SIZE AND INTERFACIAL LAYER DEPENDENCE OF OPTICAL BISTABILITY IN CYLINDRICAL METAL-DIELECTRIC COMPOSITION

Sera Habtie

A Thesis Submitted to

The Department of Physics

PRESENTED IN PARTIAL FULFILMENT OF THE
REQUIREMENTS FOR THE DEGREE MASTER OF SCIENCE

JIMMA UNIVERSITY
JIMMA, ETHIOPIA
JUNE 2015

JIMMA UNIVERSITY
DEPARTMENT OF
PHYSICS

This is to certify that the thesis prepared by **Sera Habtie** Graduate Studies entitled "**Size and Interfacial layer Dependence of optical bistability in cylindrical metal-dielectric composition**" in fulfillment of the requirements for the degree of **Master of Science** complies with the regulations of the University and meets the accepted standards with respect to originality and quality. .

Dated: June 2015

Supervisor: _____
Dr. Menberu Mengesha

Readers: _____

_____

JIMMA UNIVERSITY

Date: **June 2015**

Author: **Sera Habtie**

Title: **Size and Interfacial layer Dependence of optical bistability in cylindrical metal-dielectric composition**

Department: **Physics**

Degree: **M.Sc.** Convocation: **October** Year: **2015**

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Abstract

The optical bistability in a composite, composed of nonlinear cylindrical metallic particles with interfacial layers randomly embedded in a linear dielectric host, is reviewed. During the formation of composite, due to the diffusion and surface roughness, the interface between the nonlinear inclusions and the host matrix is not very sharp, leading to an interfacial layer, whose physical properties are different from those of either the inclusions or the host. It is shown that both interfacial property and the size of metallic particles can dramatically affect the optical bistable behavior of the metal-dielectric composite.

Acknowledgements

Prior to all I would like to thank God of my Holly ancestors, who had always been a constant source of encouragement and support. Next I would like to express my gratitude and deepest appreciation to my supervisor Dr.Menberu Mengesha who has provided me continual guidance and support during the course of my project, without his help and encouragement this work would never have been done. I would like to thank my colleagues at Molale Preparatory School for their great help and friendship. I would like thank to my colleagues and friends,Tigsit W/yes, Chilotu Haile and Debasu Ayenew for their priceless help during writing. Moreover I would like to express my heart felt thank to my wife Yeshemebet Asefa for help to continue my education.

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Chapter 1

Introduction

The search for new materials is one of the defining characteristics of modern science and technology. Novel mechanical, electrical, magnetic, chemical, biological, and optical devices are often the result of the fabrication of new materials. Recent advances in optical science and technology, such as the development of new lasers, detectors, and photonic devices, have relied heavily on advances in materials research[1].

The physics of metal-dielectric composites has recently gained increasing interest because of their unique linear and nonlinear optical properties, and their high application potential as nonlinear media and media for optical data storage [2]. The effective optical properties of composite materials were first devoted to the analysis of linear media and were recently extended to the study of materials with one nonlinear component under very restrictive conditions, that is, very low concentration of the nonlinear component with nonlinear contributions being treated as purely real and as low field approximation to the assumed leading linear behavior.

In a composite, where metal and dielectric components combine with each other in a disordered manner, the boundary conditions in the system are so complicated

that the determination of its electromagnetic response by solving Maxwell's equations becomes intractable. Fortunately, under certain conditions the situation can be simplified significantly. For the study of the optical properties of a composite system with an in homogeneity scale much smaller than the wavelength of interest, electrodynamic scattering by individual metal or dielectric particles is overshadowed by the average response of the whole system. Therefore, we can investigate the optical properties of a microscopically heterogeneous composite by evaluating the effective dielectric function of the macroscopically uniform medium. We obtain this effective dielectric function in terms of the permittivity's of the individual components as well as their respective volume fractions. This method is known as the effective medium approach. The most widely used effective medium approach for describing the optical properties of such composite material is the Maxwell- Garnett theory (MGT) [3].

1.1 Statement of the problem

The nonlinear optical properties of metal-dielectric composite materials have been the subject of numerous studies in recent years due to their technological potential applications . Because of their potential uses in optical devices, the most commonly considered materials are made of nonlinear particles embedded in linear host. The nonlinearities of such materials may be strongly enhanced relative to bulk samples of the same materials, and optical bistability may arise in them under certain conditions. These effects are the results of a possibly great enhancement of the electric field within the particles. This enhancement can be produced by an appropriate ratio of the host-to-particle complex dielectric permittivity and by a modification of the field inside a given particle by neighboring particles.

In general the effects of interfacial layer and particle size on optical bistability were not well understood for non-linear composites in which the host medium is non-linear. The purpose of the study is, therefore, the investigations of the important features of optical bistability for such system.

1.2 Objectives of the Study

1.2.1 General objective

To understand the optical properties of nonlinear cylindrical metal- linear dielectric composite materials that are separated by an interfacial layer. In doing so the optical bistability phenomena will be investigated.

1.2.2 Specific objective

The specific objectives of this study are:

To describe the influence of interfacial layer on the bistability property,

To explain the influence of size on the optical bistability.

1.3 Significance of the Study

This study will have its own contribution for the advancement of utilization 0f optical bistability in modern world. Thus, the engineering of nonlinear optical materials will be highly developed and it will provide good opportunity on knowledge and application for the research.

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Chapter 2

Literature Review

2.1 Material Response to Electromagnetic Radiation (EMR)

Electromagnetic radiation is a form of radiant energy released by certain electromagnetic processes [4]. EMR consisting of electromagnetic waves, including radio waves, infrared, visible light, ultraviolet, X-rays and gamma rays. Visible light is one type of electromagnetic radiation, other familiar forms are invisible electromagnetic radiations such as X-rays and radio waves. Electromagnetic waves are waves which can travel through the vacuum of outer space. Mechanical waves, unlike electromagnetic waves, require the presence of a material medium in order to transport their energy from one location to another.

When materials are exposed to electromagnetic radiation, it is important to be able to predict and alter their responses. This is possible when we are familiar with their optical properties and understand the mechanisms responsible for their optical behaviors. By "optical property" is meant a material's response to exposure to electromagnetic radiation and, in particular, to visible light. For example, in discussing on optical fiber materials, we note that the performance of optical fibers is increased

by introducing a gradual variation of the index of refraction at the outer surface of the fiber. This is accomplished by the addition of specific impurities in controlled concentrations. For instance we know light is one type of electromagnetic radiation.

The response of materials to light is described by a number of quantities, often called "optical constants." Among these the following are some of them:(ε) dielectric constant (ε), the electrical conductivity (σ), the susceptibility(χ), the refractive index (n), the extinction coefficient (k), the electromagnetic skin depth (δ), the surface impedance (Z), and many others. These quantities are neither constant nor independent. They are functions of the frequency, temperature, pressure, external magnetic field, and many other things. By knowing two of these, one that describes the absorption in the solid (such as the electrical conductivity or the extinction coefficient) and one that describes dispersion (such as the dielectric constant or the refractive index), all of the others may be calculated [5] Classically, EMR consists of electromagnetic waves, which are synchronized oscillations of electric and magnetic fields that propagate at the speed of light. The oscillations of the two fields are perpendicular to each other and perpendicular to the direction of energy and wave propagation, forming a transverse wave. Electromagnetic waves can be characterized by either the frequency or wavelength of their oscillations to form the electromagnetic spectrum, which includes, in order of increasing frequency and decreasing wavelength: radio waves, microwaves, infrared radiation, visible light, ultraviolet radiation, X-rays and gamma ray [5]. Electromagnetic waves are produced whenever charged particles are accelerated, and these waves can subsequently interact with any charged particles. EM waves carry energy, momentum and angular momentum away from their source particle and can impart those quantities to matter with which they interact. In the

classical sense, electromagnetic radiation is considered to be wave-like, consisting of electric and magnetic field components that are perpendicular to each other and also to the direction of propagation.

In the quantum theory of electromagnetism, EMR consists of photons, the elementary particles responsible for all electromagnetic interactions. Quantum effects provide additional sources of EMR, such as the transition of electrons to lower energy levels in an atom and black-body radiation. The energy of an individual photon is quantized and is greater for photons of higher frequency. This relationship is given by Planck's equation $E = h\nu$, where E is the energy per photon, ν is the frequency of the photon, and h is Planck's constant. A single gamma ray photon, for example, might carry 100,000 times the energy of a single photon of visible light.

2.1.1 Electromagnetic Radiation in Vacuum

EMR can travel not only through air and solid materials, but also through the vacuum of space. If there is no medium, there are neither electric nor magnetic dipoles nor free currents. Maxwell's equations describe how electromagnetic radiation is propagated. He showed that a varying magnetic field induces an associated varying electric field perpendicular to the magnetic field and this varying electric field in turn induces an associated varying magnetic field in the plane of the initial magnetic field. Together these two varying fields form an electromagnetic wave propagating at the speed of light in a direction perpendicular to both the electric and magnetic fields. In his electromagnetism law, James Clerk Maxwell, predicted the possibility of electromagnetic waves, that is waves consisting of oscillating electric and magnetic fields. Further, he predicted that the fields would oscillate in a direction perpendicular to the wave

velocity, that is, the waves would be transverse. According to his prediction, all electromagnetic radiation traverses a vacuum at the same velocity, that of light namely, $3 \times 10^8 m/s$ (186,000 miles/s). This velocity (c) is related to the electric permittivity of a vacuum ϵ_o and the magnetic permeability of a vacuum μ_o as;

$$c = \frac{1}{\sqrt{\epsilon_o \mu_o}} \tag{2.1.1}$$

Finally, from known electric and magnetic constants, he was able to calculate that the velocity of these waves in vacuum should be about $3 \times 10^8 m/s$. This value is identical to the speed of light in vacuum which had been measured fairly accurately by that time. These predictions immediately suggested that light is an electromagnetic wave.

2.1.2 Electromagnetic Radiation in Material Medium

In any kind of media EMR moves with speed less than speed of EMRs in vacuum, Because any transferring medias have ability of resisting the flow of energies in it. For that matter it is impossible to say that speed of light in a medium like water or air is $3 \times 10^8 m/s$. The optical phenomena that occur within solid materials involve interactions between the electromagnetic radiation and atoms, ions, and/or electrons. Two of the most important of these interactions are electronic polarization and electron energy transitions.

Electronic Polarization

One component of an electromagnetic wave is simply a rapidly fluctuating electric. For the visible range of frequencies, this electric field interacts with the electron cloud surrounding each atom within its path in such a way as to induce electronic polarization, or to shift the electron cloud relative to the nucleus of the atom with

each change in direction of electric field component. Electric polarization has two basic consequences. These are some of the radiation energy may be absorbed and light waves are retarded in velocity as they pass through the medium. The second consequence is manifested as refraction

Electron Transitions

The absorption and emission of electromagnetic radiation may involve electron transitions from one energy state to another. An electron may be excited from an occupied state at energy E_2 (the second energy level), to a vacant and higher lying one, denoted E_4 (Fourth energy level), by the absorption of a photon of energy. The change in energy experienced by the electron, ΔE, depends on the radiation frequency as follows: $\Delta E = h\nu$ where, again, h is Planck's constant and v is the frequency of the wave. At this point it is important that several concepts be understood. First, since the energy states for the atom are discrete, only specific ΔE 's exist between the energy levels; thus, only photons of frequencies corresponding to the possible ΔE's for the atom can be absorbed by electron transitions. Furthermore, all of a photon's energy is absorbed in each excitation event

2.2 Linear and Nonlinear Optics

In linear optics it is assumed that an optical disturbance propagating through an optical medium can be described by a linear wave equation and the polarization density depends linearly on the electric field strength. Nonlinear optics is the discipline in physics in which the electric polarization density of the medium is studied as a nonlinear function of the electromagnetic field of the light [6]. Being a wide field

of research in electromagnetic wave propagation, nonlinear interaction between light and matter leads to a wide spectrum of phenomena, such as optical frequency conversion, optical solitons, phase conjugation, and Raman scattering. In addition, many of the analytical tools applied in nonlinear optics are of general character, such as the perturbative techniques and symmetry considerations, and can equally well be applied in other disciplines in nonlinear dynamics. The field of nonlinear optics is complex and encompasses myriads of interesting effects and practical applications. In spite of its richness, most of the effects can be described accurately with just a few equations. Nonlinear optics is therefore limited to a simple analysis of Maxwell's equations which govern the propagation of light. In dielectric media and in absence of free charges or currents the Maxwell's equations[7] are given by

$$\nabla \cdot \mathbf{D} = 0 \tag{2.2.1}$$

$$\nabla \cdot \mathbf{B} = 0 \tag{2.2.2}$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \tag{2.2.3}$$

$$\nabla \times \mathbf{H} = \mathbf{J} + \frac{\partial \mathbf{D}}{\partial t} \tag{2.2.4}$$

where $\mathbf{D}$ is the electric displacement $\mathbf{J}$ is the current density, $\mathbf{B}$ and $\mathbf{E}$ are the magnetic and electric fields respectively. $\mathbf{H}$ and $\mathbf{D}$ are related to the magnetic and electric fields respectively as

$$\mathbf{B} = \mu\mathbf{H}, \mathbf{D} = \varepsilon_o\mathbf{E} + \mathbf{P} and \mathbf{J} = \sigma\mathbf{E} \tag{2.2.5}$$

where σ is the conductivity of the media, P is the total polarization, ε_0 and μ_0 are the permittivity and permeability of free space respectively. The total polarization is the summation of linear and nonlinear polarization parts. In the linear part of polarization, the polarization is directly proportional to the electric field. Which can be expressed as:

$$\mathbf{P} = \varepsilon_o \chi \mathbf{E} \tag{2.2.6}$$

Therefore the total polarization can be expressed as;

$$\mathbf{P} = \mathbf{P}_l + \mathbf{P}_{nl} \tag{2.2.7}$$

where $\mathbf{P}_l$ and $\mathbf{P}_{nl}$ are the linear and nonlinear polarizations respectively

$$\mathbf{P} = \varepsilon_o \chi \mathbf{E} + \mathbf{P}_{nl} \tag{2.2.8}$$

Then it is possible to express the electric field displacement vector as;

$$\mathbf{D} = \varepsilon_o \mathbf{E} + \varepsilon_o \chi \mathbf{E} + \mathbf{P}_{nl} \tag{2.2.9}$$

Substituting equation (2.2.9) into (2.2.3) gives us;

$$\nabla \times \mathbf{H} = \frac{\partial(\sigma \mathbf{E} + \varepsilon_o \mathbf{E} + \varepsilon_o \chi \mathbf{E} + \mathbf{P}_{nl})}{\partial t} \tag{2.2.10}$$

Then we can now use these Maxwell's equations to develop the wave equation for this phenomenon. Let's us determine the time derivative of equations (2.2.3) and (2.2.10). It becomes;

$$\nabla \times \frac{\partial \mathbf{E}}{\partial t} = -\frac{\partial^2 \mathbf{B}}{\partial t^2} \tag{2.2.11}$$

$$\nabla \times \frac{\partial \mathbf{H}}{\partial t} = \varepsilon_o (1 + \chi) \frac{\sigma^2 \mathbf{E}}{\partial t^2} + \frac{\partial^2 \mathbf{P}_{nl}}{\partial t^2} + \sigma \frac{\partial \mathbf{E}}{\partial t} \tag{2.2.12}$$

And the curl of these two equations also looks like as follows;

$$\nabla \times \nabla \times \mathbf{E} = -\nabla \frac{\partial \mathbf{B}}{\partial t} = -\mu \nabla \times \frac{\partial \mathbf{H}}{\partial t} \tag{2.2.13}$$

$$\nabla \times \nabla \times \mathbf{H} = \varepsilon_o(1+\chi)\nabla \times \frac{\partial \mathbf{E}}{\partial t} + \sigma \nabla \times \mathbf{E} \tag{2.2.14}$$

Then combining equation (2.2.11) with (2.2.13):

$\nabla \times \frac{\partial \mathbf{E}}{\partial t} + \nabla \times \nabla \times \mathbf{E} = -\frac{\partial^2 \mathbf{B}}{\partial t^2} - \mu \nabla \times \frac{\partial \mathbf{H}}{\partial t}$

$\Rightarrow \nabla \times \nabla \times \mathbf{E} = -(\nabla \times \frac{\partial \mathbf{E}}{\partial t} + \frac{\partial^2 \mathbf{B}}{\partial t^2}) - \mu(\varepsilon_o(1+\chi)\frac{\partial^2 \mathbf{E}}{\partial t^2} + \frac{\partial^2 \mathbf{P}_{nl}}{\partial t^2} + \sigma \frac{\partial \mathbf{E}}{\partial t})$

$\Rightarrow \nabla \times \nabla \times \mathbf{E} = -\mu(\varepsilon_o(1+\chi)\frac{\partial^2 \mathbf{E}}{\partial t^2} + \frac{\partial^2 \mathbf{P}_{nl}}{\partial t^2} + \sigma \frac{\partial \mathbf{E}}{\partial t})$

Because $\nabla \times \frac{\partial \mathbf{E}}{\partial t} + \frac{\partial^2 \mathbf{B}}{\partial t^2} = 0$

$$\Rightarrow \nabla \times \nabla \times \mathbf{E} + \mu\varepsilon_o(1+\chi)\frac{\partial^2 \mathbf{E}}{\partial t^2} + \frac{\partial^2 \mathbf{P}_{nl}}{\partial t^2} + \sigma\mu \frac{\partial \mathbf{E}}{\partial t} = 0 \tag{2.2.15}$$

On the other hand combining equation (2.2.12) with (2.2.14) leads us;

$\nabla \times \frac{\partial \mathbf{H}}{\partial t} + \nabla \times \nabla \times \mathbf{H} = \varepsilon_o(1+\chi)\frac{\partial^2 \mathbf{E}}{\partial t^2} + \frac{\partial^2 \mathbf{P}_{nl}}{\partial t^2} + \sigma \frac{\partial \mathbf{E}}{\partial t} + \varepsilon_o(1+\chi)\nabla \times \frac{\partial \mathbf{E}}{\partial t} + \sigma \nabla \times \mathbf{E}$

Rearranging and eliminating similar terms in the above equation leads us;

$$\nabla \times \nabla \times \times \mathbf{H} + \mu\varepsilon_o(1+\chi)\frac{\partial^2 \mathbf{H}}{\partial t^2} + \mu\sigma \frac{\partial \mathbf{H}}{\partial t} = 0 \tag{2.2.16}$$

But since we are not interested on the magnetic field we will ignore equation (2.2.16) and manipulating equation (2.2.15) using the following condition can leads as to the required wave equation. From identity we know that;

$$\nabla \times \nabla \times \mathbf{E} = \nabla(\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E} \tag{2.2.17}$$

But from Maxwell's equation we know that;

$\nabla . \mathbf{E} = 0$

As a result equation (2.2.17) can be reduced to;

$$\nabla \times \nabla \times \mathbf{E} = -\nabla^2 \mathbf{E} \tag{2.2.18}$$

Substituting equation (2.2.18) into (2.2.15) gives the general wave equation for nonlinear polarization, which is expressed as;

$$\nabla^2\mathbf{E} = \mu\varepsilon_o(1+\chi)\frac{\partial^2\mathbf{E}}{\partial t^2} + \mu\frac{\partial^2\mathbf{P}_{nl}}{\partial t^2} + \sigma\mu\frac{\partial\mathbf{E}}{\partial t} \tag{2.2.19}$$

But since all are constants it is possible to express the coefficients of the second term as

$\varepsilon = \varepsilon_o(1+\chi)$

Therefore equation (2.2.19) can be expressed as;

$$\nabla^2\mathbf{E} = \mu\varepsilon\frac{\partial^2\mathbf{E}}{\partial t^2} + \mu\frac{\partial^2\mathbf{P}_{nl}}{\partial t^2} + \sigma\mu\frac{\partial\mathbf{E}}{\partial t} \tag{2.2.20}$$

2.2.1 Nonlinear Polarization

In linear optics, the polarization density depends linearly on the electric field strength in a manner that can often be described by the relationship.

$\mathbf{P} = \varepsilon_o\chi\mathbf{E}$

where ε_o is the permittivity of free space and χ is the electric susceptibility of the medium. As the magnitude of the applied electric field increases, the linear relationship between $\mathbf{P}$ and $\mathbf{E}$ breaks down and we enter the realm of nonlinear optics. Then, the resulting polarization can be expressed as a series in increasing powers of the electric field. Mathematically, such a series can be expressed as,

$\mathbf{P}(t) = \epsilon_o[\chi^{(1)}\mathbf{E}(t) + \chi^{(2)}\mathbf{E}^2(t) + \chi^{(3)}\mathbf{E}^3(t) + ...]$

where we ignored the vector nature of the fields as well as dispersion, for simplicity. The expansion coefficients, $\chi^{(1)}$ and $\chi^{(2)}$, etc, are identified as linear and nonlinear susceptibilities, respectively.In general, the nonlinear susceptibilities depend on the frequencies of the applied fields, but under our present assumption of instantaneous response, we take them to be constants[8].We shall refer to $\mathbf{P} = \chi^{(2)}\mathbf{E}^{(2)}$ as the

second-order nonlinear polarization and $\mathbf{P} = \chi^{(3)}E^{(3)}$ as the third-order nonlinear polarization.

2.2.2 Nonlinear Optical Materials

A NLO material is a compound in which a nonlinear polarization is invoked on application of an intense electric field. This electric field results from the external application of an intense laser source. NLO materials for integrated nonlinear optics pose stringent problems as regards their process ability, adaptability and interfacing with other materials. These additional requirements are intrinsically related to the fabrication of nonlinear integrated devices, which besides efficiently performing the expected nonlinear operation, must be miniaturized, compact, reliable and with precisely reproducible characteristics in large-scale production and long term operation. On the basis of the three types of cohesive forces that bind the charges and polarization together, the NLO materials can be classified into the following cases: ionic crystals,which essentially consist of oxygen-polyhedra based solids,covalent crystals essentially dealing with semiconductors and molecular crystals that with organic materials, disordered and amorphous solids, in particular glasses and polymers, composites and inhomogeneous artificial solids [9].

The nonlinear frequency preserving effects are the ones that are most seriously considered in integrated optical devices. They can be either all optical or hybrid (parametric) effects. All optical nonlinearities essentially involve valence electron motion and are in general weaker than the hybrid ones where the ionic motion, vibrational, orientational or translational can set up very large nonlinearities; the situation however is reversed as regards the speed of establishing and erasing these nonlinearities, the electronic polarization being much faster than the ionic ones. The magnitude

and speed of the nonlinearities are essential characteristics in any assessment of the material for NLO devices[10].

2.3 Optical Bistability

A nonlinear system is said to be optically bistable if the system exhibits two output intensities for the same input intensity when the input is varied over a range of values [11,12]. Optical bistability is a rapidly expanding field of current research, because of its potential application for laser pulse shaping, optical switching, signal processing, memory elements, and because of the interesting phenomena it encompasses [11,13].

Optical bistability is usually studied by characterizing the transmitted or reflected intensity as a function of input intensity in hysteresis-like curves by rapidly switching of the excitation source. Since the first observation of optical bistability in a passive, unexcited medium of sodium (Na) vapor in 1974 [11], bistability has been observed in many different materials including GaAs [14], InSb [15], InAs [16], and in GaAs/GaAl Assuper lattices [17]. Optimizing the devices by decreasing their size, switching times, and operating power and operating them at room temperature have been the focus of intense research activities in recent times [11].

2.4 Local Responses of material

It is customary to write Ohm's law as;

$$\mathbf{J} = \sigma \mathbf{E}$$

where $\mathbf{J}$ is current density at point, σ is the conductivity of the material and E is the surrounding electric field. But this equation has a direct implication that some sort of assumption has been taken, the assumption is that the response is local[18].

Because, that the current density **J** at point depends only on the electric field **E** at that point and the conductivity σ. This statement is never completely true; the current possesses a certain momentum and, once set in motion, takes a certain time (e. g., the relaxation time τ) or distance (e. g., the mean free path) to relax back to zero. Even though, local electrodynamics is the usual case, because the mean free path is typically short compared to the lengths over which the field itself varies, and so the current is indeed related to the field by a local relation. However, in pure metals at low temperatures, the mean free path can become longer than the electromagnetic penetration depth. In that case, currents generated within the skin depth can travel deep into the metal, where the electric field is essentially zero. Nonlocal effects appear also in superconductors [5].

2.5 Local Field Effects and Effective Medium Theory

Clausius Mossotti(Lorentz-Lorenz) relation tend to obscure the physical origin of local-field effects by proceeding from the macroscopic dielectric function of the equivalent homogeneous system to the microscopic parameters of the model. The microscopic and macroscopic aspects can be made clearer by reversing the order, that is, by first obtaining the microscopic solution and then implementing the definition of macroscopic quantities as averages of their microscopic counter parts. This approach also leads naturally into a treatment of effective medium theory and description of the dielectric response heterogeneous materials [19]. Maxwell's equations are completely analogous in that they also express dependences among macroscopic average or observable quantities, in this case the electromagnetic fields E, D, and $\mathbf{B} = \mu\mathbf{H}$

If the material is considered especially homogeneous the dielectric function ε, then a uniform applied field will generate uniform dipole moment per unit volume, $\mathbf{p}$, where $\mathbf{p}$ and $\mathbf{E}$ are related to $\mathbf{D}$ and $\mathbf{E}$ by the macroscopic definitions as

$$\mathbf{D} = \varepsilon\mathbf{E} = \mathbf{E} + 4\pi\mathbf{P} \tag{2.5.1}$$

The polarizabilities α for the given dipole moment per unit volume is given by

$$\mathbf{P} = \alpha\mathbf{E}_{loc} = \alpha(\mathbf{E} - \frac{4\pi}{3}\mathbf{P}) \tag{2.5.2}$$

$\mathbf{E}_{loc} = \mathbf{E} + \frac{4\pi}{3}\mathbf{p} = \mathbf{E} + \frac{\mathbf{p}}{3\varepsilon_o}$

The microscopic approach is ideally suited for treating heterogeneous materials which are mixtures of constituents of different polarizabilities where the polarization $\mathbf{P}$is given by

$$\mathbf{P} = (N_a\alpha_a + N_b\alpha_b)\mathbf{E}_{LOC} \tag{2.5.3}$$

$$\mathbf{E} = \mathbf{E}_{LOC} - \frac{4\pi}{3}\mathbf{P} \tag{2.5.4}$$

where N_a and N_b are the volume densities of phases a and b.

The dielectric function of the mixture can be calculated from equation (2.5.1) i.e. again with polarization

$$(\mathbf{P}) = N\alpha\mathbf{E}_{loc} = \varepsilon_o\chi\mathbf{E}$$

$$\varepsilon_o\chi\mathbf{E} = N\alpha(\mathbf{E} + \frac{\mathbf{p}}{3\varepsilon_0})$$

$$\varepsilon_o\chi\mathbf{E} = \frac{N\alpha}{3\varepsilon_o}(3 + \chi)\varepsilon_0\mathbf{E}$$

or $\chi = \frac{N\alpha}{3\varepsilon_o}(3+\chi)$, but $\chi = \varepsilon - 1$, and rearranging this we get

$\Rightarrow \frac{\varepsilon-1}{\varepsilon+2} = \frac{1}{3\varepsilon_o}(N\alpha)$, this is clausius moossotti, where N is the number density

From equation (2.5.1) since

$\mathbf{P} = (N_a\alpha_a + N_b\alpha_b)\mathbf{E}_{LOC}$ which $E = E_{LOC} - \frac{4\pi}{3}P$ and then

$$\Rightarrow \frac{\varepsilon-1}{\varepsilon+2} = \frac{4\pi}{3}(N_a\alpha_a + N_b\alpha_b) \tag{2.5.5}$$

In the heterogeneous dielectric and effective medium theory, it is convenient to write equation (2.5.5) in terms of the dielectric functions ε_a and ε_b of the phases a and b

$$\frac{\varepsilon-1}{\varepsilon+2} = f_a\frac{\varepsilon_a-1}{\varepsilon_a+2} + f_b\frac{\varepsilon_b-1}{\varepsilon_b+2} \tag{2.5.6}$$

with $f_a = \frac{N}{V}\pi r_a^2 L_a$ and $f_b = \frac{N}{V}\pi r_b^2 L_b$

are volume fraction occupied by the phases a and b and V is the total volume of the cylindrical system. But where r is located in the region a and b, performing the volume averages using the above expressions the macroscopic parameters **E** and **P** yields

$$\frac{\varepsilon-\varepsilon_b}{\varepsilon+2\varepsilon_b} = f_a\frac{\varepsilon_a-\varepsilon_b}{\varepsilon_a+2\varepsilon_b} \tag{2.5.7}$$

Equation (2.5.7) is the Maxwell-Garnett effective-medium expression [20] Equations (2.5.6) and (2.5.7) have the same general formula gives

$$\frac{\varepsilon-\varepsilon_h}{\varepsilon+2\varepsilon_h} = f_a\frac{\varepsilon_a-\varepsilon_h}{\varepsilon_a+2\varepsilon_h} + f_b\frac{\varepsilon_b-\varepsilon_h}{\varepsilon_b+2\varepsilon_h} \tag{2.5.8}$$

where ε_h is the dielectric function of a host medium. Thus ε_h equals 1(void) and ε_b for the Lorentz-Lorenz and Maxwell Garnett expressions, respectively.

If $f_a > f_b$, more appropriate choice for ε_h in the Maxwell Garnett case is ε_a. However; the resulting values of ε are different for the two choices. In the above formulation this

is equivalent to choosing $\varepsilon_h = \varepsilon$, in which case the left-hand side of equation(2.5.8) vanishes and

$$0 = f_a \frac{\varepsilon_a - \varepsilon}{\varepsilon_a + 2\varepsilon_0} + f_b \frac{\varepsilon_b - \varepsilon}{\varepsilon_b + 2\varepsilon_0} \tag{2.5.9}$$

This is the Bruggeman effective-medium expression, or in conventional terminology, the effective-medium approximation (EMA)

Chapter 3

Materials and Methodology

3.1 Materials

The study deals on determining the size and interfacial layer dependence on optical bistability in cylindrical geometry .The study is supported by different materials such as books, published articles, thesis and dissertations, which are the main sources of the theory. More over MatLAB Software is used to solve the problem numerically as well as graphically.

3.2 Methodology

3.2.1 Analytical

In this thesis one of the method used to determine the size and interfacial layer dependence of optical bistability in metal-dielectric composition is analytical method. That is the optical bistable behavior of gold is obtained using laplace equation and its solution as well as the continuity equation analytical method.

3.2.2 Computational

We employed computational method using Matlab/Fortran program by developing suitable computer codes for studying the properties of nonlinear optical media of composite materials. To determine the effects of interfacial layer and particle size on optical bistability in cylindrical geometry computational and graphical techniques are employed. In general mathematical important optical relations, size and interfacial layer dependence model equations and the result of original and review figures in the thesis are interpreted computationally and graphically with the help of MatLAB software.

Chapter 4

The effect of interfacial layer and the trend of optical bistability on cylindrical metal dielectric composite

4.1 Introduction

In this part of the thesis we are going to observe the interfacial effect on two different models. On the first model the core is considered as metallic and nonlinear, whereas on the second model the core is considered as dielectric and nonlinear. The host environment is dielectric for the cylindrical metallic core and metallic for dielectric core and considered as linear. The surface (layer) between the two systems is considered as interface. The composite is considered as a three phase system and the interface has either metallic or dielectric property.

Mathematical formalism of the problem

To study the interfacial effects of the interface separating the metallic core of radius a and the dielectric host matrix we considered the interface layer as a shell with radius

$a + \delta$.Interfacial effects are known to be important in different systems and can dramatically change the effective behavior. For example, thermal or electrical resistance at the interface due to roughness can significantly reduce the effective conductivity and debonding at the interface can erode effective behavior of the composite.

The mathematical formalism of this three phase is employed from the well-known Laplace's equation $\nabla^2\phi = 0$. Because of translational symmetry along the z-axis ,ϕ is independent of z, and we need only consider the problems in $r - \theta$ plane.

Assuming the quasistatic approximation that is valid, the two dimensional electrostatic potential can be expressed in the cylindrical coordinate system. With origin at the center of cylinder and Z-axis parallel to applied electric field $\mathbf{E}_0$ the electric potential ϕ is symmetric with respect to ϕ satisfying $\nabla^2\phi = 0$.

The most general solution of this Laplace equation is

$$\phi(r,\theta) = A_0 + B_0 ln(r) + \sum_{n-1}^{\infty}(A_n r^n + C_n r^{-n})[Bcos(n\theta) + Dsin(n\theta)]$$

At infinity the potential is that of a homogenous electric field i,e $\phi = -\mathbf{E}_0 r cos\theta$

According to the electric potential the system are given by the solution of Laplace equations in cylindrical coordinates [21]. The potential distribution for the three phase system core, shell and host is given by

$$\phi_c = -\mathbf{E}_0 A r cos\theta, r < a$$

$$\phi_s = -\mathbf{E}_0(Br - \tfrac{Ca2}{r})cos\theta, a < r < a + \delta$$

$$\phi_h = -\mathbf{E}_0(r - \tfrac{G(a+\delta)^2}{r})cos\theta, r > a + \delta$$

A structural parameter $\sigma = (\frac{a}{a+\delta})^2$ with $(0 \leq \sigma \leq 1)$ is defined to describe the thickness of the shell. With the condition of the quasistatic approximation, the size

of the particle is smaller than the wavelength of the incident wave. We have chosen the center of the cylindrical to be the origin of the coordinate system, and the field at large distance from the cylindrical is uniform. So the potential is continuous everywhere. Further boundary conditions are obtained from the cylindrical surface. These are the normal component of the electric displacement $\mathbf{D}$ across the surface, because due to $\nabla.\mathbf{D} = \frac{\rho}{\varepsilon_o}$, we can state that the sources of $\mathbf{D}$ are the true charges only. Furthermore the tangential component of the field intensity should be continuous. The boundary conditions for core-shell and shell-host matrix are mathematically described as follows: The normal component is given by:

$$\varepsilon_c \frac{\partial \phi_c}{\partial r}|_{r=a} = \varepsilon_s \frac{\partial \phi_s}{\partial r}|_{r=a}$$

and

$$\frac{\partial \phi_c}{\partial \theta}|_{r=a} = \frac{\partial \phi_s}{\partial \theta}|_{r=a}$$

where as the tangential component is given by:

$$\varepsilon_s \frac{\partial \phi_s}{\partial r}|_{r=a+\delta} = \varepsilon_h \frac{\partial \phi_h}{\partial r}|_{r=a+\delta}$$

and

$$\frac{\partial \phi_s}{\partial \theta}|_{r=a+\delta} = \frac{\partial \phi_h}{\partial \theta}|_{r=a+\delta}$$

Using the normal component part, we obtain

$$\varepsilon_c \frac{\partial \phi_c}{\partial r}|_{r=a} = -\varepsilon_c (A E_0 cos\theta)$$

$$\frac{\partial \phi_c}{\partial \theta}|_{r=a} = (ArE_0 sin\theta)$$

$$\varepsilon_s \frac{\partial \phi_s}{\partial r}|_{r=a} = \varepsilon_s(-B - c\frac{a^2}{a^2})E_0 cos\theta)$$

$$\frac{\partial \phi_s}{\partial \theta}|_{r=a} = (Ba - c\frac{a^2}{a})E_0 sin\theta$$

Also using the tangential component part we obtain:

$$\varepsilon_s \frac{\partial \phi_s}{\partial r}|_{r=a+\delta} = \varepsilon_s(-B - C\frac{a^2}{(a+\delta)^2}E_0 cos\theta$$

$$\frac{\partial \phi_s}{\partial \theta}|_{r=a+\delta} = B(a+\delta) - C\frac{a^2}{(a+\delta)}E_0 sin\theta$$

$$\varepsilon_h \frac{\partial \phi_h}{\partial r}|_{r=a+\delta} = \varepsilon_h(-1 - G\frac{(a+\delta)^2}{(a+\delta)^2}E_0 cos\theta$$

$$\frac{\partial \phi_h}{\partial \theta}|_{r=a+\delta} = (a - G\frac{(a+\delta)^2}{a}E_0 sin\theta$$

After differentiation and some mathematical manipulation, the solution (undetermined coefficients) of the system of linear equations as follows

$$A - B + C = 0$$

$$\varepsilon_s B - \varepsilon_h G + \varepsilon_s \frac{a^2}{(a+\delta)^2}C - \varepsilon_h = 0$$

$$\varepsilon_s B + \varepsilon_s C - \varepsilon_c A = 0$$

$$B - C\frac{a^2}{(a+\delta)^2} + G - 1 = 0$$

Now using a four by four matrix and solving linear equation, we obtain:

$$A = \frac{4\varepsilon_h \varepsilon_s}{(\varepsilon_c+\varepsilon_s)(\varepsilon_s+\varepsilon_h)-(\varepsilon_c-\varepsilon_s)(\varepsilon_h-\varepsilon_s)\sigma}$$

$$B = \frac{2\varepsilon_h(\varepsilon_c+\varepsilon_s)}{(\varepsilon_c+\varepsilon_s)(\varepsilon_s+\varepsilon_h)-(\varepsilon_c-\varepsilon_s)(\varepsilon_h-\varepsilon_s)\sigma}$$

$$C = \frac{2\varepsilon_h(\varepsilon_c-\varepsilon_s)}{(\varepsilon_c+\varepsilon_s)(\varepsilon_s+\varepsilon_h)-(\varepsilon_c-\varepsilon_s)(\varepsilon_h-\varepsilon_s)\sigma}$$

$$G = \frac{(\varepsilon_c+\varepsilon_s)(\varepsilon_s-\varepsilon_h)+(\varepsilon_c-\varepsilon_s)(\varepsilon_s+\varepsilon_h)\sigma}{(\varepsilon_c+\varepsilon_s)(\varepsilon_s+\varepsilon_h)-(\varepsilon_c-\varepsilon_s)(\varepsilon_h-\varepsilon_s)\sigma}$$

The uniform electric field $\mathbf{E}_o$ inside the metallic particle is

$$\mathbf{E}_c = \frac{4\varepsilon_h\varepsilon_s}{(\varepsilon_c+\varepsilon_s)(\varepsilon_s+\varepsilon_h)-(\varepsilon_c-\varepsilon_s)(\varepsilon_h-\varepsilon_s)\sigma}\mathbf{E}_0$$

The induced dipole moment of cylindrical metallic particle

$$\mathbf{p} = G\mathbf{E}_0(a+\delta)^2 = \frac{(\varepsilon_c+\varepsilon_s)(\varepsilon_s-\varepsilon_h)+(\varepsilon_c-\varepsilon_s)(\varepsilon_s+\varepsilon_h)\sigma}{(\varepsilon_c+\varepsilon_s)(\varepsilon_s+\varepsilon_h)-(\varepsilon_c-\varepsilon_s)(\varepsilon_h-\varepsilon_s)\sigma}(\delta+a)^2\mathbf{E}_0$$

4.2 Interfacial Layer Effect

Now, we consider the effect of interfacial layer through the limit $\delta \to 0$, while $\varepsilon_s \to \infty$ the interfacial property is concentrated on a surface of zero thickness and only the quantity $\delta\varepsilon_s$ is of significance, we take $I = lim_{\delta\to 0,\varepsilon_s\to\infty}\delta\varepsilon_s$

To characterize the interface between particles and dielectric host, I is the interfacial factor. It is a mixture of metal and dielectric, since I is a complex number, and I is also complex quantity. But, the real part of the dielectric function of the metallic particle is always negative number; whereas the imaginary part is a small, positive one. When I is taken as a negative (or positive) value, the interface exhibits metal-like or dielectric-like; and I=0 corresponds to the perfect interface. If we consider $\eta = \frac{\varepsilon_s}{\varepsilon_c(E_c)}$and $\sigma = (\frac{a}{a+\delta})^2$

The induced dipole moment of our system becomes

$$G\mathbf{E}_0 = \mathbf{p} = \frac{q(\mathbf{E}_c)-\varepsilon_h}{q(\mathbf{E}_c)+\varepsilon_h}(a+\delta)^2\mathbf{E}_0 \tag{4.2.1}$$

with $q(\mathbf{E}_c) = k\varepsilon_c(\mathbf{E}_c)$,where

$$K = \frac{(1+\sigma)\eta+(1-\sigma)\eta^2}{(1-\sigma)+(1+\sigma)\eta} \tag{4.2.2}$$

By taking the limit of an interfacial factor

$$k = 1 + \frac{I}{a(\varepsilon_c)} \tag{4.2.3}$$

$$Z(\mathbf{E}_c) = K\varepsilon_c(\mathbf{E}_c) = \varepsilon_c(\mathbf{E}_c) + \frac{I}{a} \tag{4.2.4}$$

$$G = \frac{(1 + \frac{I}{a\varepsilon_c})\varepsilon_c - \varepsilon_h}{(1 + \frac{I}{a\varepsilon_c})\varepsilon_c + \varepsilon_h}(a + \delta)^2 \tag{4.2.5}$$

Assuming that the inclusions are not too densely packed, the uniform field inside the nonlinear particle is

$$\mathbf{E}_c = \frac{2\varepsilon_h \langle \mathbf{E}_1 \rangle}{\varepsilon_c + \varepsilon_h + \frac{1}{a}} \tag{4.2.6}$$

For non-dilute mixture of nonlinear cylindrical metallic particles

$$\mathbf{E}_c = \frac{2\varepsilon_h \mathbf{E}_0}{(\varepsilon_c + \varepsilon_h + \frac{I}{a})(1 - \frac{fG}{a^2})} \tag{4.2.7}$$

By using equations (4.2.1),(4.2.2) and (4.2.3) we get

$$Z^3 + \alpha Z^2 + \beta Z + \gamma = 0$$

where

$$Z = \chi_c^{(3)} |\mathbf{E}_c|^2$$

$$\alpha = \frac{2[(1-f)\varepsilon_{c1} + (1+f)\varepsilon_h + \frac{I}{a}]}{1-f}$$

$$\beta = \frac{[(1-f)\varepsilon_{c1} + (1+f)\varepsilon_h + \frac{I}{a}]^2 + [(1-f)\varepsilon_{c2}]^2}{(1-f)^2}$$

$$\gamma = \frac{-4\varepsilon_h^2 \chi_c^{(3)} |\mathbf{E}_0|^2}{(1-f)^2}$$

Here, $\varepsilon_{c1} (or \varepsilon_{c2})$ is real (or imaginary) part of ε_c Equation $Z^3 + \alpha Z^2 + \beta Z + \gamma = 0$ is a cubic equation in Z, and signifies bistability.

4.3 Interfacial layer dependence of optical bistability

We graphically analyzed the effect of interfacial layer for both metallic core and dielectric core and compared the threshold magnitude of bistability.For metallic core we took cylindrical gold with real and imaginary part of dielectric constant -13.0 and 1.04 respectively.However,for dielectric core we consider a system with real and imaginer part of the dielectric constant 6.0 and 2.04 respectively.one can observe that the interfacial layer plays an important role in bistable behavior. For the first model

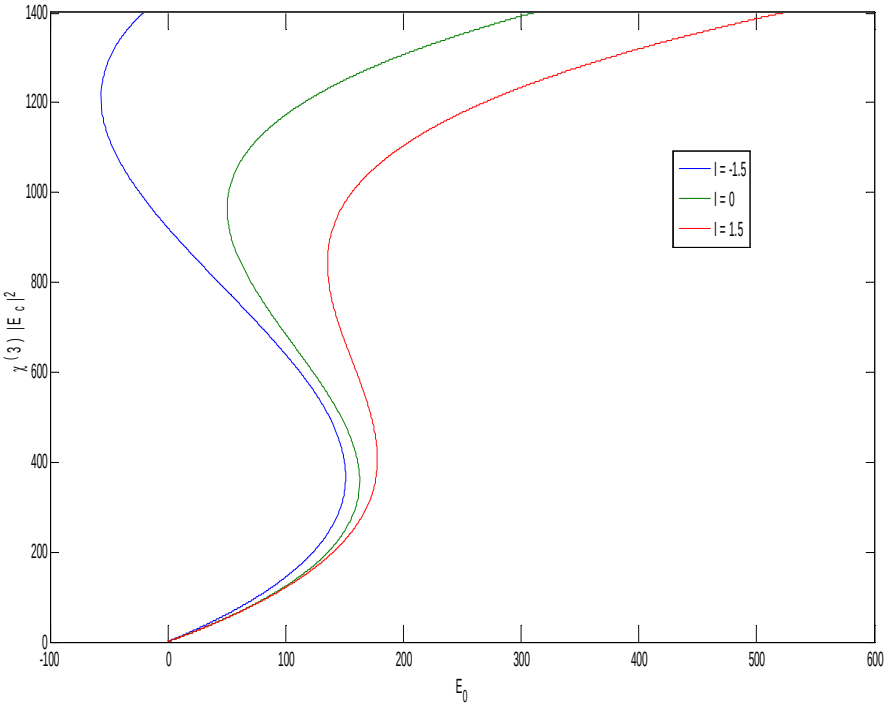

Figure 4.1: *Bistability curves for interfacial factors* $I = -1.5, 0, 1.5 and \chi_c^{(3)} = 10^{-8}$ *for the given values of dielectric constant and radius 4nm cylindrical metallic core*

the interfacial factor I changes from a negative value $I = -1.5$ to a positive value $I = 1.5$ that is from the metallic property to the dielectric property, the threshold value of the bistability decreases. From this one conclude that the dielectric like interfacial layer is favorable to reduce the threshold values.

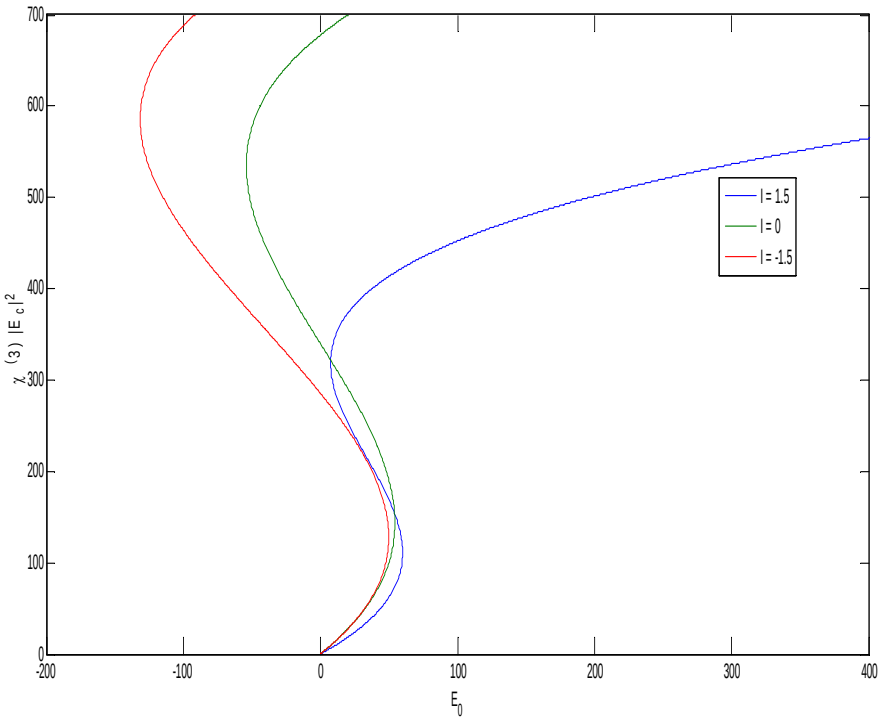

Figure 4.2: *Bistability curves for interfacial factors* $I = 1.5, 0, -1.5$ *and* $\chi_c^{(3)} = 10^{-8}$ *for the given values of dielectric constant and radius 4nm cylindrical metallic core*

However for the second model (dielectric core) the interfacial factor changes from positive value (dielectric behavior) to a negative value (metallic behavior) and the threshold value of the bistability increases. For the metallic like interfacial layer amplifies the threshold value of the bistability.

4.4 Size Dependence of Optical Bistability

In this part we analyzed the effect of size of the core and compared the threshold magnitude of the bistability. The threshold value of the optical bistability depends on the size or radius of the core, for these two models threshold bistablity for different radii is illustrated as shown the figures below.

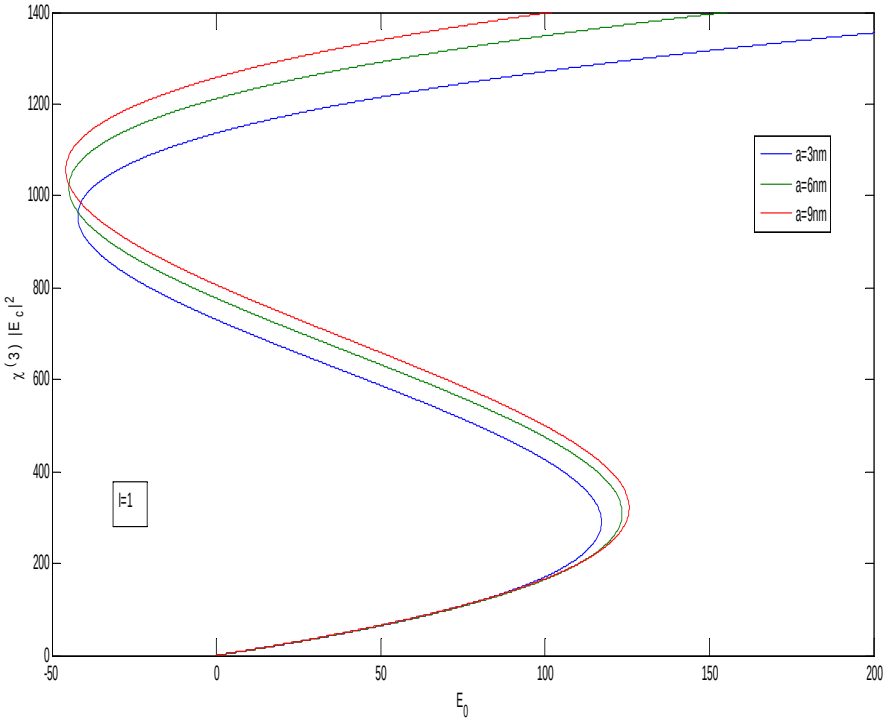

Figure 4.3: *Bistability curves for small radii* $a = 3nm, 6nm, 9nm, I = 1.5 and \chi_c^{(3)} = 10^{-8}$ *for the given values of dielectric constant and radius 4nm cylindrical metallic core*

For the first model (metallic core) the threshold values of the bistability increases with increasing the radius of the metallic core.

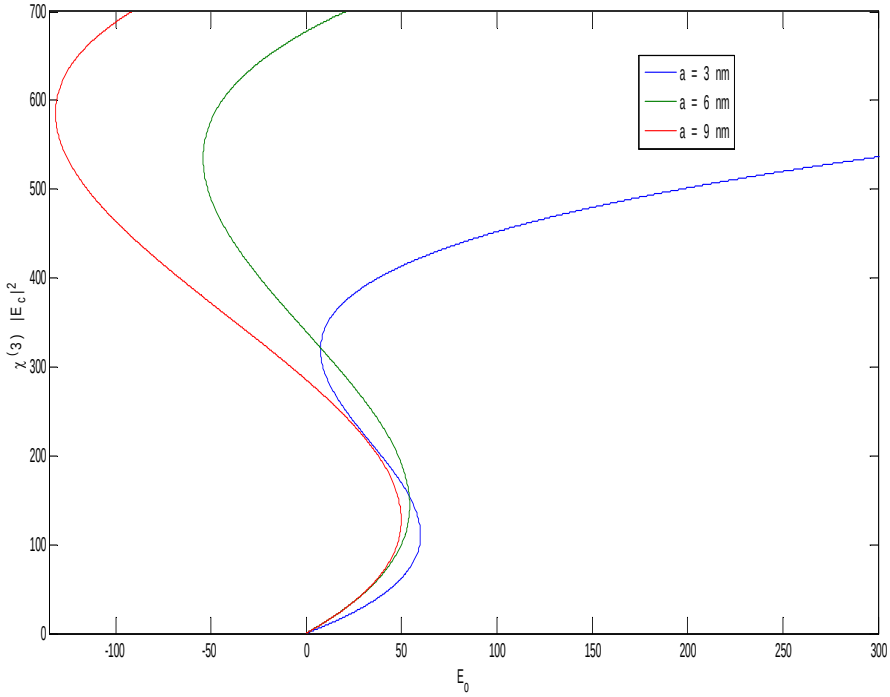

Figure 4.4: *Bistability curves for small radii* $a = 3nm, 6nm, 9nm, I = 1.5 and \chi_c^{(3)} = 10^{-8}$ *for the given values of dielectric constant and radius 4nm cylindrical dielectric core*

For the second model (dielectric core)the threshold value of the bistability decreases with the increase of the radius.

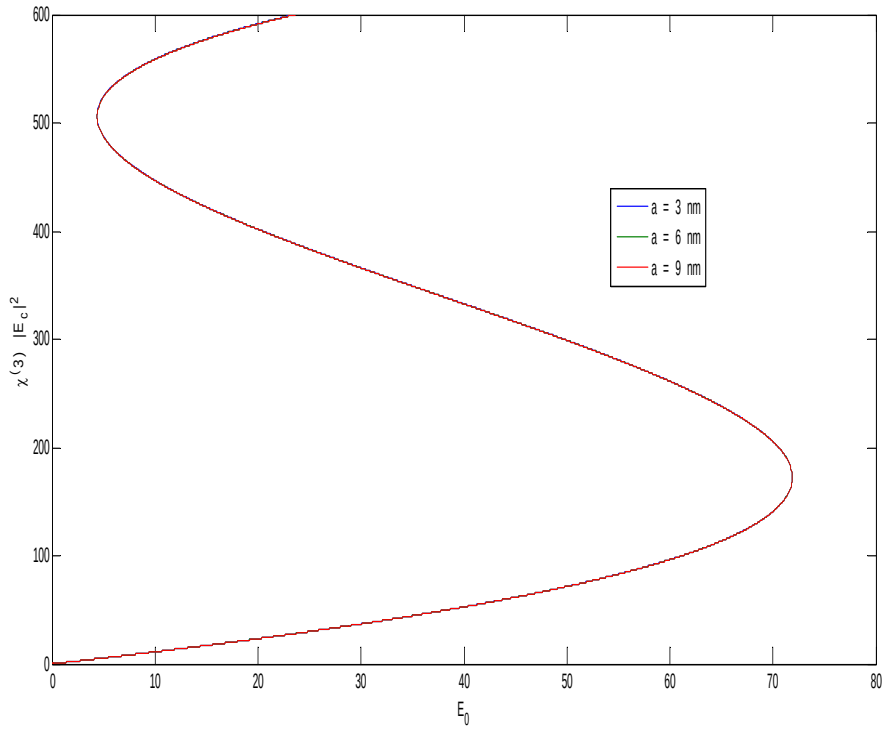

Figure 4.5: *Bistability curves for small radii* $a = 3nm, 6nm, 9nm, I = 1.5 and \chi_c^{(3)} = 10^{-8}$ *for the given values of dielectric constant and radius 4nm cylindrical metallic core*

From the above figure we conclude that, for perfect interfacial layer ($I = 0$) increment of the radius does not affect the threshold values of the bistablity as shown in figure 5.

4.5 The effect of interfacial layer on induced dipole moment

In figure 6 we observed the effect of interfacial layer on induced dipole moment. From the figure one can observe that as interfacial factor switches from negative

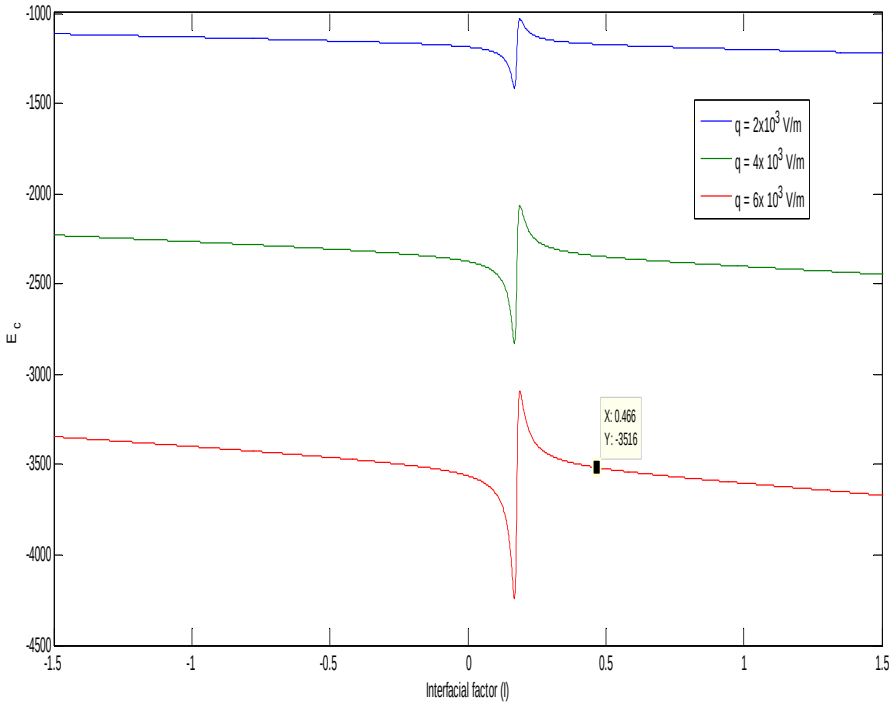

Figure 4.6: *Bistability curves for small radii* $a = 3nm, 6nm, 9nm, I = 1.5 and \chi_c^{(3)} = 10^{-8}$ *for the given values of dielectric constant and radius 4nm cylindrical metallic core*

value (metallic behavior) to a positive value (dielectric behavior) the absolute value of the local electric field increases in magnitude with respect to the magnitude of applied external electric field.

Chapter 5

Conclusion

Interfacial layer plays a great role in bistability behavior. As interfacial factor I changes from negative (metallic property) to a positive (dielectric property) the threshold value of the bistability decreases where as when I changes from positive (dielectric property) to a negative (metallic property) the threshold value increases. The interfacial layer also has effect on induced dipole moment: as interfacial factor switches from negative value to positive value the magnitude (absolute value), of the local electric field increases with respect to the magnitude of applied external electric field. More over, the effect of size on the bistability has a great impact: as the size a metallic core increases, the threshold value of the bistability increases, however as the size of the dielectric core increases the threshold value of the bistability decreases For perfect interfacial layer the size does not affect the bistability of the system.

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DECLARATION

I hereby declare that this Msc dissertation is my original work and has not been presented for a degree in any other University and that all source of materials used for the dissertation have been duly acknowledged.

Name:Sera Habtie

Signature: _____

email:hsera07@gmail.com

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

Name:Dr.Menberu Mengesha

Signature: _____

Place and date of submission:

Department of Physics

Jimma University

October,2015