OPTICAL PROPERTIES OF SMALL SPHERICAL PURE METAL IN PASSIVE AND ACTIVE HOST MATRICES

By

Girma Berga Kereta

A THESIS SUBMITTED TO THE DEPARTMENT OF PHYSICS PRESENTED IN PARTIAL FULFILLMENT OF REQUIREMENTS FOR THE DEGREE OF MASTER OF SCIENCE IN PHYSICS (CONDENSED MATTER PHYSICS)

WOLKITE UNIVERSITY

WOLKITE, ETHIOPIA

December 17, 2021

Wolkite University

Department of Physics

The undersigned hereby certify that they have read and recommend to the Department of Physics for acceptance of a **thesis** entitled "**Optical Properties of Small Spherical Pure Metal in Active and Passive Host Matrices**" by **Girma Berga** in partial fulfillment of the requirements for the degree of Master of Science in Physics with specialization in (Condensed Matter physics).

Date: December, 2021

Advisor: ______________________

Dr. Sisay Shewamare

Internal Examiner: ______________________

External Examiner: ______________________

Chairperson: ______________________

Name of chair person

______________________ __________ __________

SGS Approval Signature Date

Final approval and acceptance of the thesis is contingent upon the submission of the final copy of the thesis to the School of Graduate Studies (SGS) through the Department/School of Graduate Committee (DGC/SGC) of the candidate's department.

Stamp of SGS Date: ______________________

Wolkite University

School of Graduate Studies

As members of the Board of Examiners of the final Master's degree open defense, we certify that we have read and evaluated the **thesis** prepared by **Girma Berga** under the title "**Optical Properties of Small Spherical Pure Metal in Active and Passive Host Matrices**" and recommend that it be accepted as fulfilling the thesis requirement for the degree of Master of Science in Physics with Specialization in **Condensed Matter Physics**.

Name of the Chairperson	Signature	Date
Name of Internal Examiner	Signature	Date
Name of External Examiner	Signature	Date

Final approval and acceptance of the thesis is contingent upon the submission of the final copy of the thesis to the School of Graduate Studies (SGS) through the Department/School Graduate Committee (DGC/SGC) of the candidate's department.

Thesis approved by:

DGC/SGC	Signature	Date

Wolkite University

School of Graduate Studies

Certification of the final Thesis

I hereby certify that all the corrections and recommendation suggested by the Board of Examiners are incorporated into the final Thesis entitled **Optical Properties of Small Spherical Pure Metal in Active and Passive Host Matrix** by **Girma Berga**.

Date: December, 2021

Name of the Designate	Signature	Date

Stamp of SGS Date: ____________________

WOLKITE UNIVERSITY

Date: **December, 2021**

Author: **Girma Berga Kereta**

Title: **Optical Properties of Small Spherical Pure Metal in Active and Passive Host Matrix**

Department: **Physics**

Degree: **MSc.**

Convocation: **December**

Year: **2021**

Permission is herewith granted to Wolkite University to circulate and to have copied for non-commercial purposes, at its discretion, the above title upon the request of individuals or institutions.

Signature of Author

Acknowledgment

First of all I would like to thanks my God for his help throughout this work.

Next, I would like to express my gratitude, appreciation and respect to my advisor, Dr. Sisay Shewamare for his guidance, invaluable advice and kindness throughout this study.

Besides my advisor, I would like to thank Wolkite University physics department head person and all accademic staff members for their encouragement , motivation and material support.

Next, I would like to thank my brothers Bzuayehu, Mesfin, Adem and Mintesnot Alemu for their financial support and motivation.

Next, I would like to thank my family members Feleku, Fikirte, Werkie, Sintayehu, Senayt, Kidist, Teslima, Zuneyra, Tsegaab, Amar, Yohannes and my wife Meseret Mulugeta.

I would also like to thank all of my friends Endalkachew, Ashenafi, Murad, Tessema, Reshad for their sharing of ideas on the progress of the coureses. Moreover, I would like to acknowledge Shewa Getachew for what he has done in supporting me technically when writng the thesis with softwares LATEX and MATLB.

The last but not the least, I would like to thank Wolkite University, Guraghe Zone Education department, Wolkite City Administration Education Sector, and Yaberus General Secondary and Preparatory School for helping me to achieve my goal.

Abstract

In this thesis, we have studied the optical properties of small spherical pure metal in active and passive host matrix. One of the optical properties we have investigated by this work is the local field enhancement factor for small spherical pure metal in passive and active host matrix. The results show that for small spherical pure metal there is only one maxima of the local field enhancement factor in both the passive and active host matrix. We present an analytical and numerical method for optical bistability of small spherical pure metal in passive and active host matrix. Using the derived analytical and numerical results we calculated the cubic equation of the optical bistability of small spherical pure metal embedded in passive and active host matrix. To observe considerable differences on the onset and offset parts of the plots of optical bistability for the active host matrix, we took the integer multiples for the imaginary part of dielectric function of the host matrix (ϵ''_h), (i.e. $\epsilon''_h = -2$, $\epsilon''_h = -3$ and $\epsilon''_h = -4$). The third main target of study was to derive the analytical and numerical results of the real and imaginary parts of refractive indices for small spherical pure metal embedded in a host matrix. We have plotted the graphs of the real and imaginary parts of the refractive indices of the analytical and numerical results for small spherical pure metal in active and passive host matrix. The numerical and analytical results show that the local field enhancement factor is extremely enhanced, the optical bistability activation and the real and imaginary parts of the refactive index are increased when the small spherical pure metal is embedded in active host matrix (i.e. the natural or original property of the imaginary part of the dilectric function of the host matrix is affected by applying additionalexternal dilectric function on the host matrix).

Contents

List of Figures

List of Tables

List of Abbrivations and Symbols

DF – Dielectric Function

EM – Electro-Magnetic

LSP – Localized Surface Plasmons

OB – Optical Bistability

SP – Surface Plasmon

$\vec{B}$ – Magnetic induction

$\vec{D}$ – Electric displacement

$\vec{E}$ – Electric field strength

$\vec{E}_{Loc}$ – Local electric field

$\vec{H}$ – Magnetic field strength, or magnetic intensity

$\vec{J}$ – Current density

N – The concentration of electrons

n – Complex refractive index

n' – Real part of refractive index

n' – Imaginary part of refractive index

f – Volume fill fraction of the spherical metal

$\vec{M}$ – Magnetization

$\vec{P}$ – Polarization

ϵ – Dielectric function/permitivity

ϵ_h – Dielectric function of the host matrix

ϵ_h' – real part of the DF of the host matrix

ϵ_h'' – imaginary part of the DF of the host matrix

ϵ_m – Dielectric function of small spherical pure metal

ϵ'_m – real part of the DF of small spherical pure metal

ϵ''_m – imaginary part of the DF of small spherical pure metal

α – dimensionless polarizability

α' – real part of dimensionless polarizability

α'' – imaginary part of dimensionless polarizability

μ – permeability of medium

ν – Collision frequency

ρ – charge density

ω – frequency of incident wave

ω_p – plasma frequency

γ – gamma

χ – electric susceptibility

χ' – real part of susceptibility

χ'' – imaginary part of susceptibility

z – dimensionless frequency

Chapter 1

Introduction

1.1 Background of the study

The linear and nonlinear response of the medium strongly effects on the propagation of electromagnetic wave in the optical material and can even result in the permanent modification of its physical properties. In turn, the linear and nonlinear optical features of composite materials with metal nanostructures are dominated by surface plasma oscillations. The fact that the surface plasmon (SP) strongly depends on size, shape, distribution of metal nanoparticles as well as on surrounding dielectric matrix offers an opportunity for manufacturing of new promising nonlinear materials, nano devices and optical element..
Composite materials, consisting of small, nonlinear metallic particles (existing in the shape of sphere or cylinder) randomly embedded in a linear dielectric host, are well known for their complex responses to incident light fields [1, 2].
Nonlinear optical effects are useful for many different applications. Some examples of areas of science and technology in which nonlinear optics discussed in nonlinear microscopy [3] ultrafast laser systems, wavelength conversion [4] , material processing [5], optical routing and switching based on optically induced bistability both on networks and on chips [6].
Metals have a fast and strong nonlinear response [7] and may be good candidates for nonlinear optical applications if they are combined with dielectrics [8, 9].

Combining metals and dielectrics has two main purposes. One is allowing light to enter more deeply into metals, and the other is achieving light localization which in turn leads to an enhanced nonlinear response. An outstanding property of metal nanoparticle is the presence of extinction bands in the visible or infrared that result from the so called plasmon resonances these resonances do not exist in bulk metals, can be interpreted as a result of the confinement of free electrons in a space smaller than one wavelength of the light and controlled by changing the shape of the nanoparticle and its orientation with respect to the electric field. Plasmon resonances in metal nanoparticles can be put to many practical uses for example they enhance the sensitivity of optical sensors [10] and show great potential to be applied in the therapy of cancer [11]. Plasmon resonances are also the reason why the nonlinear optical properties of nanoparticle systems are enhanced with respect to those of bulk metals [12].

Optical properties of matter are consequences of how it reflect, transmit, and absorb visible light. Noble metal nanoparticles attract great interest because of their outstanding optical properties which arise from their ability to resonate with light. Resonant excitation of localized surface plasmons (LSP) on nanoparticles give rise to a variety of effects, such as frequency-dependent absorptions [13].

In optics of metals, strong frequency dependence of dielectric function ($\epsilon(\omega)$) is of basic importance in shaping their optical and transport properties. Significance of indexes of refraction of noble metals in basic issues and applications has been a motivation to many experimental studies intended to increase the accuracy of measurements of their frequency dependence A. Derkachova et.al [14, 15].

1.2 Statement of the problem

Metals have a fast and strong nonlinear response and may be good candidates for nonlinear optical applications if they are combined with dielectrics. In different research works, the application of the nonlinear optical materials are observed by affecting the composite properties of the pure metal. In this thesis we are studied the impact of the surrounding the active and passive matrix without consideration of the composite properties of the pure metal.

1.3 Objectives

1.3.1 General objectives

The general objective of this paper is to study the optical properties of small spherical pure metal in a passive and active host matrix analytically and numerically.

1.3.2 Specific objectives

The specific objectives of this study are:

- To study the local field enhancement of small spherical pure metal in a passive and active host matrix analytically and numerically.
- To study optical bistability of small spherical pure metal in a passive and active host matrix analytically and numerically.
- To study the refractive-index of Small Spherical pure metal in a passive and active host matrix analytically and numerically.

1.4 Significance of the study

The linear and nonlinear response of the medium strongly effect on the propagation of electromagnetic wave in the optical material and can even result the permanent modification of its physical properties, the linear and nonlinear optical features of the materials are dominated by surface plasma oscillations and the surrounding dielectric properties. Therefore, this study would contribute for the advancement of optical properties of the surrounding dielectric materials which provide good opportunity on knowledge and application for the researcher and others who will be interested to do more investigation on the field.

1.5 Thesis outline

Chapter two introduces related literature reviews, from electrodynamical properties of materials to optical properties of metals.
Chapter three describes the enhancement factor of the local field of small spherical pure metal in passive and active host matrix, there, the highest enhancement local field factor is achieved as the original property of the dielectric function of the host matrix is varying.
Chapter four presents the optical bistability of small spherical pure metal in passive and active host matrix, here highest activation of optical bistability is achievd with varying the natural property of the host matrix.
Chapter five presents the refractive index of small spherical pure metal in passive and active host matrix, similarly the real and imaginary parts of the refractive index is maximum in active host matrix.
conclusions are drawn in chapter six.

Chapter 2

Literature Review

2.1 Electrodynamical Properties of Materials

2.1.1 Maxwell Equations and Constitutive Relation

All the electromagnetic phenomena are governed by Maxwell's equations, which are a set of equations describing the interrelationship between fields, sources, and materials properties. The electromagnetic properties of a material are determined by two material parameters: the permittivity ϵ and permeability μ, describing the coupling of a material to the electric and magnetic components of an electromagnetic wave, respectively. 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. These constitutive relations in cgs units are:

$$\vec{D}(r,t) = \vec{E}(r,t) + 4\pi\vec{P}(r,t), \tag{2.1}$$

$$\vec{B}(r,t) = \vec{H}(r,t) + 4\pi\vec{M}(r,t) \tag{2.2}$$

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.

$$\nabla \cdot \vec{D}(r,t) = 4\pi\rho(r,t), \tag{2.3}$$

$$\nabla \cdot \vec{B}(r,t) = 0, \tag{2.4}$$

$$\nabla \times \vec{E}(r,t) = \frac{1}{c}\frac{\partial \vec{B}(r,t)}{\partial t}, \tag{2.5}$$

$$\nabla \times \vec{H}(r,t) = \frac{4\pi}{c}\vec{J}(r,t) + \frac{1}{c}\frac{\partial \vec{D}(r,t)}{\partial t}, \tag{2.6}$$

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. The current density also given by the following equation

$$J = \sigma E \tag{2.7}$$

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, Eqn. (2.1)

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 [16].

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 [17]. 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 of 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; where as the nonlinear optical response of a medium depends no longer linearly on the strength of the applied optical field, $\vec{E}(t)$. In linear optics, the polarization $\vec{P}(\mathrm{t})$ of a medium is defined by

$$\vec{P}(t) = \chi^{(1)}\vec{E}(t) \tag{2.8}$$

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 $\vec{P}(\mathrm{t})$ as a power series in the field strength $\vec{E}(\mathrm{t})$ at

$$\vec{P}(t) = \chi^{(1)}\vec{E}(t) + \chi^{(2)}\vec{E}^2(t) + \chi^{(3)}\vec{E}^3(t) + ... = \vec{P}^{(1)}(t) + \vec{P}^{(2)}(t) + \vec{P}^{(3)}(t) + ... \tag{2.9}$$

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 $10^{15} cm^3$ [18], the influence of the local field effects become significant and must be taken in to 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:

$$\vec{E}_{Loc} = L\vec{E} \tag{2.10}$$

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

$$\vec{E}_{Loc} = \vec{E} + \frac{4\pi}{3}\vec{P} \tag{2.11}$$

Eqn. (2.10) is commonly known as the "Lorentz local field" [18]. The concept of a local field was originally introduced by Lorentz [19] and it is further derive the Lorentz-Lorenz (or Clausius-Mossotti) relation for the dielectric permittivity ϵ and microscopic polarizability α. Let us assume for now that the medium is lossless and dispersionless. We represent the dipole moment induced in a typical molecule (or atom) of the medium as

$$P = \alpha E_{Loc} \tag{2.12}$$

The macroscopic polarization of the medium is derived over the investigated volume V , as follows [20]:

$$P(\omega) = \frac{1}{v}\int_v p(\omega)dv = N\alpha(\omega)E \tag{2.13}$$

The macroscopic polarization of the material is also given by the equation

$$P = Np \tag{2.14}$$

where N denotes molecular (or atomic) number density. Using equations (2.12) and (2.13), we find that the polarization and macroscopic field are related by

$$P = N\alpha[E + \frac{4\pi}{3}P] \tag{2.15}$$

We assume the polarization P to be linear in the average field:

$$P = \chi^{(1)}E \tag{2.16}$$

where $\chi^{(1)}$ is the linear optical susceptibility of the medium. Substituting the expressions (2.14) and (2.15), solving for $\chi^{(1)}$, and eliminating the field $\vec{E}$, we find that

$$\chi^{(1)} = \frac{N\alpha}{1 - \frac{4\pi}{3}N\alpha} \tag{2.17}$$

Expressing the optical susceptibility of the medium

$$\chi^{(1)} = \frac{\epsilon^{(1)} - 1}{4\pi} \tag{2.18}$$

$\epsilon^{(1)}$ is the dielectric permittivity of the medium, we obtain the well-known Lorentz-Lorenz as:

$$\frac{\epsilon^{(1)} - 1}{\epsilon^{(1)} + 2} = \frac{4\pi}{3}N\alpha \tag{2.19}$$

Through rearrangements of Eqn. (2.18) we can express the linear susceptibility as

$$\chi^{(1)} = \frac{\epsilon^{(1)} + 2}{3} N\alpha \tag{2.20}$$

Substituting the expression (2.19) in to (2.15) then (2.15) in to (2.13) and using the relationship $P(\omega) = \alpha(\omega)E_{Loc}(\omega)$ between the local field and dipole moment, we obtain the equation relating the local field to the average field.

$$E_{Loc} = \frac{\epsilon^{(1)} + 2}{3} E \tag{2.21}$$

where,

$$L = \frac{\epsilon^{(1)} + 2}{3} \tag{2.22}$$

is known in the literature as the Lorentz local-field correction factor [21].
The expression (2.20) for the local-field correction factor is valid in the case of homogeneous media, where all the particles (molecules or atoms) are of the same sort. It is also valid in materials where the emitters enter as inclusions that do not influence the correlation between the host molecules or atoms [22].

2.3 Optical Properties in Metals

2.3.1 Optical Enhancement in Metals

A characteristic property of metal nanoparticles is the enhancement of the local electromagnetic fields near the surface of the particles.
There are different factors which can enhance the local fields: The plasmon resonance,always enhances the local field near the resonance frequency. At the resonsnce, the absorption and scattering cross sections are enhanced, and the local electromagnet fields in the structure also enhanced. For the large resonance enhancement, a narrow width of the resonance

is advantageous, as it typically leads a strong response at the center of the resonance. The strongest local fields are thus obtained usually close to the resonance. In addition tiny features, in the particles, such as sharp tips small gaps can further enhance the fields [23] and combining a metal with dielectric materials also enhances the local field.

2.3.2 Optical Bistability

The concept of optical bistable switching or bistability has been explored to implement all-optical transistors, switches, logical gates, and memory. It has been employed widely in all-optical signal processing. In systems having optical bistability, the output intensity is a strong nonlinear function of the input intensity. These bistable systems might even display a hysteresis loop. One of the performance indicators of a bistable system is its ability to operate at low input intensity [24].

Optical bistability can be achieved in composite materials made of a nonlinear dielectric and a metal of which the dielectric constant has a negative real part and a small imaginary part [25].

A system is said to be optically bistable if it has two output states I_T for the same value of the input I_I over some range of input values. Thus a system having the transmission curve of Fig. 2.1 is said to be bistable between $I_\downarrow$ and $I_\uparrow$. Such a system is clearly nonlinear, i.e., I_T is not just a multiplicative constant times I_I. In fact, if I_I is between $I_\downarrow$ and $I_\uparrow$, knowing I_I does not reveal I_T. Nonlinearity alone is insufficient to assure bistability. It is feedback that permits the nonlinear transmission to be multivalued, i.e., bistable. It is this restricted definition of optical bistability defined by Fig. 2.1, with the nonlinear medium unexcited, that is adopted here. This definition implies that the bistable system can be cycled completely and repeatedly by varying the input intensity [26].

A powerful principle that could be explored to implement all-optical transistors, switches, logical gates, and memory, is the concept of optical bistability. In systems that display optical bistability, the outgoing intensity is a strongly non-linear function of the input

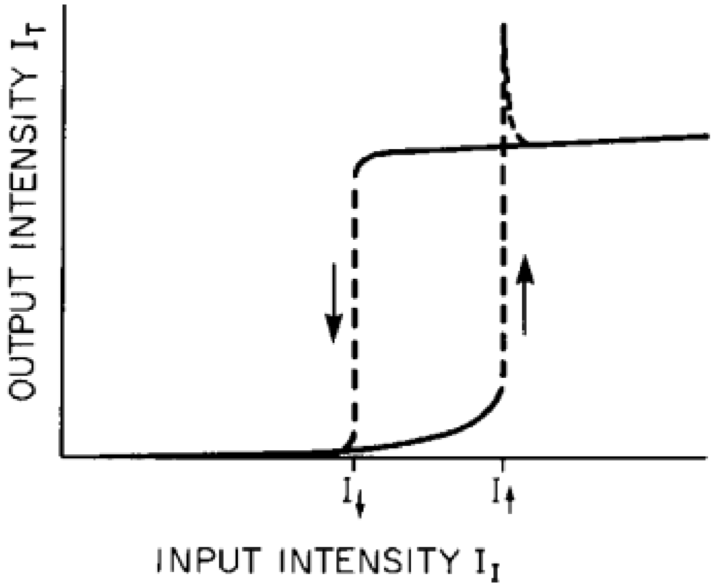

Figure 2.1: Characteristic curve for an optical bistable system.

intensity, and might even display a hysteresis loop [27].

2.3.3 Refractive Index

The refractive index is one of the most important parameters for an optical medium, defined as $n = c/v$, it measures the comparative velocity of light in different media. When a light beam travels across the boundary between two different materials, it bends owing to the change in refractive index at the interface. This phenomenon, refraction, gives the reason why a water pool appears shallower than it actually is, why a straw placed partially in water at a slant seems to bend towards the surface, and why people can use eyeglasses to adjust the path of light in front of their eyes and alleviate the effects of conditions such as myopia [28]. The concept of refractive index engineering of materials was raised more than 100 years ago, and was described by the Clausius–Mossotti relation. Nowadays, the rapid development of nanotechnology opens a new gate to achieving precise engineering of materials' refractive indices and realizing novel applications. Refractive index (n) is one of the most important physical quantities of the materials that directly influences the device's performance, since it determines how the light interacts with the materials and devices. It describes how light or electromagnetic waves propagate through an optical medium. If a material absorbs light, the refractive index equation can be modified using a complex term: $n = n' + in''$, where n is the complex refractive index, n' is the real refractive index, n'' is the imaginary part of the complex refractive index and it is the extinction coefficient, which relates to the amount of light that is absorbed. Different optical phenomena such as reflection, refraction, interference and total internal reflections can happen when light travels through one medium to another due to the refractive index difference [29].

Increasing the refractive index available for optical and nanophotonic systems opens new vistas for design: applications ranging from broadband metalenses to ultrathin photovoltaics to high-quality-factor resonators, higher index directly leads to better devices with greater functionality [30]. In the case of metal dielectric composite media, their application can practically be limited by absorbtion of incident electromagnetic radiation due to the presence of metal components. In a number of papers [31,32], it was proposed to use active (amplifying) host matrix in order to compensate absorption at metallic inclusion.

In our thesis, we also think to affect the imaginary part of the dielectric function of the host matrix by applying external dielectric function, this resembles increasing the optical density of the small spherical pure metal embedded in the host matrix, to compensate the refracive index.

The index represented by n and given by the square root of the dielectric constant (ε), which is in turn related to the atomic polarizability (α) using

$$D = \varepsilon E \tag{2.23}$$

$$D = E + 4\pi P \tag{2.24}$$

$$P = \alpha E \tag{2.25}$$

Considering the above equations, (2.23) to (2.25), the complex dielectric constant is given by [33]

$$\varepsilon = 1 + 4\pi\alpha \tag{2.26}$$

and the refractive index is given by [28],

$$n = \sqrt{\varepsilon} \tag{2.27}$$

Chapter 3

Local field Enhancement for Small Spherical Pure Metal in Passive and Active Host Matrix

Nonlinear optical materials have many interesting phenomena that have potential uses in optical devices and technology. However, in the current state, nonlinear optical materials produce very weak nonlinear optical responses, requiring large laser intensity to realize nonlinear phenomena. For applications of nonlinear optical phenomena to be used in realistic devices, the overall optical nonlinearity of materials must be significantly enhanced by many orders of magnitude.

To support the enhancement of the small metallic particle in a host matrix there are different mechanisms such as using composite, but in our thesis work, we are thinking to affect the imaginary part of the dielctric function of the host matrix by appliying additional dielectric function to it.

A host matrix is a medium with a complex dielectric function (ϵ_h), in which we put a small spherical pure metal whose optical property is to be studied. The property of the dielectric functuon of the host matrix will be affected by varying the imaginary part of its dielectric function. This phenomenon resembles as that of increasing the optical density of small spherical pure metal in the host matrix.

Then this host matrix is said to be active host matrix. From the field-dependent permittivity definition of optical nonlinear materials, it is clear that, there are two pathways for enhancement of nonlinearity. The first approach is the microscopic manipulation of materials that generate enhanced nonlinearity that do not occur in nature. From the microscopic approach, the goal is to yield a larger $\chi^{(2)}$ or $\chi^{(3)}$ in the nonlinear material of interest. For this to work, suitable The second approach is the macroscopic organization of materials in structures that generate enhanced nonlinearity. For the macroscopic approach, the goals is to yield a larger $|E|$ or $|E|^2$ within the nonlinear material of interest.

3.1 Enhancement factor in Passive and Active Host Matrix

Let us consider a small spherical pure metal embedded in a linear host matrix. In the

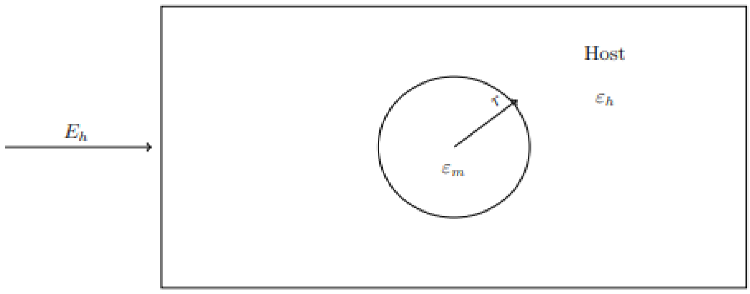

Figure 3.1: Applied field (E_h) incidence to a small spherical pure metal of radius r and DF ϵ_m embedded in a rectangular host matrix with DF of ϵ_h.

electrostatic approximation, when a wave length of the incident electromagnetic radiation is much greater than a typical size of the nanoparticle, the distribution of the electric potential in the system is described by the following expressions [34, 35].

$$\phi_m = -AE_h r cos\theta, \qquad (r < r_1) \qquad (3.1)$$

$$\phi_h = -E_h(r - \frac{B}{r^2})cos\theta, \qquad (r > r_1) \qquad (3.2)$$

They are the solutions of the Laplace equations in small spherical pure metal and in the host matrix respectively. Here ϕ is potential, 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 nanoparticle and the z axis is along E_h), A and B are unknown coefficients or constants, r is the radius of the small spherical pure metal.

Applying the continuity conditions of the potential at the boundaries of the small spherical pure metal, we get [36],

$$A = 1 - \frac{B}{{r_1}^3} \tag{3.3}$$

Again applying the continuity conditions of the displacement vector at the boundaries of the small spherical pure metal, we have [36]

$$\varepsilon_m A = \varepsilon_h (1 + \frac{2B}{{r_1}^3}) \tag{3.4}$$

From the respective results of the two boundary conditions, we obtain expressions for the unkown coefficients A and B.

$$A = \frac{3\varepsilon_h}{2\varepsilon_h + \varepsilon_m} , \tag{3.5}$$

$$B = \frac{\varepsilon_m - \varepsilon_h}{2\varepsilon_h + \varepsilon_m} \cdot {r_1}^3 \tag{3.6}$$

We consider that, the complex dielectric functions of the metal (ϵ_m) and that of the host matrix (ϵ_h) are expressed interms of their real and imaginary parts respectively

$$\varepsilon_m = \epsilon'_m + i\epsilon''_m , \quad \epsilon_h = \epsilon'_h + i\epsilon''_h \tag{3.7}$$

Inserting equation (3.7) in equation (3.5), we get,

$$A = \frac{3(\epsilon'_h + i\epsilon''_h)}{(2\epsilon'_h + \epsilon'_m) + i(2\epsilon''_h + \epsilon''_m)} \tag{3.8}$$

We find the modules of Eq. (3.8)

$$|A|^2 = \frac{9(\varepsilon'^2_h + \epsilon''^2_h)}{(2\epsilon'_h + \epsilon'_m)^2 + (2\epsilon''_h + \epsilon''_m)^2} \tag{3.9}$$

Equation (3.9) is known as local field enhancement factor. The dielectric function (DF) of the metal (ϵ_m) in the Drude form is,

$$\epsilon_m = \epsilon_\infty - \frac{{\omega_p}^2}{\omega(\omega + i\nu)} \tag{3.10}$$

The real and imaginary parts of DF of the metal are,

$$\epsilon'_m = \epsilon_\infty - \frac{\omega_p^2}{\omega^2 + \nu^2}\ , \qquad \epsilon''_m = \frac{\nu\omega_p^2}{\omega(\omega^2 + \nu^2)} \tag{3.11}$$

where ω_p is the plasma frequency given by ${\omega_p}^2 = Ne^2/(\epsilon_0 m)$, ω is the frequency of the incident wave, m is the mass of electron, N is the concentration of electron, setting $\frac{\omega}{\omega_p} = z$ and $\frac{\nu}{\omega_p} = \gamma$, therefore, the real and the imaginary part of equation (3.11) is given as,

$$\varepsilon'_m = \varepsilon_\infty - \frac{1}{z^2 + \gamma^2}\ , \quad \varepsilon''_m = \frac{\gamma}{z(z^2 + \gamma^2)} \tag{3.12}$$

3.1.1 The Enhancement Factor of Small Spherical Pure Metal in a Passive Host Matrix

Passive host matrix is a medium that is considering as a natural property of its dielectric function is not affected, because, there is no external dielectric function added to it, therefore, the imaginary part of the dielectric function of the host matrix is zero ($\epsilon''_h = 0$).

The plot of the enhancement factor stated on Eqn. (3.9) for a small spherical pure metal in a passive host matrix is shown below.

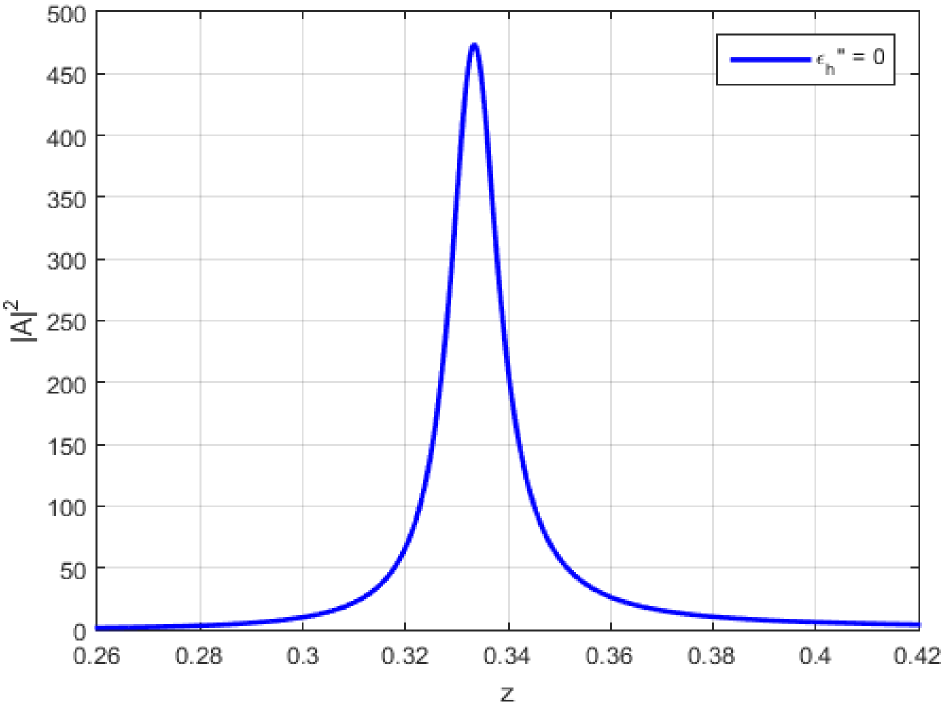

Figure 3.2: The plot of Enhancement factor $|A|^2$ versus dimensionless frequency z for a small Spherical pure metal in a passive host matrix ($\epsilon''_h = 0$) and with other numerical parameters $\nu = 1.68 \times 10^4$, $\omega_p = 1.46 \times 10^{16}$, $\gamma = 0.0115$, $\varepsilon_h = 2.5$, $\varepsilon_\infty = 4.5$.

From figure 3.2, we obtain the maximum value of amplitude, $|A|^2 = 473$ at the frequency of $z = 0.3334$.

3.1.2 The Enhancement Factor of Small Spherical Pure Metal in Active Host Matrix

Active host matrix is a medium that is considering as a natural property of its imaginary part of dielectric function is affected by applying additional dielectric function on it, therefore, the imaginary part of the dielectric function of the host matrix is different from zero ($\epsilon''_h \neq 0$). The imaginary part of the dielectric function of the host matrix in active host matrix has a negative sign because of that seems additional dielectric function is given to the host matrix.

The graph of the local field enhancement factor expressed on Eqn. (3.9) for asmall spherical pure metal in active host matrix for different values of ϵ_h'' is shown below.

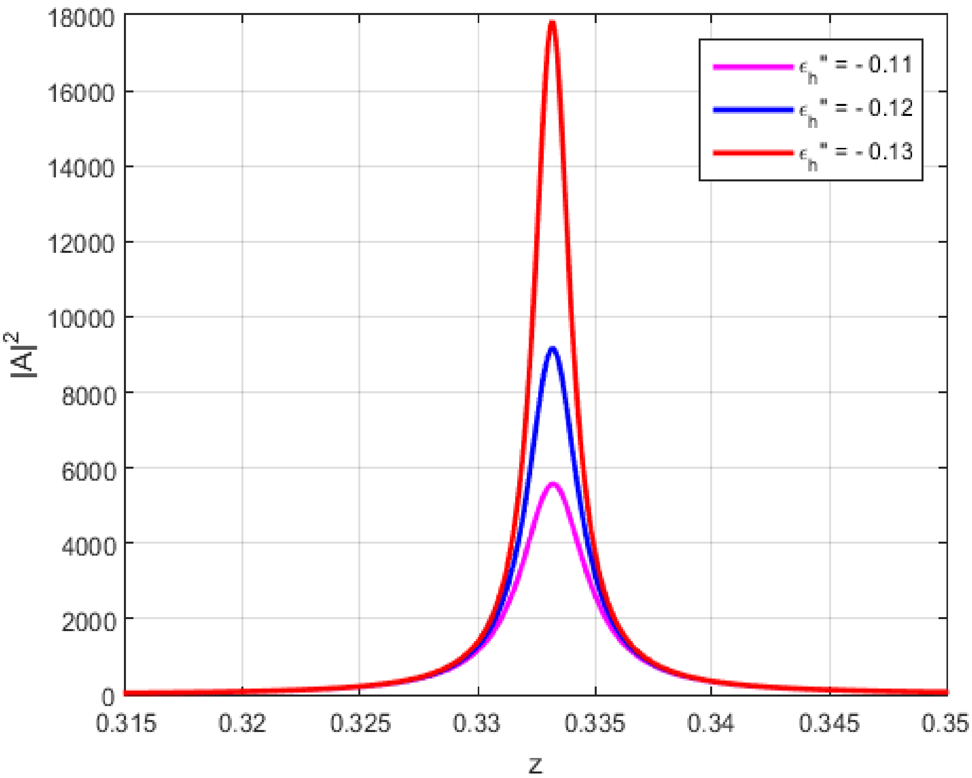

Figure 3.3: The enhancement factor versus the dimensionless frequency (z) for a small spherial pure silver metal in active host matrix ($\epsilon_h'' = -0.11$, $\epsilon_h'' = -0.12$, $\epsilon_h'' = -0.13$ respectively), with parameters, $\nu = 1.68 \times 10^4$, $\omega_p = 1.46 \times 10^{16}$, $\gamma = 0.0115$, $\varepsilon_h = 2.5$, $\varepsilon_\infty = 4.5$,.

From figure 3.3, we obtained that there are different maximum value of the local field enhancement factor $|A|^2$ for different values of active host matrix ($\epsilon_h'' < 0$). When affecting the natural property of the DF of the host matrix by applying additional dielectric function on it, the amplitude of the graph or $|A|^2$ increases.

Therefore, for the active host matrix ($\epsilon_h'' < o$), the local field Enhancement factor $|A|^2$ can be maximized by varying the original property of imaginary part of the dielectric function of the host matrix, this resembles like tha of increasing the density of the small spherical pure metal. The dependence between the magnitude of ϵ_h'' and $|A|^2$ is shown in the Table below.

ϵ_h''	$\lvert A\rvert^2$	z
-0.11	5568	0.3332
-0.12	9170	0.3332
-0.13	17840	0.3332

Table 3.1: The dependence between different values of ϵ_h'' enhancement factor ($|A|^2$) for a small spherical pure metal in active host matrix.

From Table 3.1, we ovserved that in active host matrix, as varying the imaginary part of the dielectric function of the host matrix by applying additional dielectric function on it, this is like increasing the density of the small spherical pure metal, then different amplitude of maximum values obtained at the same resonance frequency.

When $\epsilon_h'' = -0.11$, we obtained a maximum amplitude of 5568 at a frequency $z = 0.3332$, when $\epsilon_h'' = -0.12$, we obtained the maximum amplitude of 9170 at $z = 0.3332$ and when $\epsilon_h'' = -0.13$, we obtained the maximum amplitude of $|A|^2 = 1.784 \times 10^4$ at $z = 0.3332$.

Therefore, applying additional dielectric function on the host matrix,enhances the local electric field in the small spherical metal.

Chapter 4

Optical Bistability of Small Spherical Pure Metal in Passive and Active Host Matrix

Metallic nanoparticles have the ability to sustain coherent electron oscillations known as surface plasmon (SP) leading to electromagnetic fields confined to their surface. The formation of surface plasmon (SP) are due to the electric field of an incoming radiation that induces the formation of a dipole or a polarization of charges on the nanoparticle surface.

In this chapter we derived the analytical expression for the Optical bistability of a small Spherical pure metal in relation to the enhancement factor ($|A|^2$), that we discussed on the previous chapter.

Let the electromagnetic wave incident on a spherical pure metallic particle embedded in a dielectric host matrix. The dielectric function (DF) of the particle is assumed to depend on frequency ω and the local electric field $\vec{E}$ (inside the particle). Since the local field in nonlinear metal components cannot be solved exactly, we shall use mean-field approximation to estimate the field-dependent permittivity of metal,and may be presented in the form [16] $\epsilon(\omega, \vec{E})$ and it is used in the presence of small field $\chi|E|^2$.

$$\epsilon_m(\omega, \vec{E}) = \epsilon_m(\omega) + \chi(\omega)|\vec{E}|^2 \tag{4.1}$$

The real and imaginary parts of $\epsilon(\omega, \vec{E})$ in terms of χ' and χ'' are:

$$\varepsilon'_m = \epsilon'(\omega) + \chi'|\vec{E}|^2, \tag{4.2}$$

$$\varepsilon''_m = \varepsilon''(\omega) + \chi''|\vec{E}|^2. \tag{4.3}$$

Recalling eqn. (3.8) for the expression of the unknown coefficient A,

$$A = \frac{3(\varepsilon'_h + i\epsilon''_h)}{(2\varepsilon'_h + \varepsilon'_m) + i(2\epsilon''_h + \varepsilon''_m)} \tag{4.4}$$

The local field ($\vec{E}$) in the small spherical pure metal can be obtained with the help of relation

$$\vec{E} = A\vec{E}_h \tag{4.5}$$

In general, eqn. (4.4) 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 expressed using equations (4.2) and (4.3) for ϵ'_m and ϵ''_m, respectively, as

$$|A|^2 = \frac{9(\epsilon'^2_h + \epsilon''^2_h)}{(2\epsilon'_h + \epsilon'(\omega) + \chi'|E|^2)^2 + (2\epsilon''_h + \epsilon''(\omega) + \chi''|E|^2)^2} \tag{4.6}$$

We have taken the square modulus of eqn. (4.4) and multiplied both sides by χ, to get

$$\chi|E|^2 = |A|^2\chi|E_h|^2 \tag{4.7}$$

Let, $2\epsilon_h' + \epsilon'(\omega) = \tilde{\epsilon}$, $\quad 2\epsilon_h'' + \epsilon''(\omega) = \tilde{\epsilon}'$, $\quad \frac{2(\tilde{\epsilon}\chi' + \tilde{\epsilon}'\chi'')}{\chi} = a$,

and $\quad \tilde{\epsilon}^2 + \tilde{\epsilon}'^2 = b$, then, we get

$$|A|^2 = \frac{9({\epsilon_h'}^2 + \epsilon_h''^2)}{b + a\chi|E|^2 + (\chi|E|^2)^2} \tag{4.8}$$

Substituting equation (4.8) in to (4.7), we get

$$\chi|E|^2 = \frac{9({\epsilon_h'}^2 + \epsilon_h''^2)}{b + a\chi|E|^2 + (\chi|E|^2)^2}\chi|E_h|^2. \tag{4.9}$$

Let us introduce with the following denotations for, $X = \chi|E|^2$, and for $Y = \chi|E_h|^2$, using some rearrangements, we obtain the cubic equation for Y in terms of X,

$$X = \frac{9({\epsilon_h'}^2 + \epsilon_h''^2)}{b + aX + X^2}Y, \tag{4.10}$$

$$X^3 + aX^2 + bX = \eta Y, \tag{4.11}$$

where $\eta = 9(\epsilon_h'^2 + \epsilon_h''^2)$. Equation (4.11) determines a dependence of the local field (X) on the applied field (Y). Then, we express Y in terms of X to get equation (4.12).

$$Y = \frac{X^3 + aX^2 + bX}{9({\epsilon_h'}^2 + \epsilon_h''^2)} \tag{4.12}$$

Equation (4.12) is the mathematical expresion for the optical bistability.

4.1 Optical Bistability of Small Spherical Pure Metal in Passive Host Matrix

Referring subsection (3.1.1), in the passive host matrix, the natural property of the dielectric function of the host matrix is not affected by applying additional dielectric function, then $\epsilon_h'' = 0$.

The plot of Equation (4.12) for a small spherical pure metal in passive host matrix is shown on the figure below.

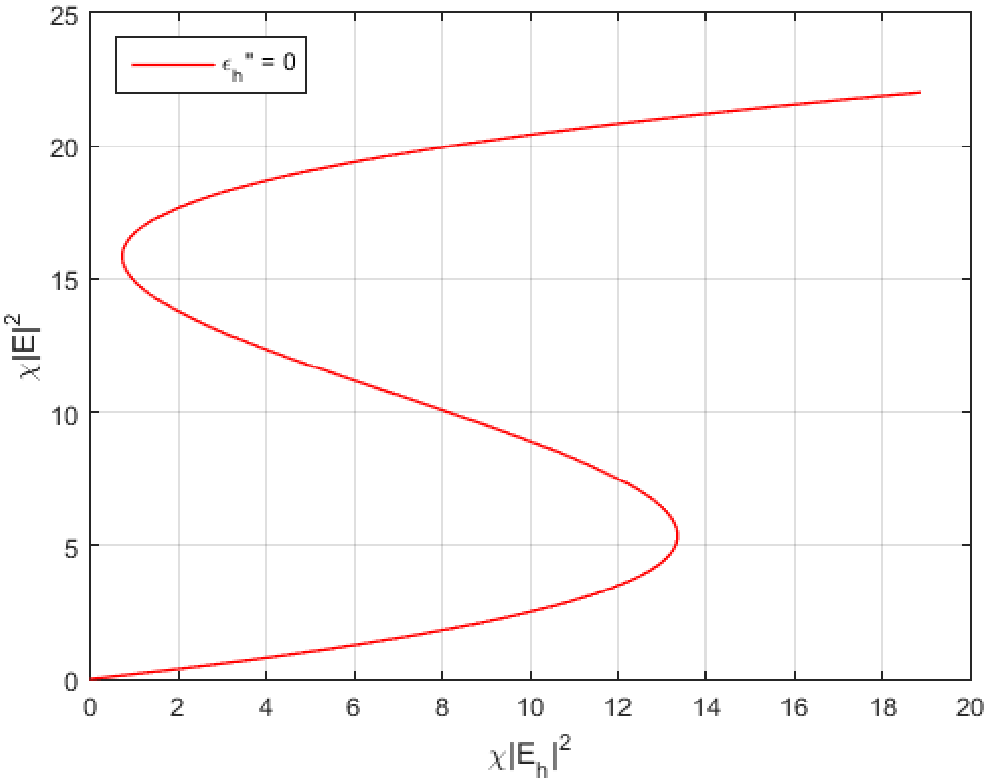

Figure 4.1: Local field $\chi|E|^2$ versus applied field $\chi|E_h|^2$ at frequency $z = 0.2$, for a small spherical pure metal in passive host matrix ($\epsilon_h'' = 0$) with parameters , $\epsilon_h' = 2.25$, $\gamma = 0.0115$.

ϵ_h''	Switching up $\chi\|E_h\|^2$ (onset)	Switching down $\chi\|E_h\|^2$ (offset)	rangewidth (threshold width) of $\chi\|E_h\|^2$ $(\Delta\chi\|E_h\|^2)$
0	13.35	0.716	12.634

Table 4.1: The switching up and down value of Optical Bistability for a small spherical pure metal in a passive host matrix.

The switching up and switching down input intensity of figure 4.1 is shown in Table 4 .1. There, we observed that the range width (threshold width) of $\chi|A|^2$ or $(\Delta\chi|E_h|^2)$=12.634, which is wider as compared to that of the active host matrix mentioned on the next section.

4.2 Optical Bistability of a Small Spherical Pure Metal in Active Host Matrix

Refering subsection 3.1.2, we have seen that in the active host matrix, the imaginary part of the dielectric function of host matrix (ϵ_h'') is affected by applying additional dielectric function on it, that is like that of increasing the density of the small spherical metallic particle, then, $\epsilon_h'' \neq 0$. In our mathematical calculation, to observe considerable differences on the onset and offset parts of the plots of optical bistability for the active host matrix, we took the integer values for the imaginary part of the dielectric function of the host matrix ϵ_h'', (i.e., $\epsilon_h'' = -2$, $\epsilon_h'' = -3$ and $\epsilon_h'' = -4$). The negative sign implies that additional dielectric function is given to the host matrix. The plot of Equation (4.12) for a small spherical pure metal in avtive host matrix is shown on the figure below.

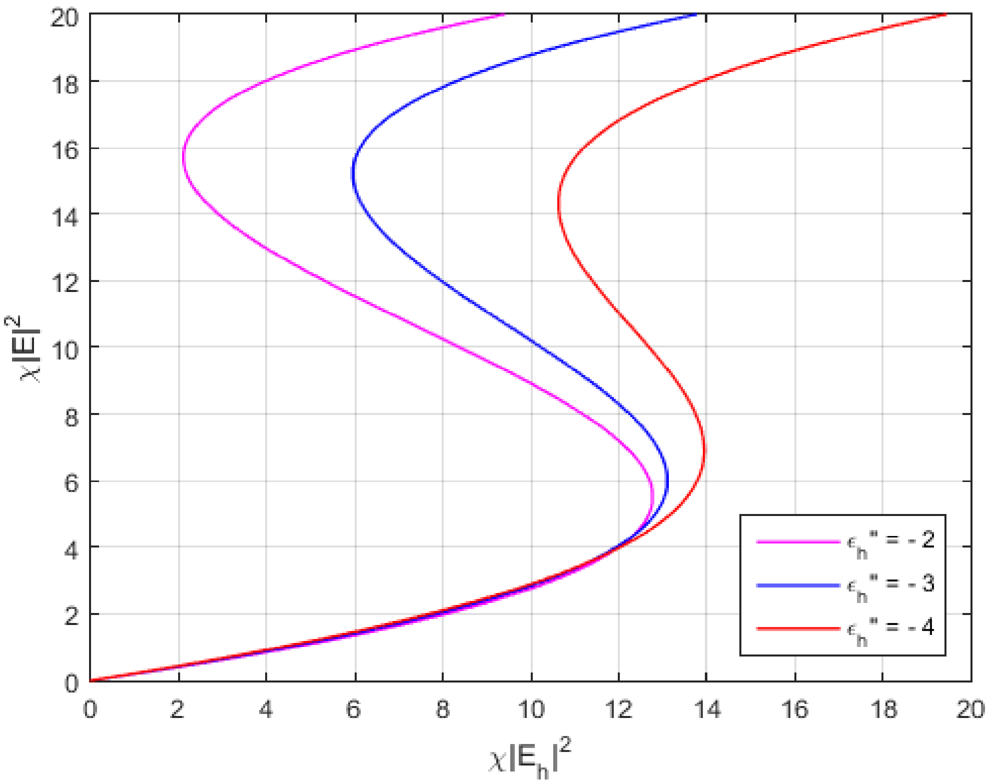

Figure 4.2: Local field $\chi|E|^2$ versus applied field $\chi|E_h|^2$ at frequency $z = 0.2$, with parameters , $\epsilon_h' = 2.25$, $\gamma = 0.0115$ for a small spherical pure silver particle in active host matrix ($\epsilon_h'' = -2$, $\epsilon_h'' = -3$, and $\epsilon_h'' = -4$).

From Fig. 4.2, optical bistability is illustrated in the Y-X plane and we observed S-like curves. Increasing of $|E_h|^2$ from zero to and gives increasing of $|E|^2$ monotonically. However, after a certain value of $|E_h|^2$, decreasing the value of $|E_h|^2$ gives increasing of the local field $|E|^2$. Using linear stability analysis this branch of solution is unstable. That means if the system is initially in this state, it will rapidly switch to one of the stable solutions through the growth of small perturbations. So, with increasing of $|E_h|^2$ from zero to and when it passes the turning point in the lower branch then immediately it switching up to the upper branch. On the other hand, if the input intensity is slowly decreased, the system will remain on the upper branch and the output intensity will continue and at the turning point switching down to the lower branch.

The switching up and switching down input intensity for different values of magnitude of ϵ_h'' in active host matrix are listed out in Table 4.2.

ϵ_h''	Switching up $\chi\|E_h\|^2$ (onset)	Switching down $\chi\|E_h\|^2$ (offset)	range width of $\chi\|E_h\|^2$ ($\Delta\chi\|E_h\|^2$) (threshold width)
− 2	12.77	2.103	10.667
− 3	13.1	5.959	7.141
− 4	13.95	10.63	3.32

Table 4.2: $\chi|E_h|^2$ at different values of ϵ_h'' for a small spherical pure metal (silver) in active host matrix (Switching up and switching down of OB).

From the Table, we observed that both the swithing up and the switching down input intensities increase. But the input intensity gap between switching up and switching down input intensities decreases as the magnitude of ϵ_h'' increases, and this results a narrow width range of optical bistability. i.e., when $\epsilon_h'' = -2$, we obtained the width range of $\Delta\chi|E_h|^2 = 10.667$, when $\epsilon_h'' = -3$, we obtained the width range of $\Delta\chi|E_h|^2 = 7.141$, and when $\epsilon_h'' = -4$, we obtained the width range of $\Delta\chi|E_h|^2 = 3.32$. This shows that as the imaginary part of the dielectric function of the host matrix increases, the width range of optical bistability or the threshold width narrowns. This narrow threshold width enables the system to oscillate highly, or increases the activation of the system.

Chapter 5

Refractive Index of Small spherical Pure Metal in Passive and Active Host Matrix

It is shown that we calculated the refractive index of a small spherical pure metal for both real and imaginary parts in passive and active host matrix. It is important to derive for the dimensionless polarisability (α) and the dielectric function (ϵ) to calculate the refractive index.

5.1 Polarizability

The polarizability of an atom or molecule is the physical magnitude that describes the response of the electron cloud to an external field M. Domingo et. al [37]. Polarizability is an important fundamental property related with refraction. In the electrostatic approximation when the wavelength of the incident electromagnetic radiation is much greater than the typical size of the particle, referring Eqn. (3.6) of section 3.1, the unknown coefficient B is

$$B = \frac{\varepsilon_m - \varepsilon_h}{2\varepsilon_h + \varepsilon_m} \cdot {r_1}^3 \tag{5.1}$$

The dimensionless polarisability (α) can be expressed interms of B and r_1^2. Letting $B = \alpha r_1^3$, then, the dimensionless polarizability (α) is

$$\alpha = \frac{\varepsilon_m - \varepsilon_h}{\varepsilon_m + 2\varepsilon_h} \tag{5.2}$$

The complex dimensionless polarizability (α) and the complex Dielectric function (ϵ) can be expressed in their real and imaginary parts respectively as

$$\alpha = \alpha' + i\alpha'' \quad and \;\; \varepsilon = \varepsilon' + i\varepsilon'' \tag{5.3}$$

we plug equation (5.3) in to equation (5.2), we have

$$\alpha = \frac{(\varepsilon'_m - \varepsilon'_h) + i(\varepsilon''_m - \varepsilon''_h)}{(\varepsilon'_m + 2\varepsilon'_h) + i(\varepsilon''_m + 2\varepsilon''_h)}. \tag{5.4}$$

The real and imaginary parts of the polarizability of the small spherical pure metal, are respectively:

$$\alpha'_m = \frac{(\varepsilon'_m - \varepsilon'_h)(\varepsilon'_m + 2\varepsilon'_h) + (\varepsilon''_m - \varepsilon''_h)(\varepsilon''_m + 2\varepsilon''_h)}{(\varepsilon'_m + 2\varepsilon'_h)^2 + (\varepsilon''_m + 2\varepsilon''_h)^2} \tag{5.5}$$

and

$$\alpha''_m = \frac{(\varepsilon'_m + 2\varepsilon'_h)(\varepsilon''_m - \varepsilon''_h) - (\varepsilon'_m - \varepsilon'_h)(\varepsilon''_m + 2\varepsilon''_h)}{(\varepsilon'_m + 2\varepsilon'_h)^2 + (\varepsilon''_m + 2\varepsilon''_h)^2} \tag{5.6}$$

5.2 Refractive Index of Small Spherical pure Metal in Passive and Active Host Matrix

The refractive index is one of the most important parameters for an optical medium. Defined as $n = c/v$, it measures the comparative velocity of light in different media. When a light beam travels across the boundary between two different materials, it bends owing to the change in refractive index at the interface.

This phenomenon gives the reason why a water pool appears shallower than it actually is, why a straw placed partially in water at a slant seems to bend towards the surface, and why people can use eyeglasses to adjust the path of light in front of their eyes and alleviate the effects of conditions such as myopia. The refractive index is a complex number, $n = n' + in''$, where the imaginary part (n'') characterizes the losses in the material. Using the electromagnetic description of light given by Maxwell's equations, the refractive index (n) is related to basic material parameters, namely the permittivity or the dielectric function(ϵ) and the permeability μ [28].

The famous Clausius-Mossoti formula is used to write an equation for the complex dielectric function of the small spherical pure metal embedded in active and passive host matrix.

$$\frac{\varepsilon_m - \varepsilon_h}{\varepsilon_m + 2\varepsilon_h} = \frac{4\pi}{3} BN, \tag{5.7}$$

where B is a constant that is given by equation (5.1), and N is a density number of atoms in small spherical pure metal. Let us introduce a new function f which is called the volume fill fraction of spherical metallic particle in a host matrix, it can be expressed interms of N and r^3 as:

$$f = \frac{4\pi}{3} r_1^3 N \ , \tag{5.8}$$

and N is expressed as

$$N = \frac{3}{4\pi r_1^3} f, \tag{5.9}$$

finally, summarising equations, (5.1), (5.7) and (5.8) we get,

$$\frac{\epsilon_m - \epsilon_h}{\epsilon_m + 2\epsilon_h} = f\alpha \tag{5.10}$$

$$\Rightarrow \epsilon - \epsilon_h = (\epsilon + 2\epsilon_h) f\alpha$$
$$\epsilon(1 - f\alpha) = \epsilon_h(1 + 2f\alpha)$$

$$\epsilon = \epsilon_h(1 + 3f\frac{\alpha}{1 - f\alpha}) \tag{5.11}$$

where ϵ is the complex dielectric function and it can be expressed interms of the real and imaginary parts of the polarisability α as follows

$$\epsilon = \epsilon_h(1 + 3f\frac{\alpha' + i\alpha''}{1 - f(\alpha' + i\alpha'')}), \tag{5.12}$$

where, $\epsilon_h = \epsilon'_h + i\epsilon''_h$ and $\epsilon = \epsilon' + i\epsilon''$, therefore, the real and iaginary parts of the dielectric function expressed on equation (5.12) respectively, are

$$\epsilon' = \epsilon'_h + 3f\frac{(\alpha' - f|\alpha|^2)\epsilon'_h - \alpha''\epsilon''_h}{(1 - f\alpha')^2 + (f\alpha'')^2}, \tag{5.13}$$

$$\epsilon'' = \epsilon''_h + 3f\frac{(\alpha' - f|\alpha|^2)\epsilon''_h + \alpha''\epsilon'_h}{(1 - f\alpha')^2 + (f\alpha'')^2}. \tag{5.14}$$

Recalling equation (2.26), the relation between the complex refractive index (n) and the complex dielectric function (ε), and squaring both sides, we have

$$n^2 = \epsilon \tag{5.15}$$

expressing n and ϵ in terms of the real and imaginary parts, we have
$n = n' + in''$ and $\epsilon = \epsilon' + i\epsilon''$, then

$$n'^2 - n''^2 + 2in'n'' = \epsilon' + i\epsilon'' \tag{5.16}$$

By substituting the real parts of the polarizability in to the real parts of the dielectric function, and the imaginary parts of the polarizability in to the imaginary parts of the dielectric function and then by equating the real and imaginary parts of the refractive index with the real and imaginary parts of the dielectric functions, we obtain the following equations for the real and imaginary relations respectively [28].

$$n'^2 - n''^2 = \epsilon' , \qquad 2n'n'' = \epsilon'' \tag{5.17}$$

The above equation yields the expression for the real n' and imaginary n'' parts of the refractive index of small spherical pure metal embedded in a linear dielectric host matrix, respectively as [28]:

$$n' = \sqrt{\frac{1}{2}(\sqrt{\epsilon'^2 + \epsilon''^2} + \epsilon')} \tag{5.18}$$

$$n'' = \sqrt{\frac{1}{2}(\sqrt{\epsilon'^2 + \epsilon''^2} - \epsilon')} \tag{5.19}$$

5.2.1 The real part of refractive index of a Small Spherical Pure Metal in a Passive Host Matrix

Referring equation (5.18), the real part of refractive index is expressed in terms of the real and imaginary parts of the dielectric functions of the metal and that of the host matrix. In the passive material, the natural property of the dielectric function of the host matrix is not affected because, there is no additional dielectric function applied to the host matrix, then the imaginary part of the dielectric function of the host matrix is zero ($\epsilon''_h = 0$). The graph of the real part of refractive index expressed on Eqn. (5.18) has one maximum value and one minimum value. The graph of the real part of refractive index versus the dimensionless frequency z in a passive host matrix is plotted below.

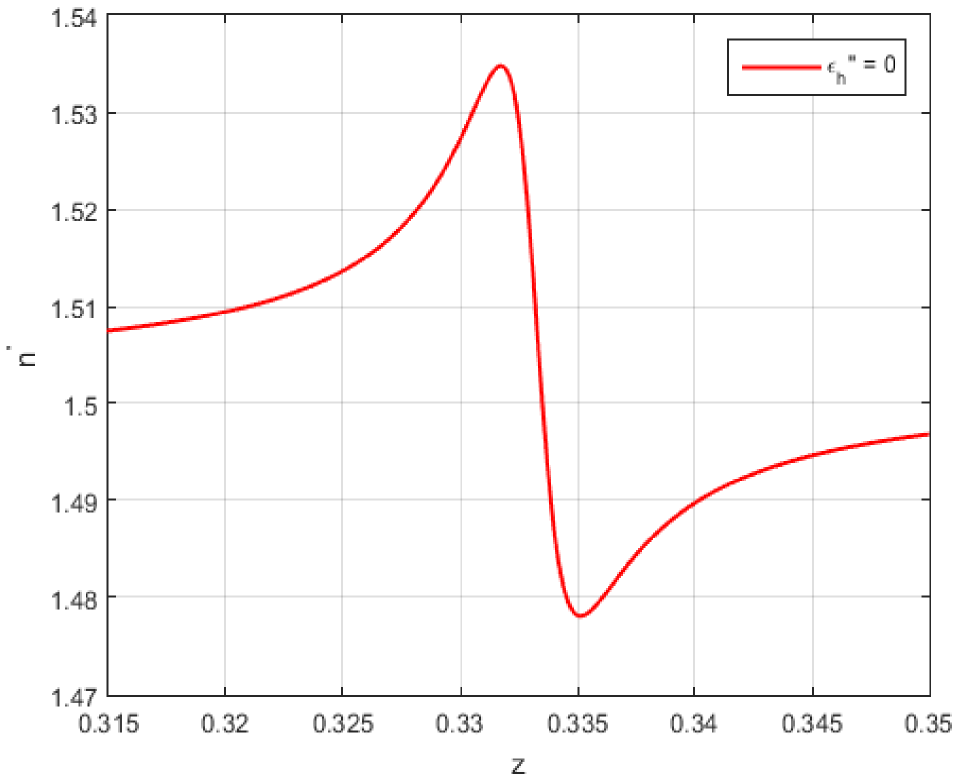

Figure 5.1: The real part of refractive index n' versus z for small spherical pure silver metal in passive host matrix ($\epsilon_h'' = 0$) and $f = 0.001$ where other parameters specified as in fig. 4.1.

Fig. 5.1 shows the graph of n' versus the dimensionless frequency z in a passive host matrix ($\epsilon_h'' = 0$) for a small spherical pure metal. From the graph, we observed that there are one maximum and one minimum values of n', the refractive index rises to a maximum value,then drops to the minimum, then rises again. The drop in refractive index happens over wider size gap of frequency range and this agrees with the definition of anomalous dispersion. The real part of the refractive index n' increases with increasing the dimensionless frequency z on the resonance region this phenomenon agrees with the definition of normal dispersion. The relation between the magnitude of ϵ_h'' and the frequency range for the real part of refractive index in a passive host matrix is illustrated on the table 5.1.

ϵ_h''	$Refracted\ \ width\ (\Delta z)$
0	0.0110

Table 5.1: The dependence between ϵ_h'' and frequncy range for the real part of refractive index in passive host matrix.

From Table 5.1, in passive host matrix ($\epsilon_h'' = 0$), we obtained wider refracted width (Δz) of magnitude 0.0110.

5.2.2 The real part of refractive index of a Small Spherical Pure Metal in Active Host Matrix

Reffering equation (5.18), the real part of refractive index is expressed in terms of the real and imaginary parts of the Dielectric function of the metal and that of the host matrix. In the active material, the natural property of the DF of the host matrix is affected by applying additional dielectric function on the host matrix. Therefore, the imaginary part of the dielectric function of the host matrix is different from zero ($\epsilon_h'' \neq 0$). Then, the real part of the refractive index is also affected. The graph of the real refractive index in active host matrix expressed on Eqn. (5.18) is plotted below.

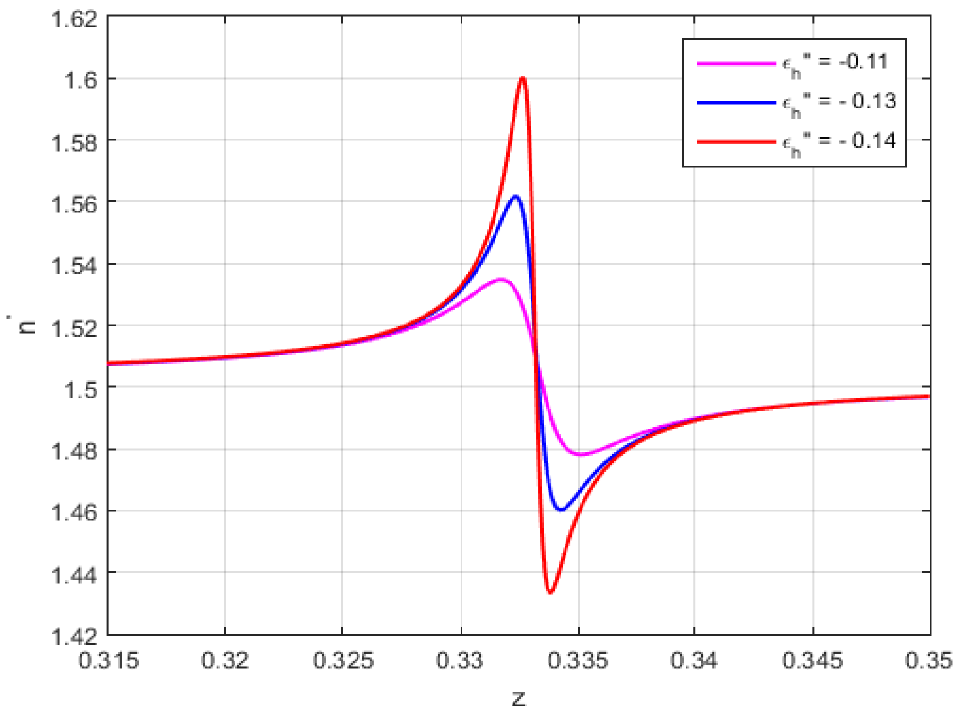

Figure 5.2: Real part of refractive index n' versus z for a small spherical pure silver metal in active host matrix ($\epsilon_h'' = -0.11$, $\epsilon_h'' = -0.13$ and $\epsilon_h'' = -0.14$) respectively, where other parameters specified as in fig.3.1. and $f = 0.001$.

Figure 5.2 shows the graph of n' versus the dimensionless frequency z in active host Matrix ($\epsilon_h'' < 0$) for a small spherical pure metal. There are one maximum and one minimum values of n' for each value of the imaginary part of the dielectric function of the host matrix (ϵ_h'').

From the graph, we observed that, as the dimensionless frequency z increases, the refractive index rises to a maximum value,then drops to the minimum, then rises again. The drop in refractive index happens over different size gaps of frequency ranges, and this agrees with the definition of anomalous dispersion. The real part of the refractive index (n') increases with increasing the dimensionless frequency (z) on the resonance region this phenomenon agrees with the definition of normal dispersion.

The relation between the magnitude of ϵ_h'' and the frequency range for the real part of refractive index in active host matrix is shown in the table below.

ϵ_h''	$Refracted\ \ width\ (\Delta z)$
-0.11	0.0034
-0.13	0.0020
-0.14	0.0011

Table 5.2: The dependence between ϵ_h'' and frequncy range for the real part of refractive index in active host matrix.

From Table 5.2, we observed that, as the imaginary part of the DF of the host matrix increases, this resembles as that of the optical density of the small spherical pure metal increases, then the incident light refracts or bends most, therfore, the refracted width (Δz) decreases.

When $\epsilon_h'' = -0.11$, we obtained the refracted width (Δz) of 0.0034, when $\epsilon_h'' = -0.13$, we obtained the refracted width (Δz) of 0.0020 and when $\epsilon_h'' = -0.14$, we obtained the refracted width (Δz) of 0.0011. Therefore, from the results we understand that, the most the external DF applied to the host matrix, the least the frequency range or refracted width occurs. This narrow range of refracted width results high resonance of light on the system.

5.2.3 The Imaginary part of refractive index of a Small Spherical Pure Metal in Passive Host Matrix

Reffering equation (5.19), for the Imaginary part of refractive index is expressed in terms of the real and imaginary parts of the Dielectric function of the metal and that of the host matrix. In the passive material, the imaginary part of the dielectric function of the host matrix is zero ($\epsilon_h'' = 0$). The graph of the imaginary part of refractive index expressed on Eqn. (5.19) has only one maximum value ($n'' = 0.03262$) at a resonance frequency ($z = 0.3333$).

The graph of the imaginary part of refractive index (n") versus the dimensionless frequency (z) in passive host matrix is plotted below.

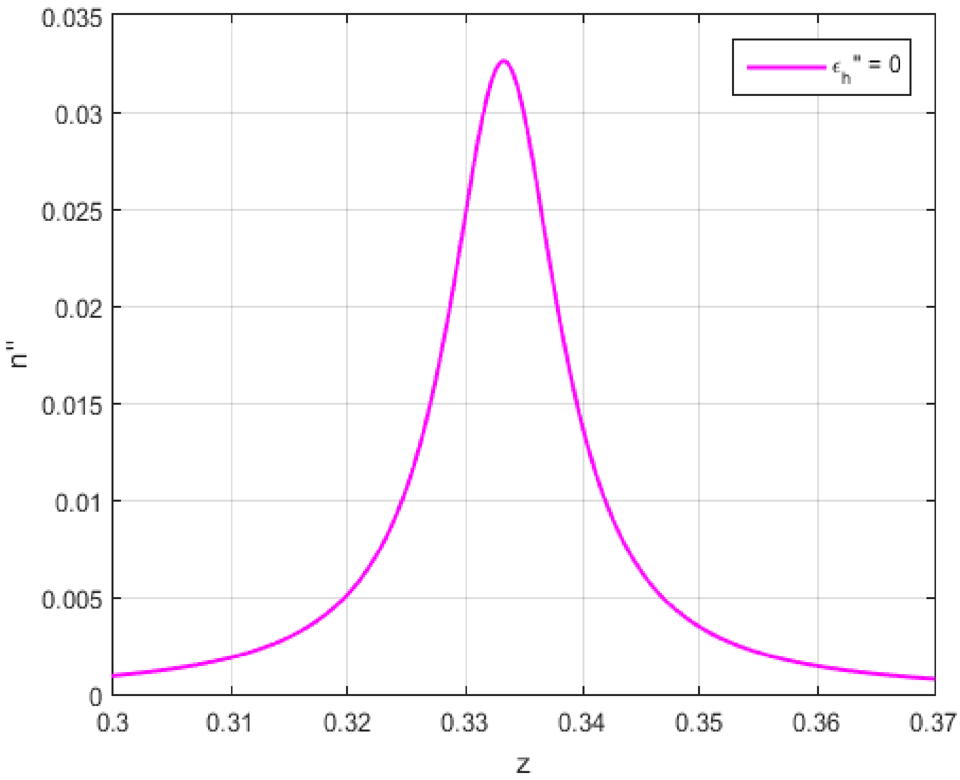

Figure 5.3: Imaginary part of refractive index n'' versus z for small spherical pure metal in passive host matrix, with parameters $\epsilon_h' = 2.25$, f $= 0.001$, $\gamma = 0.0115$, $\epsilon_h'' = 0$.

5.2.4 The Imaginary part of refractive index of a Small Spherical Pure Metal in Active Host Matrix

Reffering equation (5.19), the Imaginary part of refractive index is expressed in terms of the real and imaginary parts of the Dielectric function of the metal and that of the host matrix. In the active material, the imaginary part of the dielectric function of the host matrix has contribution in the enhancement of the imaginary part of refractive index (n''), and it can be varying by applying additional dielectric function on the host matrix, this is like as that of increasing the optical density of the small spherical pure metal, thus, the imaginary part of the refractive index (n'') is enhanced, or the amplitude increased.
The graph of the imaginary part of refractive index expressed on Eqn. (5.19) is plotted below.

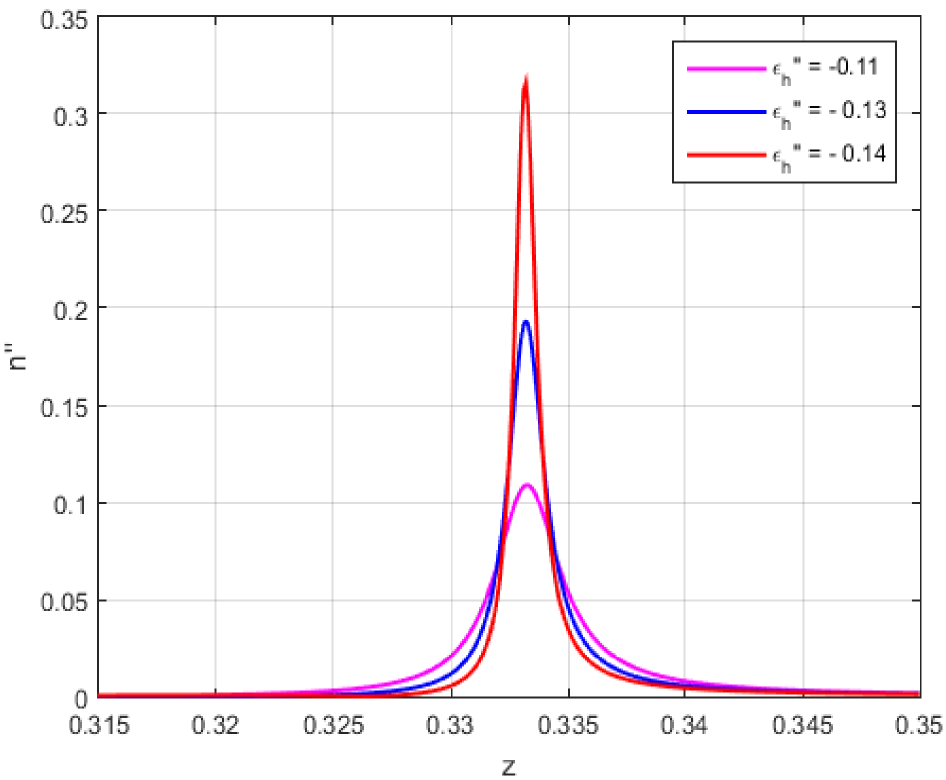

Figure 5.4: Imaginary part of refractive index (n'') versus z for small spherical pure metal in active host matrix, with parameters $\epsilon'_h = 2.25$, $f = 0.001$, $\gamma = 0.0115$, $\epsilon''_h = -0.11$, $\epsilon''_h = -0.13$, $\epsilon''_h = -0.14$ plotted on one plane.

Figure 5.4 shows the imaginary part of the refractive index (n'') of small spherical pure metal versus the dimensionless frequency (z) in active host matrix, with numerical values of parameters ($\epsilon'_h = 2.25,\ \epsilon_\infty = 4.5, \omega_p = 1.6 \times 10^{16},\ \gamma = 0.0115,\ f = 0.0001$).
The dependency between the maximum value of the Imaginary part of refractive index (n'') and the imaginary part of the dielectric function (ϵ''_h) in active host matrix is shown on the table given below.

ϵ''_h	$Mxmvalueofn''$	z
− 0.11	0.1088	0.3332
− 0.13	0.1934	0.3332
− 0.14	0.3159	0.3332

Table 5.3: The dependency between the maximum value of the imaginary refractive index and ϵ''_h in active host matrix.

From figure 5.4 and Table 5.3, we observed that, as the imaginary part of the dielectric function of the host matrix increases, this resembles as that of increasing the optical property of the small spherical pure metal, then the maximum value of Imaginary part of refractive index also increases.
Hence, the large value of n'' agrees with the definition of strong absorption of incident light.

Chapter 6

Conclusions

In this thesis, we studied the optical properties of a small spherical pure metal in passive and active host matrix.

We have calculated the local field enhancement factor ($|A|^2$) for a small spherical pure metal embedded in a linear host matrix. We plotted the graph of enhancement factor ($|A|^2$) versus dimensionless frequency (z) for a small spherical pure silver metal in a passive active host matrix and observed that, there is only one maximum values of the local field enhancement factor ($|A|^2$) for the passive and active matrices. When the natural property of the dielectric function of the host matrix is varying by applying additional dielectric function to it, like as that of increasing the optical density of the small spherical pure metal, then the local field enhancement factor increased.

We calculated optical bistability within a certain input intensity and we found interesting findings on threshold range differences for the optical bistability in the passive and active matrices, the threshold range is affected as the natural property of the dielectric function of the host matrix is varying. In our mathematical calculation, to observe considerable differences on the onset and offset parts of the plots of optical bistability for the active host matrix, we took the integer value of the imaginary part of dielectric function of the host matrix ϵ_h'' (i.e., $\epsilon_h'' = -2$, $\epsilon_h'' = -3$ and $\epsilon_h'' = -4$). We have observed that both the swithing up and the switching down input intensities increased. But the input intensity gap between switching up and switching down input intensities (the threshold range) decreases as the imaginary part of the dielectric function of the host matrix ϵ_h'' increased by applying additional dielectric function on it, that is like as the optical density of the small spherical metal increased. This narrower threshold range enables the system to oscillate highly, increases the activation of the system, or makes the switcing up and switching down process to be achieved with in short range of time easily.

We have calculated the real and imaginary parts of the refractive index of a small spherical pure metal embedded in passive host matrix and active host matrix. We observed that the real part of refractive index (n') has one maximum value and one minimum value in the passive and active host matrix, and as the value of the imaginary part of the dielectric function of the host matrix (ϵ_h'') increases like as that of the optical density of the small spherical pure metal increases, thus, the incident light refracts (bends) most, due to this effect, the refracted width (Δz) decreases. This narrow range of refracted width results high resonance offree electrons on the small spherical pure metal.

The imaginary part of the refractive index has only one maximum value both in passive and active host matrices. And we observed that, n'' or the amplitude increases as the imaginary part of the dielectric function of the host matrix (ϵ_h'') increases this is like the optical density of the small spherical pure metal increases.

Generally, the analytical and numerical results show that varying the natural property of the dielectric function of the host matrix or applying additional dielectric function on the host matrix, increases the optical density of the small spherical pure metal and, therefore greately affects the local field enhancement factor ($|A|^2$), the optical bistability and the real and imaginary parts of refractive index (n).

Bibliography

[1] C. F. Bohren and D. R. Huffman, "Absorption and scattering of," *Light by Small*, 1983.

[2] U. Kreibig and M. Vollmer, "Theoretical considerations," in *Optical properties of metal clusters*, pp. 13–201, Springer, 1995.

[3] B. R. Masters and P. So, *Handbook of biomedical nonlinear optical microscopy*. Oxford University Press, 2008.

[4] V. G. Dmitriev, G. G. Gurzadyan, and D. N. Nikogosyan, *Handbook of nonlinear optical crystals*, vol. 64. Springer, 2013.

[5] R. W. Boyd and S. J. Bentley, "Recent progress in quantum and nonlinear optical lithography," *Journal of modern optics*, vol. 53, no. 5-6, pp. 713–718, 2006.

[6] H. Nihei and A. Okamoto, "Photonic crystal systems for high-speed optical memory device on an atomic scale," in *Optical Engineering for Sensing and Nanotechnology (ICOSN 2001)*, vol. 4416, pp. 470–473, International Society for Optics and Photonics, 2001.

[7] T. Tokizaki, A. Nakamura, S. Kaneko, K. Uchida, S. Omi, H. Tanji, and Y. Asahara, "Subpicosecond time response of third-order optical nonlinearity of small copper particles in glass," *Applied physics letters*, vol. 65, no. 8, pp. 941–943, 1994.

[8] V. Shalaev, "Nonlinear optics of random media, vol. 158 of," *Springer Tracts in Modern Physics*, 2000.

[9] R. S. Bennink, Y.-K. Yoon, R. W. Boyd, and J. Sipe, "Accessing the optical nonlinearity of metals with metal–dielectric photonic bandgap structures," *Optics Letters*, vol. 24, no. 20, pp. 1416–1418, 1999.

[10] L. A. Lyon, M. D. Musick, and M. J. Natan, "Colloidal au-enhanced surface plasmon resonance immunosensing," *Analytical chemistry*, vol. 70, no. 24, pp. 5177–5183, 1998.

[11] P. Fortina, L. J. Kricka, D. J. Graves, J. Park, T. Hyslop, F. Tam, N. Halas, S. Surrey, and S. A. Waldman, "Applications of nanoparticles to diagnostics and therapeutics in colorectal cancer," *Trends in biotechnology*, vol. 25, no. 4, pp. 145–152, 2007.

[12] D. Ricard, P. Roussignol, and C. Flytzanis, "Surface-mediated enhancement of optical phase conjugation in metal colloids," *Optics letters*, vol. 10, no. 10, pp. 511–513, 1985.

[13] J. Feldmann, "Plasmon resonances in large noble-metal clusters," *New Journal of Physics*, vol. 4, pp. 93–1, 2002.

[14] M. Guerrisi, R. Rosei, and P. Winsemius, "Splitting of the interband absorption edge in au," *Physical Review B*, vol. 12, no. 2, p. 557, 1975.

[15] P. B. Johnson and R.-W. Christy, "Optical constants of the noble metals," *Physical review B*, vol. 6, no. 12, p. 4370, 1972.

[16] V. Mal'nev and S. Shewamare, "Slow and fast light in metal/dielectric composites with passive and active host matrices," *Physica B: Condensed Matter*, vol. 426, pp. 52–57, 2013.

[17] A. Lagendijk, B. Nienhuis, B. A. van Tiggelen, and P. de Vries, "Microscopic approach to the lorentz cavity in dielectrics," *Physical review letters*, vol. 79, no. 4, p. 657, 1997.

[18] J. Guo, J. Cooper, and A. Gallagher, "Selective reflection from a dense atomic vapor," *Physical Review A*, vol. 53, no. 2, p. 1130, 1996.

[19] J. J. Maki, M. S. Malcuit, J. Sipe, and R. W. Boyd, "Linear and nonlinear optical measurements of the lorentz local field," *Physical review letters*, vol. 67, no. 8, p. 972, 1991.

[20] R. W. Boyd, "Nonlinear optics, academic press," *San Diego*, vol. 155, 1992.

[21] O. Knudsen, "Ow richardson and the electron theory of matter, 1901–1916," *Histories of the Electron: The Birth of Microphysics*, pp. 227–254, 2001.

[22] C.-K. Duan, M. F. Reid, and Z. Wang, "Local field effects on the radiative lifetime of emitters in surrounding media: virtual-or real-cavity model?," *Physics Letters A*, vol. 343, no. 6, pp. 474–480, 2005.

[23] H. Husu, "Electromagnetic resonances and local fields in the linear and nonlinear optical response of metal nanostructures," 2011.

[24] Q. M. Ngo, K. Q. Le, D. L. Vu, *et al.*, "Optical bistability based on fano resonances in single-and double-layer nonlinear slab waveguide gratings," *JOSA B*, vol. 31, no. 5, pp. 1054–1061, 2014.

[25] O. Levy and D. J. Bergman, "Intrinsic optical bistability and resonances in nonlinear composites," *Physica A: Statistical Mechanics and its Applications*, vol. 207, no. 1-3, pp. 157–162, 1994.

[26] H. Gibbs, "Optical bistability: Controlling light with light," *Orlando*, 1985.

[27] M. Soljacic, M. Ibanescu, C. Luo, S. G. Johnson, S. Fan, Y. Fink, and J. D. Joannopoulos, "All-optical switching using optical bistability in nonlinear photonic crystals," in *Photonic crystal materials and devices*, vol. 5000, pp. 200–214, International Society for Optics and Photonics, 2003.

[28] W. Cai and V. M. Shalaev, *Optical metamaterials*, vol. 10. Springer, 2010.

[29] C. Li, "Refractive index engineering and optical properties enhancement by polymer nanocomposites," 2016.

[30] H. Shim, F. Monticone, and O. D. Miller, "Fundamental limits to the refractive index of transparent optical materials," *arXiv preprint arXiv:2103.15697*, 2021.

[31] A. N. Oraevsky and I. E. Protsenko, "Optical properties of heterogeneous media," *Quantum Electronics*, vol. 31, no. 3, p. 252, 2001.

[32] S. G. Moiseev, E. A. Pashinina, and S. V. Sukhov, "On the problems of transparency of metal–dielectric composite media with dissipative and amplifying components," *Quantum Electronics*, vol. 37, no. 5, p. 446, 2007.

[33] F. Wooten, *Optical properties of solids*. Academic press, 2013.

[34] T. Pan, J. Huang, and Z. Li, "Optical bistability in metal/dielectric composite with interfacial layer," *Physica B: Condensed Matter*, vol. 301, no. 3-4, pp. 190–195, 2001.

[35] L. Gao, "Optical bistability in composite media with nonlinear coated inclusions," *Physics Letters A*, vol. 318, no. 1-2, pp. 119–125, 2003.

[36] T. Tamasgen, S. Shewamare, and G. Melese, "Induced optical bistability in a composite of ellipsoidal metal/dielectric nanoparticles of dielectric core," 2016.

[37] K. J. Miller, "Calculation of the molecular polarizability tensor," *Journal of the American Chemical Society*, vol. 112, no. 23, pp. 8543–8551, 1990.