Linear Host and $\vec{E}_h$= Applied Field

They are the solutions of the Laplace equations of the dielectric core, interfacial layer, metal inclusion and the host matrix, respectively. Here $\vec{E}_h$ is the applied field, r and θ are the coordinates of the observation point (the beginning of the coordinate in the center of the inclusion and the z axis is along $\vec{E}_h$), A, B, C, D, E, and F are unknown coefficients.

From the continuity conditions of the potential and the displacement vector at the boundaries: Dielectric core-interfacial layer, interfacial layer-concentric metal shell and concentric metal shell-host matrix, we obtain a system of linear algebraic equations for A, B, C, D, E, and F given by Eq.(4.1.17). where r_1, t and r_2 are the radii of the dielectric core, thickness of the interfacial layer and the radii of the metal shell of the inclusion, respectively. Further, we need only the coefficient A that enter into the potential of the local field in the dielectric core. They can be presented in the form

$$A = 9\frac{\varepsilon_m\varepsilon_h}{2p\Delta_I} \tag{4.1.18}$$

$$\begin{aligned}
B &= \frac{6\varepsilon_h\varepsilon_m}{2p\Delta_I}, \\
C &= \frac{-3\varepsilon_h\varepsilon_m}{2p\Delta_I}r_1^3, \\
D &= \frac{3\varepsilon_h(\varepsilon_d + \frac{2I}{r_1} + 2\varepsilon_m)}{2p\Delta_I}, \\
E &= \frac{3\varepsilon_h(\varepsilon_d + \frac{2I}{r_1} - \varepsilon_m)}{2p\Delta_I}r_1^3, \\
F &= \frac{\varepsilon_m[-\varepsilon_h(\frac{3}{p} - 1) + 2(\varepsilon_d + \frac{2I}{r_1})(\frac{3}{2p} - 1)] + \varepsilon_m^2 - \varepsilon_h(\varepsilon_d + \frac{2I}{r_1})}{2\Delta_I}r_2^2,
\end{aligned} \tag{4.1.19}$$

$$\Delta_I = \varepsilon_m^2 + Q\varepsilon_m + (\varepsilon_d + \frac{2I}{r_1})\varepsilon_h. \tag{4.1.20}$$

Here

$$Q = (\varepsilon_d + \frac{2I}{r_1})(3/2p - 1) + \varepsilon_h(3/p - 1) \tag{4.1.21}$$

where $p = 1 - (r_1/r_2)^3$ is a metal fraction in the inclusion, ε_d, ε_s, ε_m, and ε_h are the dielectric functions (DFs) of the dielectric core, interfacial layer, metal shell and the host matrix, respectively. We note that the expressions Eq.(4.1.18) have been used in [27] while studying the optical induced bistability in dielectric matrix with spherical metal inclusions with small nonlinear dielectric core. With interfacial layer; the expression Eq.(4.1.18) gives the corresponding result of [1] for the polarizability of a coated sphere. The local field E in the dielectric core can be obtained with the help of relation

$$\vec{E} = |A|\vec{E}_h, \tag{4.1.22}$$

where $|A|$ is given by Eq.(4.1.18). In general, it is a complex function. Further, it would be convenient to deal with the real quantity $|A|^2$, which we call the enhancement factor. It can be presented as follows

$$|A|^2 = \frac{81}{4p^2} \frac{\varepsilon_h^2(\varepsilon_m'^2 + \varepsilon_m''^2)}{((\varepsilon_m'^2 - \varepsilon_m''^2 + Q\varepsilon_m' + (\varepsilon_d + \frac{2I}{r_1})\varepsilon_h)^2 + \varepsilon_m''^2(Q + 2\varepsilon_m')^2)}. \quad (4.1.23)$$

This quantity Eq.(4.1.23) is calculated by considering the limiting transition parameters. The limit, $t \to 0$, while $\varepsilon_s \to \infty$, the interfacial property is concentrated on a surface of zero thickness and only the quantity $t\varepsilon_s$ is of significance, we take

$$I = \lim_{t\to 0, \varepsilon_s \to \infty} \varepsilon_s t$$

To characterize the interface between dielectric core and the concentric metallic particles. Here, I is just called the interfacial factor. The interfacial layer is, in fact, the mixture of metal and dielectric; since ε_s is a complex number, I is also a complex quantity. But, the real part of the dielectric function of metallic particle is always a large negative number, whereas the imaginary part is a small, positive one. Thus, for simplicity, in the limit case, we may neglect the imaginary part.

The DF of metal ε_m is chosen in the Drude form.

$$\varepsilon_m = \varepsilon_\infty - \tfrac{\omega_p^2}{\omega(\omega + i\nu)}.$$

where ω_p is the plasma frequency given by $\omega_p^2 = \frac{Ne^2}{(\varepsilon_0 m)}$, ω is the frequency of the incident wave, e is the charge of electron, m is the mass of electron, N is the concentration of electron. Its real ε_m' and imaginary ε_m'' parts are

$$\begin{aligned} \varepsilon_m' &= \varepsilon_\infty - \frac{1}{z^2 + \gamma^2}, \\ \varepsilon_m'' &= \frac{\gamma}{z(z^2 + \gamma^2)}. \end{aligned} \quad (4.1.24)$$

The dielectric function of the dielectric core and the dielectric function for the host are taken to be linear for the seek of simplicity. That means we will ignore the imaginary parts of ε_d and ε_h. For the analytic analysis, we consider the practically non-decaying plasma vibrations in the metal when γ is negligible. A maximum of the enhancement factor in this case is obtained setting zero the first term in the denominator 4.1.23. It gives the quadratic equation with respect to ε'_m

$$\varepsilon'^2_m + Q\varepsilon'_m + (\varepsilon_d + \frac{2I}{r_1})\varepsilon_h = 0. \tag{4.1.25}$$

4.2 Numerical Description of Interfacial Layer Effect of Enhancement Factor

4.2.1 Numerical Description for Pure Metal Composite with Interfacial

We start our numerical calculations with the enhancement factor of a composite of spherical metal with interfacial inclusion $|A|^2$ obtained from Eq.(4.1.7) versus z for different values interfacial factor $I = -2$, $I = 0$, and $I = 2$.

Table 4.1: Maximum values of enhancement factor of local field $|A|^2$ verses the resonant frequency z for pure Metal composite with interfacial layer of different values of I.

I	$\|A\|^2$	z
-2	671.3	0.354
0	470.6	0.33
2	343.9	0.316

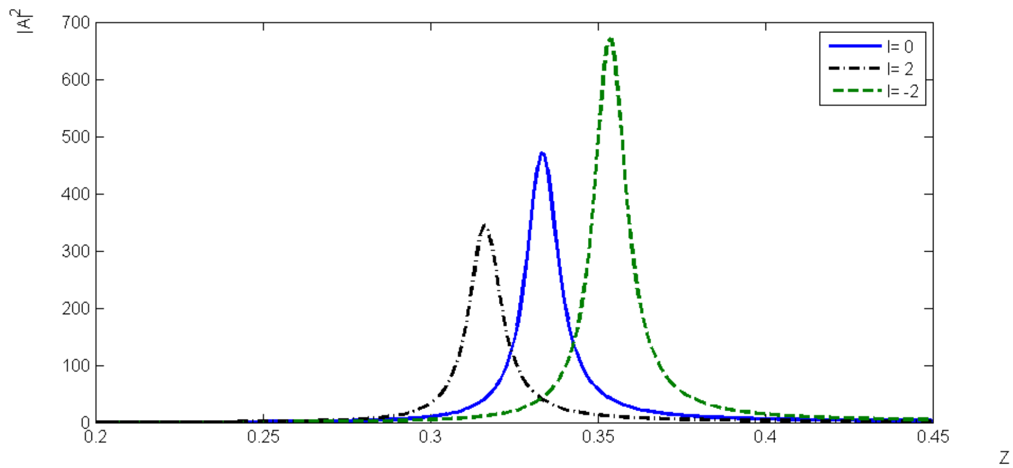

Figure 4.3: The enhancement factor $|A|^2$ for composites of small spherical silver metal particle embedded in dielectric host matrix versus z. We use the following parameters of the system: $\omega_p = 1.46 \times 10^{16}$ (silver plasma frequency), $\nu = 1.68 \times 10^{14}$, $\gamma = 1.15 \times 10^{-2}$; $\varepsilon_\infty = 4.5$, $\varepsilon_h = 2.25$, $\varepsilon_d = 6$, $I = -2, 0, 2$.

In Fig.4.3, we present $|A|^2$ of composites of pure small spherical Metal Composite with Interfacial Layer embedded in linear host matrix verses the resonant frequency Z for three different values of Interfacial Layers $I = -2$, $I = 0$ and $I = 2$.

It is shown that with the change of I from positive value to negative value, namely, with the transition of the interfacial layer from dielectric property to metallic property, the threshold values of the Enhancement factor of the local field increases.

4.2.2 Numerical Description for Metal/Dielectric composite with Interfacial

In the real inclusions, γ is not extremely small but finite. The behavior of $|A|^2$ as a function z in this case can be analyzed only numerically. The results of this study

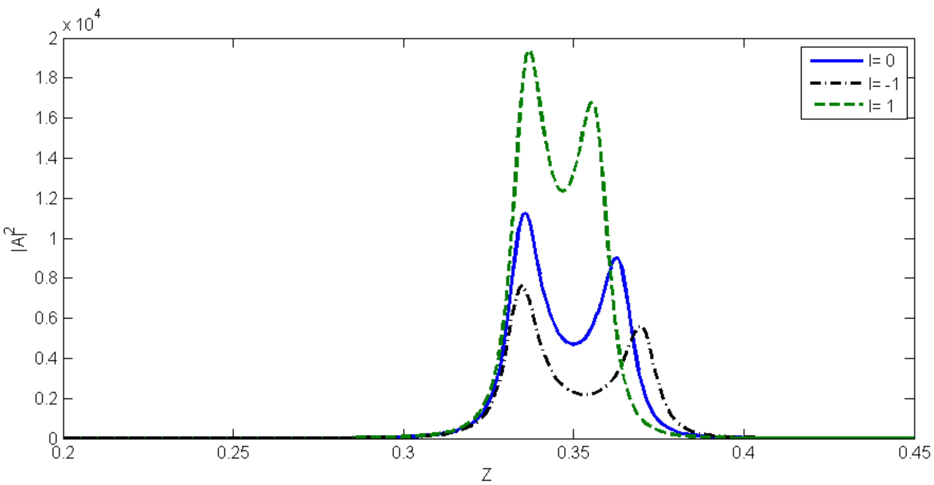

Figure 4.4: The enhancement factor $|A|^2$ for composites of small spherical silver metal/dielectric nanoparticle separated by interfacial layer embedded in dielectric host matrix versus z. We use the following parameters of the system: $\omega_p = 1.46 \times 10^{16}$ (silver plasma frequency), $\nu = 1.68 \times 10^{14}, \gamma = 1.15 \times 10^{-2}$; $\varepsilon_\infty = 4.5, \varepsilon_h = 2.25$, $p = 0.99$, $\varepsilon_d = 6, I = 0, I = 1, I = -1$.

are presented below. They qualitatively confirm the above reported analytical ones.

Table 4.2: Maximum values of enhancement factor of local field $|A|^2$ verses the resonant frequency z of Metal/Dielectric composite with interfacial layer for different values of I at $p = 0.99$.

I	$\|A\|_1^2$	z_1	$\|A\|_2^2$	z_2
-1	7633	0.335	5606	0.37
0	1.124×10^4	0.336	8999	0.363
1	1.941×10^4	0.337	1.677×10^4	0.355

We start our numerical calculations with the enhancement factor of the local field of composites of small spherical metal/dielectric inclusion with interfacial layer $|A|^2$. It can be obtained from Eq.(4.1.23) by setting $I = 0$, $I = -1$ and $I = 1$.

$$|A|^2 = \frac{81}{4p^2} \frac{\varepsilon_h^2(\varepsilon_m'^2 + \varepsilon_m''^2)}{((\varepsilon_m'^2 - \varepsilon_m''^2 + Q\varepsilon_m' + \varepsilon_d\varepsilon_h)^2 + \varepsilon_m''^2(Q + 2\varepsilon_m')^2)}. \tag{4.2.1}$$

In Fig.4.4, we present $|A|^2$ of composites of small spherical metal/dielectric separated by interfacial layer verses the resonant frequency Z for three different interfacial layer values; $I = -1$, $I = 0$ and $I = 1$. we find that the interfacial layer plays an important role in enhancement factor behavior. It is shown that with the change of I from negative value to positive value, namely, with the transition of the interfacial layer from dielectric property to metallic property, the threshold values of the enhancement factor of the local field increase. Compared with the case of no interfacial layer, the metal-like interfacial layer makes the threshold values increase, while the dielectric-like interfacial layer makes the threshold values decrease.

4.3 Analytical Description of Interfacial Layer Effect on the Optical Induced Bistability for Metal/Dielectric Composite

Consider a composite; composed of four-component particles composite system: small spherical metallic particle with radius of r_2 and dielectric constant of ε_m is randomly embedded in the linear host with dielectric constant of ε_h, The dielectric core of radius r_1 and dielectric constant of ε_d is interfaced with Interfacial layer with dielectric constant of ε_s and radius of $r_1 + t$ in small spherical metallic particle. Then the electrical potential in the system can be expressed by Eq.(4.1.17).

By introducing the interfacial factor I Now, we consider the effect of interfacial layer through the limit, $t \to 0$, while $\varepsilon_s \to \infty$, the interfacial property is concentrated on a surface of zero thickness and only the quantity $t\varepsilon_s$ is of significance, we take

$$I = \lim_{t\to 0, \varepsilon_s \to \infty} \varepsilon_s t$$

To characterize the interface between dielectric core and the concentric shell of metallic particles. Here, I is just called the interfacial factor. The interfacial layer is, in fact, the mixture of metal and dielectric; since ε_s is a complex number, I is also a complex quantity. But, the real part of the dielectric function of metallic particle is always a large negative number, whereas the imaginary part is a small, positive one. Thus, for simplicity, in the limit case, we may neglect the imaginary part. The dielectric function for a metal in the inclusion is given to be in the Drude form equation (4.1.24). The DF of the core ε_d, in general case, includes a nonlinear part with respect to the local field.

$$\varepsilon_d = \varepsilon_{d0} + \chi|\mathbf{E}|^2, \tag{4.3.1}$$

where ε_{d0} is the linear part of DF, χ is the nonlinear Kerr coefficient, and $\mathbf{E}$ is the local field in the core. The enhancement factor of the local field in the inclusion for the weak incident fields $\chi|\mathbf{E}|^2 << \varepsilon_{d0}$, the local field is presented as in the form of equation $\mathbf{E} = |A|\mathbf{E}_h$. In this relation $|A|$ is given by equation (4.1.18) which is a complex quantity. Further, it would be convenient to consider with $|A|^2$, which represent a real quantity. We call $|A|^2$ the enhancement factor and express it as equation (4.1.23). Here ε'_m and ε''_m are the real and imaginary part of ε_m given by equation (4.1.24), respectively. For the sake of simplicity, we ignore the imaginary parts of ε_h.

From $\mathbf{E} = |A|\mathbf{E}_h$ we obtain the cubic equation for the square modulus of the local field $X = \chi|\mathbf{E}|^2$ in the form

$$\alpha X^3 + \beta X^2 + \delta X = \eta Y, \tag{4.3.2}$$

where

$$\begin{aligned}
&\alpha = d_2^2 + d_4^2, \\
&\beta = 2(d_1 d_2 + d_3 d_4), \\
&Y = \chi|\mathbf{E}_h|^2, \\
&\delta = d_1^2 + d_3^2, \\
&\eta = \tfrac{81\varepsilon_h^2(\varepsilon_m'^2 + \varepsilon_m''^2)}{4p^2},
\end{aligned} \tag{4.3.3}$$

with $a = 3/p - 1$, $b = 3/2p - 1$, $c = \varepsilon_{d0} + 2I/r_1$, $d_1 = \varepsilon'_m \varepsilon_h a + \varepsilon'_m bc + \varepsilon_m'^2 - \varepsilon_m''^2 + \varepsilon_h c$ $d_2 = \varepsilon'_m b + \varepsilon_h$, $d_3 = \varepsilon''_m \varepsilon_h a + \varepsilon''_m bc + 2\varepsilon'_m \varepsilon''_m$,and $d_4 = \varepsilon''_m b$. The quantity X depends on the applied field $\mathbf{E}_h$ and the parameters of the composite. If the cubic equation Equation (4.3.2) has one real positive root, the local field in the nanoparticle is a single-valued function of the applied field. If it has three real positive roots, the local field is not a single-valued function of the applied field and the system becomes unstable. This is what we called optical induced bistability (OIB) [16]. OIB is usually illustrated in the $Y - X$ plane and connected with S-like curves showing that single value of $|E_h|^2$ is able to activate three different values of the local field $|E|^2$. This phenomenon is called the Induced Optical Bistability [1, 28].

4.4 Numerical Description of Interfacial Layer Effect on the Optical Induced Bistability for Metal/Dielectric Composite

Now, I is only a real number. When I is taken as a negative (or positive) value, the interface exhibits dielectric-like (or metal-like); and $I = 0$ corresponds to no interface. Numerical results for three different interfacial layers $I = -2, 0$ and 2 by Eq. (4.3.2) and MatLab software version 7.10.0.499 (R2010a) codes we obtain the following Fig 4.5. Interfacial effect plays an important role in a variety of systems, and it can dramatically alter the systems' optical behavior. In this paper, the effect of interfacial layer on the optical bistability is qualitatively studied, by introducing the interfacial factor I. It is shown that interfacial property can dramatically affect the optical bistable behavior. This plot (fig 4.5) exhibits bistable response clearly, and the part of the curves with negative slope are unstable. We find that the interfacial layer plays an important role in bistable behavior. It is shown that with the change of I from positive value to negative value, namely, with the transition of the interfacial layer from metallic property to dielectric property, the threshold values of the bistability decrease. Compared with the case of no interfacial layer, the metal-like interfacial

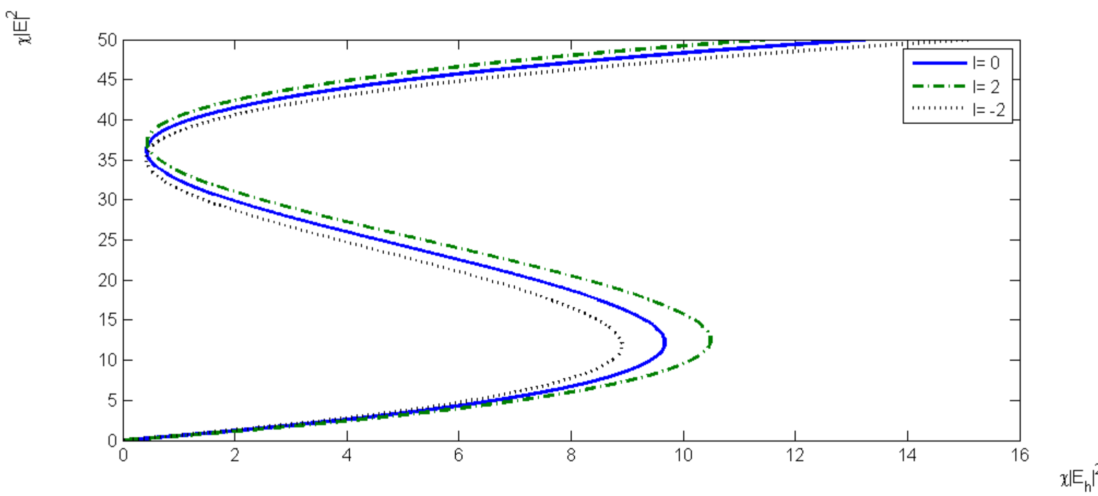

Figure 4.5: Optical Induced Bistability (IOB)in composites of small spherical metal/dielectric separated by interfacial layer in linear host matrix : $\varepsilon_{d0} = 6.$, $\varepsilon_\infty = 4.5$, $\varepsilon_h = 2.25$, $p = 0.99$, $\gamma = 0.0115$, $I = -2, 0, 2$; The local field $\chi\left|E\right|^2$ versus the applied field $\chi\left|E_h\right|^2$ at $z = 0.2$.

layer makes the threshold values increase, while the dielectric-like interfacial layer makes the threshold values decrease. It is shown in Fig.5.1 that, with increasing I, both the threshold values and bistable region are increasing, and we have known that the threshold values will increase for the case of metal-like interfacial layer; hence, the threshold values will increase.

Chapter 5

Conclusion and Recommendation

In this thesis, we studied the effect of interfacial layers on the enhancement factor of local field for pure spherical metal composite in host matrix, small spherical metal/dielectric composite randomly embedded in linear host matrix and the Optical induced Bistability (OIB) of small spherical metal/dielectric composite randomly embedded in linear host matrix. We have solved the enhancement factor of local fields for pure metal with interfacial layer, the enhancement factor of local field for metal/dielectric composites with interfacial embedded in a linear host matrix and Optical Induced Bistability (OIB) metal/dielectric composites with interfacial embedded in a linear host matrix. We have solved Laplace equation with Drude model together with Taylor. In this we have found Several interesting phenomena which are related with the enhancement factor and Optical Induced Bistability. Furthermore, we noted interfacial Layer I with thickness t and dielectric function ε_s in between dielectric core and concentric metal shell, the enhancement factor of a composites of small spherical metal/dielectric composite with Interfacial in between randomly embedded in linear host matrix; $|A|^2$ is extremely increased, meanwhile the corresponding peak is enhanced when the interfacial layer is changed from dielectric-like to metal-like, the peak corresponding to metal-like interfacial layer can be enhanced strongly.

For a given applied field; the square amplitude of the incident electric field $|E|^2$, the effect of dielectric-like interfacial layer on the induced optical bistability (IOB) is less than metal-like and (or without) interfacial layer.

Generally the analytical and numerical results show that the interfacial layer can greatly affect the Enhancement Factor of the local field and Optical Induced Bistability in metal inclusions with small dielectric cores, and that the metal-like interfacial layer is favorable to increase the threshold value of the OIB domain.

The future work will aim at studying the change that will be observed by varying the parameters considered in this study such as the metal fraction in the inclusion p.

Bibliography

[1] C. F. Bohren and D.R. Huffman, *Absorption and scattering of light by small particles*, Wiley, New York, 1983.

[2] U. Kreibig and M. Vollmer, *Optical properties of metal clusters*, Springer, Berlin, 1995.

[3] V. M. Shalaev, *Electromagnetic properties of small-particle composites*, Phys. Reports **61**, 272 (1996).

[4] K. M. Leung, *Optical bistability in the scattering and absorption of light from nonlinear microparticles*, Phys. Rev. A **33**, 2461,(1986).

[5] D. Stroud D. J. Bergman, Ohad Levy, *Theory of optical bistability in weakly nonlinear composite medium*, **49**, 129–134 (1994).

[6] , S. Arnold, T. R. Keeffe,K. M. Leung, L. M. Floan, T. SCALESE, and A. Pluchino, *Optical bistability of an aqueous aerosol particle detected through light scattering: theory and experiment*, Appl. Optics, **29**,3473,(1990).

[7] R. Nathanshon Levy and D. J. Bergman, *Electrical resonance and optical bistability in periodic composite materials*, J. Appl. Phys. **77**,4263–4273 (1995).

[8] S. Torquato and M. D. Rintoul, Phys. Rev. Lett. **75** 4067,(1995).

[9] L. GAO J. P. HUANG and Z. Y. LI, *Solid state*, Commun. **115**,347 (2000).

[10] L. GAO and J. Z. Y. LI, Appl. Phys. **87**,1620 (2000).

[11] B. A. van Tiggelen B. N. A. Lagendijk and P. de Vries, *Microscopic approach to the lorentz cavity in dielectrics*, Phys. Rev. Lett. **79**,657–660 (1997).

[12] J. Cooper J. Guo and A. Gallagher, *Selective refraction from a dense atomic vapour*, Phys. Rev. A **53**, 1130–1138 (1996).

[13] H. A. Lorentz., *Theory of electrons.* Teubner, Leipzing, 2rd ed. edition, 1916.

[14] J. D. Jackson, *Classical electrodynamics*, 3rd ed. Wiley, New York, 1998.

[15] J.van Kranendonk and J. E. Sipe, *Foundations of the macroscopic electromagnetic theory of dielectric media*, Elsevier, Amsterdam (1977).

[16] *Nonlinear optics*, Academic Press, 1992.

[17] J. Goldhar A. Szöke, V.Daneu and N.A. Kurnit, *Bistable optical element and its application*, Appl. Phys. Lett. **15** , 376 (1969).

[18] Zhenya Li Lei Gao, Liping GU, *Optical bistability in nonlinear composite media*, Phys. stat. sol. (b) **218**, 591 (2000).

[19] H. M. Gibbs, *Optical bistability: Controlling light with light, quantum electronics-principles and applications*, Academic Press, 1985.

[20] W. Bogaerts D. Van Thourhout G. Morthier G. Priem, P. Dumon and R. Baets, *Optical bistability and pulsating behaviour in silicon-on-insulator ring resonator structures*, Opt. Express **13**, 9623–9628 (2005).

[21] D. J. Hagan G. Assanto, Z. Wang and E. W. Vanstryland, *All-optical modulation via nonlinear cascading in type ii second-harmonic generation*, Appl. Phys. Lett. **67**, 2120–2122 (1995).

[22] H. Nihei and A. Okamoto, *Photonic crystal systems for high-speed optical memory device on an atomic scale*, Proc. SPIE **4416**, 470–473 (2001).

[23] , D. A. Mazurenko, R. Kerst, J. I. Dijkhuis, A. V. Akimov, V. G. Golubev, D. A. Kurdyukov, A. B. Pevtsov, and A. V. Sel'kin, *Ultrafast optical switching in three-dimensional photonic crystals*, Phys. Rev. Lett.**91**,213903,2003.

[24] T. Yanagawa K.Kurokawa Y. Hosoya, T. Suga, *Linear and nonlinear optical properties of sol-gel-derived au nanometer-particle doped alumina*, J.Appl. Phys. Lett. **81**, 1475 (1997).

[25] J. Solis C. N. Afonso D. H. Osborne R. F. Hanglud Jr. A. K. Petford-Long. R. Serna, J. M. Ballesteros, *Laser-induced modification of the nonlinear optical response of laser deposited* $cu{:}al_2o_3$, ThinSolid Films **318**, 96 (1998).

[26] Z. Y. Li T. Pan, J. P. Huang, *Optical bistability in metal/dielectric composite with interfacial layer*, Physica B **301**, 190 (2001).

[27] V. N. Mal'nev Sisay Shewamare, *Two optical bistability domains in composites of metal nanoparticles with nonlinear dielectric core*, Physica B **407**, 4837–4842 (2012).

[28] R. Inguva M.H. Birnboim N. Kalyaniwalla, J.W. Haus, *Intrinsic optical bistability for coated spheroidal particles*, Phys. Rev. A **42**, 5613(1990).

DECLARATION

I hereby declare that this thesis is my original work and has not been presented for a degree in any other University. All sources of material used for the thesis have been duly acknowledged.

Sori Abdissa

email: sori.abdissa@yahoo.com

This thesis has been submitted for examination with my approval as University advisor.

Dr.Sisay Shewamare

Jimma University email: Sisayshewa20@yahoo.com