TWO PLASMA RESONANCES IN METAL COVERED NANOPARTICLES WITH SMALL DIELECTRIC CORE

BY

Seyoum Eyasu Gebray

A THESIS SUBMITTED TO
THE DEPARTMENT OF PHYSICS

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

ADDIS ABABA UNIVERSITY
ADDIS ABABA,ETHIOPIA
JUNE 2012/13

ADDIS ABABA UNIVERSITY
SCHOOL OF GRADUATE STUDIES

This is to certify that the thesis prepared by **BYSeyoum Eyasu Gebray**, entitled "**Two Plasma Resonances in Metal Covered Nanoparticles with Small Dielectric Core**" and submitted in partial fulfillment of the requirements for the degree of **Master of Science**, compiles with the regulations of the University and meets the accepted standards with respect to originality.

Signed by the Examining Committee:

Examiner:________________________ Signature:________ Date:________________
Examiner:________________________ Signature:________ Date:________________
Advisor: ________________________ Signature:________ Date:________________

Abstract

It was generally accepted that metal covered inclusions in a dielectric host matrix have one maxima of the enhancment factor at one resonant frequency close to the frequency of the surface plasmon. Recently,Sisay shewamare, V.N.Mal'nev with coauthors have shown that the local field in the metal covered nanoinclusion with small dielectric core has two maxima at two different resonant frequencies. The second maxima is not important in metal covered inclusions with small dielectric cores. But for large metal fractions, the maxima of the enhancment factor become the same order of magnitude and must be theken into account.

The aim of this thesis is to check the dependence of the resonant frequencies and the enhancment factor of the local field on the parameters of the composite(metal fraction,dielectric function of the core, metal shell, and host matrix) in the spherical inclusions. And the second objective of the thesis is to calculate the above mention physical quantites in the composites with cylinderical inclusions. The account of two maxima of the enhancment factor of the local field affects the linear and non linear optical properties of the composites metal inclusions having small dielectric cores.

Acknowledgements

I would like to thank Prof. V. N. Mal'nev, my supervisor, for his many suggestions and constant support during this research. I am also grateful to him for his guidance through the early years of chaos and confusion.

Addis Ababa University, Ethiopia
Seyoum Eyasu
June, 2013

Table of Contents

Introduction

In recent years, the plasmonic resonances in metallic systems have [1 - 7] attracted great interest due to their ability to increase between the light and matterials interaction. It has been shown that the incident light interacting with nanometal can dramatically enhance the local electrical field by concentrating electromagnetic radiation in the composietes of metal covered nanoparticles with dielectric cores imbedded in a dielectric matrix. The amplitude of the electric field of the incident intensive radiation is enhaced in the inclusion on the frequency close to the surface frequency of the metal [7 - 9]. We analyze numerically and analytically of the enhancement factors of the spherical and cylinderical inclusions in a dielectric host matrix as the function of metal fraction and frequency range of the wave radiation in the model when a decay of the plasma vibration is negligible or for the practically non-decaying the plasma vibrations in the metal part of the inclusion corresponding to the case of very small $\gamma <<< 1$. And we have shown that the enhancement factor of the local field spherical metal inclusions with dielectric core in a dieletric host matrix has two maxima on two different frequencies that depend on the parameters of the systems [10-13] . With increasing the metal fraction of the inclusions, both maxima grow up and the second maximum must be taken in to account along with the first one while considering the electrodynamic propertiies [14-17] of the composiete . In this thesis, we study the enhancement factors of the local field in spherical and cyliderical inclusions with dielectric cores embedded in a dielectric matrix [18-20] . It will be shown that the local field in these inclusions with the small dielectric core considerably increase the local field comparing to the pure metal inclusions.

The previous work done by Sisay shewamare, and Prof. V.N.Mal'nev is the resonant frequencies and the enhancement factor of the local field on the parameters(metal fraction,dielectric function of the core, metal shell, and the host matrix). And numerically and analytically of the two maxima of the enhancement factors inmetal covered spherical inclusions. But the object of this thesis is to check the dependence of the resonant frequencies and the enhancment factor of the local field on the parameters of the composite(metal fraction,dielectric function of the core, metal shell, and host matrix) in the spherical inclusions. And (1) to calculate the two resonances frequencies and the two enhancement factors of the local field in metal covered cylinderical inclusions, (2) to derive the Mathematical expression of the enhancement factor of the local field and the two resonant frequencies in cyliderical inclusions, (3), to analyze the analytically and numerically of the enhancement factor of the local field and (4) to obtain the two resnant frequencies in cyliderical inclusions.

Chapter 1

Plasma frequency and plasma vibrations

1.1 Logitudinal plasma vibrations

Consider a one-dimensional situation in which a metal consisting entirely of one charge species is displaced from its quasi-neutral position by an infinitesimal distance δx.

An equal and opposite charge density develops on the metal site face. It is known that due to the quasineutrality of metals, the positive change of ions in the crystalline lattice is completely compensated by the negative charge of the electron. Small local violation of the quasineutrality creats an electric filed E, which act on the electron.

The equation of motion of a free electron in an electric filed is:

$$m\frac{d^2(\delta x)}{dt^2} = e\delta E_x \tag{1.1.1}$$

where m is the mass of the electron and $\delta x(t) = (\delta x)_o e^{-i\omega_p t}$ is its displacement.

The electric filed can be obtained from the equation :

$$\nabla . E = 4\pi\rho, \tag{1.1.2}$$

where ρ is the density of the charge, which in turn is :

$$\rho = e(n_i - n_e). \tag{1.1.3}$$

Here n_i, n_e are the density numbers of the ions and electrons, respctively.

The ions subsystem is the slow one and can be treated at rest, therefore, n_i = constant.

The density numbers of the elctrons can be presented as:

$$n_e = n_e^o + \delta n_e, \tag{1.1.4}$$

where due to the quasineutrality $n_e^o = n_i^o = n_0$ and $\delta n_e(x)$ is a departure from the quasineutrality. Quasi-neutrality demands that:

$$n_i \simeq n_e \equiv n_s, \tag{1.1.5}$$

where n is the number density (i.e., the number of particles per cubic meter) of species . For the small diplacment of the electron, $\delta n_e^o = -n_o \delta x$. We obtain Eq.(1.1.1) by account of Eq.(1.1.3) and Eq.(1.1.4) as follows:

$$m\frac{d^2(\delta x)}{dt^2} = -4\pi e^2 n_o \delta x. \tag{1.1.6}$$

By ccount of Eq.(1.1.6), we obtain the equation of longitudinal motion of a simple harimonic oscillator as follows:

$$\frac{d^2(\delta x)}{dt^2} + \omega_p^2 \delta x = 0. \tag{1.1.7}$$

It describes the longitudinal plasma vibration of the electrons with plasma frequency ω_p.

1.2 Plasma frequency

If the electrons in a plasma are displaced from a uniform background of ions, electric field pointing in such a direction as to restore the neutrality of the plasma by pulling the electrons back to their original positions.

Because of their inertia, the electrons will overshoot and oscillate around their equilibrium positions with a characteristic frequency known as the plasma frequency.

The equation of the plasma frequency of the free electron gas can be written as:

$$\omega_p = \sqrt{\frac{4\pi e^2 n_o}{m_e}} \tag{1.2.1}$$

This formula relaties to the plasma frequency in all systems containing electrons and ions. It is known that the density number of the electrons in metals is approximentaly

$n_o = 10^{22} cm^{-3}$. taking $e = 4.8 \times 10^{-10}$ c and $m = 10^{-27} g$, we get that $\omega_p = 5 \times 10^{15}$ cycles/s and this relaties to the optical frequency range. By using the Drude mode[7] of permittivity for metals, as the function of frequency in the following form:

$$\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma)}. \tag{1.2.2}$$

Here ω_p is the plasmon frequency of the corresponding bulk metal, and γ is the damping constant due to the electron collisions in that metal. Here, ε_∞ is ε $(\omega \to \infty)$ and when one neglect the collision (lossless), one has $\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2}$. Therefore, the permittivity is negative when the frequency is lower than ω_p. When such a metal meets another material with a positive permittivity, a wave can be bounded by their interface according to the Maxwell equations. Longitudinal density fluctuations (plasma oscillations) at plasma frequency ω_p and quanta of volume plasmons have energy:

$$\hbar\omega_p = \hbar\sqrt{\frac{4\pi n e^2}{m}}, \tag{1.2.3}$$

the Eq.(1.2.3) describes the plasmon energy of the corresponding bulk metal.

1.3 Surface Plasmons (SP)

Consider the SP at Metal-Dielectric Interface with the plasma dielectric ε_p and the dielectric permittivity ε_d. Let us consider solution of the Laplace equations for the single interface:[8]

$$\nabla^2\Phi(x, y, z) = 0, \tag{1.3.1}$$

were Φ is the electric potential.

The potential distribution on the two media and the solution, that wave like in the x-direction and exponentially decaying in the z- direction from the interface is given by:

$$\Phi(x,z) = \Phi_{op}\cos kxe^{-k_1 z}, z > 0, \tag{1.3.2}$$

$$\Phi(x,z) = \Phi_{od}\cos kxe^{k_2 z}, z < 0. \tag{1.3.3}$$

The potential distribution in Eq.(1.3.2) and Eq.(1.3.3), satisfies the Laplace equation in Eq.(1.3.1).

Now we have to satisfies the boundary conditions and the continutity equation at the interface respectivily shows as follows:

$$\Phi_p(x,z)_{z=0} = \Phi_d(x,z)_{z=0}, \tag{1.3.4}$$

$$\varepsilon_p \frac{\partial \Phi}{\partial z} \mid_{z=0} = \varepsilon_d \frac{\partial \Phi}{\partial z} \mid_{z=0} . \tag{1.3.5}$$

The potential and the normal components of the dielectric displacement and require that $\Phi_{op} = \Phi_{od}$ [8]. The second condition of continuity Eq.(1.3.5) of the displacement vector:

$$\varepsilon_p(-k)\Phi_{0p} = \varepsilon_d k \Phi_{0p}. \tag{1.3.6}$$

And additionally From Eq.(1.3.5) with ccount of Eq.(1.3.6), we obtain:

$$\varepsilon_p + \varepsilon_d = 0. \tag{1.3.7}$$

By substituting the plasma dielectric ε_p in to the Eq.(1.3.7) we get:

$$(\varepsilon_\infty - \frac{\omega_p^2}{\omega^2}) + \varepsilon_d = 0. \tag{1.3.8}$$

We obtain the equation for the frequency of the surface waves from Eq.(1.3.8) by neglect small damping constant as follows:

$$\omega_{sp}^2 = \frac{\omega_p^2}{\sqrt{\varepsilon_\infty + \varepsilon_d}}. \tag{1.3.9}$$

In plasma-vacuum , $\varepsilon_\infty = 1$ and $\varepsilon_d = 1$,then the Eq.(1.3.9) [9] gives:

$$\omega_{sp} = \frac{\omega_p}{\sqrt{2}}. \tag{1.3.10}$$

Eq.(1.3.10) discribes the frequency of the surface plasmons.

Chapter 2

Local-field and potential distribution in metal covered spherical inclusion

2.1 The model composites of nanoshells

The geometry of dielectric core-silver shell structure nanometer size sphere is shown in fig.(2.1). The dielectric core has radius $R_1 = r_1$ and dielectric fuction ε_1, the metal shell

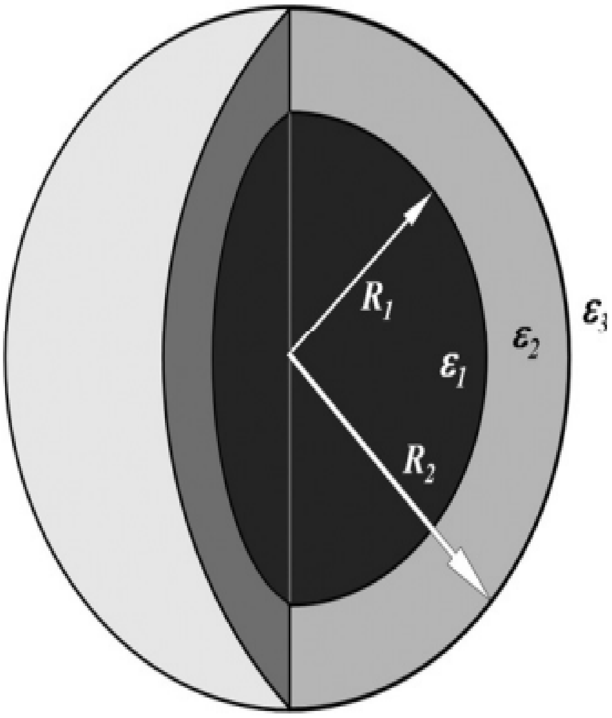

Figure 2.1: Dielectric core-metal shell structure nanoparticle geometry. Source: Applied surface (science) Phys. 253(2007)8729-8733

has radius $R_2 = r_2$, and dielectric fuction ε_2, and the host matrix has dielectric function $\varepsilon_3 = \varepsilon_h$. The dielectric fuction of the metal shell, ε_2 have real (ε_2') and imaginary (ε_2'') parts dependent on the frequency. In our calculations, the typical size of a particle d is much smaller than the wavelength of incident radiation λ, ($d << \lambda$). In this case, the particle is subjected to an almost uniform field.

2.2 The potential distribution in a spherical metal nanoparticle with dielectric core in a host matrix

According to the quasi-static theory and model composites of nanoshells, the potential distribution in each shell of the nanostructure could be derived from Laplaces equation for the potential, $\nabla \Phi_i^2 = 0$, from which we will able to calculate the electric field $E = -\nabla \Phi_i$. Due to the azimuthal symmetry of the problem, the general solution for the potential distribution in each shell ($i = 1, 2, 3$) is of the form:[8]

$$\Phi_i(r, \theta) = [A_i r + (B_i/r^2)] \cos\theta. \tag{2.2.1}$$

Let us consider the composite media, where the coated inclusions are imbedded in the linear host medium and ε_h does not depend on the electric field.

Under the quasi-static approximation, we can readily obtain the solution for the potentials inside the core, shell and host medium in an external constant electric filed E_h can be written as follows:

$$\Phi_1 = -E_h A r \cos\theta, r < r_1, \tag{2.2.2}$$

$$\Phi_2 = -E_h(Br - C/r^2) \cos\theta, r_1 \leq r \leq r_2, \tag{2.2.3}$$

$$\Phi_h = -E_h(r - D/r^2) \cos\theta, r > r_2. \tag{2.2.4}$$

Here Φ_1, Φ_2, and Φ_h are potentials in the dielectric core, metal, and the host matrix, respectively, E_h is the aplied field, r and θ are the spherical coordinates of the observation point (the $z-$ axis is chosen along the vector E_h), r_1 and r_2 are radiuses of dielectric core and the metal shell of the inclusion, respectively, and A,B,C,D, are unknown coefficients. It is easy to show that Φ_i satisfy the Laplaces equation for the potential $\nabla^2\Phi_i = 0$.

2.3 Derivation the linear algebraic equations of the unknown coefficients

To obtain a system of Linear algebraic equation for the unknown coefficients and the solution of the system, we used the continuity condition and the displacement vector on the interfaces of the dielectric core - metal and meta - host matrix.

The potential equations on the interfacs of dielectric core - metal and metal - host matrix can be expressied as follows:

$$\Phi_1 \mid_{r=r_1} = \Phi_2 \mid_{r=r_1}, \tag{2.3.1}$$

$$\Phi_2 \mid_{r=r_2} = \Phi_h \mid_{r=r_2} . \tag{2.3.2}$$

2.4 The continuity condition for the potential on the interfaces of the spherical nanoinclusions

The continuity conditions of the electric potential and the displacement vector on the interfaces of dielectric core - metal and metal - host matrix can be importantly to solving the system of linear algebraic equations for the unknown coefficients.

The continuity equations for the potential distribution on the interfaces are thus:

$$\varepsilon_1 \frac{\partial \Phi_1}{\partial r} \mid_{r=r_1} = \varepsilon_2 \frac{\partial \Phi_2}{\partial r} \mid_{r=r_1}, \tag{2.4.1}$$

$$\varepsilon_2 \frac{\partial \Phi_2}{\partial r} \mid_{r=r_2} = \varepsilon_h \frac{\partial \Phi_h}{\partial r} \mid_{r=r_2} . \tag{2.4.2}$$

By using the Eq.(2.3.1),(2.3.2) and the Eq.(2.4.1), (2.4.2),the solution of four unknown parameters A, B, C and D can be obtained as follows:[1]

$$A = \frac{9\varepsilon_h\varepsilon_2}{2p\varepsilon_2^2 + \varepsilon_2(3-2p)\varepsilon_1 + 2\varepsilon_2(3-p)\varepsilon_h + 2p\varepsilon_h\varepsilon_1}, \tag{2.4.3}$$

$$A = \frac{9\varepsilon_h \varepsilon_2}{2p\Delta}, \tag{2.4.4}$$

$$B = \frac{3\varepsilon_h(\varepsilon_1 + 2\varepsilon_2)}{2p\varepsilon_2^2 + \varepsilon_2(3-2p)\varepsilon_1 + 2\varepsilon_2(3-p)\varepsilon_h + 2p\varepsilon_1\varepsilon_h}, \tag{2.4.5}$$

$$B = \frac{3\varepsilon_h(\varepsilon_1 + 2\varepsilon_2)}{2p\Delta}, \tag{2.4.6}$$

$$C = \frac{3\varepsilon_h(\varepsilon_1 - \varepsilon_2)}{2p\varepsilon_2^2 + \varepsilon_2(3-2p)\varepsilon_1 + 2\varepsilon_2(3-p)\varepsilon_h + 2p\varepsilon_h\varepsilon_1} r_1^3, \tag{2.4.7}$$

$$C = \frac{3\varepsilon_h(\varepsilon_1 - \varepsilon_2)}{2p\Delta} r_1^3, \tag{2.4.8}$$

$$D = [1 - \frac{3\varepsilon_h(3-p)\varepsilon_2 + p\varepsilon_1}{2p\varepsilon_2^2 + \varepsilon_2(3-2p)\varepsilon_1 + 2\varepsilon_2(3-p)\varepsilon_h + 2p\varepsilon_1\varepsilon_h}] r_2^3, \tag{2.4.9}$$

$$D = [1 - \frac{3\varepsilon_h(3-p)\varepsilon_2 + p\varepsilon_1}{2p\Delta}] r_2^3, \tag{2.4.10}$$

$$\Delta = \varepsilon_2^2 + q\varepsilon_2 + \varepsilon_1\varepsilon_h.$$

Here, $q = (\frac{3}{2p} - 1)\varepsilon_1 + (\frac{3}{p} - 1)\varepsilon_h$, $p = 1 - (\frac{r_1}{r_2})^3$ is a metal fraction in the inclusion ε_1, ε_2, and ε_h are the dielectric function of the core, metal shell, and the host matrix respectively. The above formulas describe the electric field of the electromagnetic wave in the electrostatic approximation when the wavelength of the incident radiationis much larger than typical size of the inclusion ($d << \lambda$). We note that this formulas can be obtained from the corresponding results for the elliptic inclusions [11].

2.5 Local-field distribution in composite media

In the following analysis, the nanoshells is modeled as shown in Figure 2.1, with dielectrc core radius r_1, metal shell radius r_2 and dielectric constants of the core ε_1 ($\varepsilon_1 = 6$), dielectric host matrix (surrounding medium $\varepsilon_h = 2.25$), and ε_2 dielectric for the silver shell. The quasi-static approximation is a first-order approach, which describes the interaction between a silver nanoshells and electromagnetic radiation at optical frequencies .

In the quasi-static approximation, the incident electric field, is assumed not to vary spatially over the dimensions of the nanoshells.

It is valid in the limit where the wavelength of incident light λ is much greater than a typical size of the inclusion d of the nanoshell ($d << \lambda$).

As we have seen, it dramatically simplifies the mathematical analysis describing the nanoshells interaction with the electric field and, more importantly, it provides avaluable physical interpretation of local field in silver nanoshells.

According to the quasi-static theory and model composites of nanoshells, the local field distribution in each shell of the nanostructure could be derived from Laplaces equation. The solution for the electric field in the nanoshells corresponding to the core ε_1, shell ε_2 and the host medium ε_h is determined by solving Laplaces equation for the potential in each shells, then taking the gradient to determine the electric field in each of the three shells and which are determined by satisfying boundary conditions for continuity of the tangential component of the electric field and continuity of the normal component of the displacement field. Therefore, the electric fields ($E_i = -\nabla\Phi_i$) [10] are calculated to be:

$$E_1 = \frac{9\varepsilon_2\varepsilon_h}{\varepsilon_2\varepsilon_a + 2\varepsilon_h\varepsilon_b} E_h(\cos\theta r - \sin\theta\hat{\theta}), \qquad (2.5.1)$$

$$E_2 = \tfrac{3\varepsilon_h}{\varepsilon_2\varepsilon_a+2\varepsilon_h\varepsilon_b}[(\varepsilon_1 + 2\varepsilon_2) + 2(\varepsilon_1 - \varepsilon_2)(\tfrac{r_1}{r})^3]E_h \sin\theta\hat{r}$$

$$-[(\varepsilon_1 + 2\varepsilon_2) - (\varepsilon_1 - \varepsilon_2)(\frac{r_1}{r})^3]E_h \sin\theta\hat{\theta}, \qquad (2.5.2)$$

$$E_3 = (2\frac{\varepsilon_2\varepsilon_a - \varepsilon_h\varepsilon_b}{\varepsilon_2\varepsilon_a + 2\varepsilon_h\varepsilon_b}\frac{r_2^3}{r^3} + 1)E_h \cos\theta\hat{r} + (2\frac{\varepsilon_2\varepsilon_a - \varepsilon_h\varepsilon_b}{\varepsilon_2\varepsilon_a + 2\varepsilon_h\varepsilon_b}\frac{r_2^3}{r^3} - 1)E_h \sin\theta\hat{\theta}, \qquad (2.5.3)$$

$$\varepsilon_a = \varepsilon_1(3 - 2p) + 2\varepsilon_2 p,$$

$$\varepsilon_b = \varepsilon_1 p + \varepsilon_2(3 - p),$$

$$p = 1 - (\tfrac{r_1}{r_2})^3,$$

where $\hat{r}$, $\hat{\theta}$ are unit vectors a long $\vec{r}$ and $\vec{\theta}$.

The surface plasmon resonance of metal nanoparticle propagation of an electromagnetic wave in a medium containing spherical metallic nanoparticles would cause displacement of conduction electrons relative to the positively charged ionic cores.

This in turn results in dipole oscillating with the same frequency as of the incident wave. If radiuses of the nanoparticles (the diametere of the inclusions 'd') are much smaller than the wavelength λ of the incident wave, $(d << \lambda)$, the electrostatic approximation is valid and the dipole moment of the nanoparticles can be given by $(p = \varepsilon_h \alpha E_i)$ located at the dielectric core of the shell and polarizability of the core can be obitained as follows:

$$\alpha = 4\pi\varepsilon_o r_2^3 \mid \frac{\varepsilon_2\varepsilon_a - \varepsilon_h\varepsilon_b}{\varepsilon_2\varepsilon_a + 2\varepsilon_h\varepsilon_b} \mid . \tag{2.5.4}$$

Here, α is the polarzability of the nanopartcles in the electrostatic approximation.

When, the temporal oscillation of the electric field is taken into account, we arrive at a classical physical interpretation of surface plasmon resonance in a nanoshell.

In the quasi-static limit, the dominant behavior of a silver nanoshell in an electric field at optical frequencies is that of an oscillating dipole. In this classical picture, resonance occurs when the polarizability α is very large, or when the denominator of Eq.(2.5.4) $\mid \varepsilon_2\varepsilon_a + 2\varepsilon_h\varepsilon_b \mid$ is minimum or goes to zero,the polarizability shows a resonant enhancement. Then, the condition for resonance is the follows:[7,17]

$$\varepsilon_2\varepsilon_a + 2\varepsilon_h\varepsilon_b = 0. \tag{2.5.5}$$

In this simplified analysis, the polarizability becomes very large when the resonance condition is satisfied because damping effects $\gamma << 1$ due to reaction radiation have been very small or neglected.

Chapter 3

Resonant frequencies and enhancement factor of local field in metal covered spherical inclusions

3.1 Enhancement factor of the local field in spherical and cylindrical nanoinclusions

The enhancement factors of the local field of the incident electromagnetic radiation in the composites of metal covered nanopartciels with dielectric core is a great of importance because of different posible applications of these systems.

Then, the local field factor at the dielectric core or in the inner surface of the shell can been written as the magnitude of the electric field ratio $\mid E_1/E_h \mid$ with $r = r_1$, and $\theta = 0$, and the local field factor at the metal shell or at the exterior surface of the shell can been written as the magnitude of the electric field ratio $\mid E_2/E_h \mid$ with $r = r_2$, and $\theta = 0$.

The Amplitude of the electric field of the incident intensive radiation is enhaced in the inclusion (Local field) on the frequency closed to the surface frequency of the metal, $(\omega \simeq \omega_p)$. Then, the local field E_1 in the dielectric core of the inclusion can be obtained with the help of relation $E_1 = FE_h$. The enhancement factor F coincides with the coefficient A given by Eq.(2.4.4), and then, the local field in the dielectric core of the inclusion can be obtained as follows:

$$E_1 = AE_h. \tag{3.1.1}$$

Then, by subistituting the coefficient that given in Eq.(2.4.4) in to Eq.(3.1.1), the local field E_1 of the dielectri core of the inclusion in a dielectric host matrix can be obtained as follows:

$$E_1 = \frac{9\varepsilon_h\varepsilon_2}{2p\Delta}E_h. \tag{3.1.2}$$

And the modulus of the enhancemen factor in the dielectric core can be written as belows:

$$\mid A_3 \mid = \mid \frac{E_1}{E_h} \mid . \tag{3.1.3}$$

The modulus squared of the enhancement factor of Eq.(3.1.3) in dielectric core can be expressied as follows:

$$\mid A_3 \mid^2 = \mid \frac{E_1}{E_h} \mid^2 . \tag{3.1.4}$$

By subistituting the local field of dielectric core E_1 given in Eq.(3.1.2) in to Eq.(3.1.4), we obitained the modulus squared of the enhancement factort of the dielectric core of inclusion in a host matrix as follows:

$$\mid A_3 \mid^2 = \frac{81\varepsilon_h^2\varepsilon_2^2}{4p^2\Delta^2}. \tag{3.1.5}$$

Here, the delta function in the dielectric core of the inclusion:

$$\Delta = \varepsilon_2^2 + q\varepsilon_2 + \varepsilon_1\varepsilon_h. \tag{3.1.6}$$

Let us choose the dielectric function of a metal in the Drude form:

$$\varepsilon_2 = \varepsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma)}, \tag{3.1.7}$$

were ω_p is the plasma surface frequaency of the metal shell of the inclusions.

Introducing the dimeensionless frequaencies, $z = \frac{\omega}{\omega_p}$, and $\gamma = \frac{\nu}{\omega_p}$, ω is the frequency of the incident radiation, and ν is the electron collision frequency, we get :

$$\varepsilon_2 = \varepsilon_\infty - \frac{1}{z(z + i\gamma)}. \tag{3.1.8}$$

And it's real $\varepsilon_2{}'$ and imaginary $\varepsilon_2{}''$ parts are given by the following equation:

$$\varepsilon_2{}' = \varepsilon_\infty - \frac{1}{z^2 + \gamma^2}, \varepsilon_2{}'' = \frac{\gamma}{z(z^2 + \gamma^2)}. \tag{3.1.9}$$

By subistituting the Eq.(3.1.6) and Eq.(3.1.9) in to Eq.(3.1.5), the enhancement factor of the local field of the dielectric core in the inclusion can be obtained as follows:

$$\mid A_3 \mid^2 = \frac{81\varepsilon_h^2}{4p^2} \frac{\varepsilon_2'^2 + \varepsilon_2''^2}{[\varepsilon_2'^2 - \varepsilon_2''^2 + q\varepsilon_2'^2 + \varepsilon_1\varepsilon_h]^2 + \varepsilon_2''^2[q + 2\varepsilon_2']^2}. \tag{3.1.10}$$

Here ε_2' and ε_2'' are the real and the imaginary parts of ε_2 given in Eq.(3.1.9), respectively. For the sake of simplicity, we ignore the imaginary parts of ε_1 and ε_h.

The dielectric function DF of the inclusion core ε_1 in general case includes linear and nonlinear[12] part with respect to the local field:

$$\varepsilon_1 = \varepsilon_{1o} + \chi \mid E_1 \mid^2 . \tag{3.1.11}$$

Here ε_{1o} is the linear part of DF, which does not depends on E_1. And the parameter χ is the nonlinear Kerr coefficient, E_1 is the local field in the inclusion core. Then, for the week incident radiation of the local field of the inclusion core, we assumes that the above relation is weakly nonlinear, that is, the contribution of the second nonlinear part $\chi \mid E_1 \mid^2$ in the right hand of Eq.(3.1.11) is much less than that of the first linear part ε_{1o},($\chi \mid E_1 \mid^2 << \varepsilon_{1o}$).

Further, for the sake of simplicity, we ignore the imaginary parts of dielectric core ε_1 and we study the effctive linear response of strongly linear composite media when the nonlinear part in Eq.(3.1.11) vanishes.

3.2 Analysis the two resonant frequencies and the two maxima of the dielectric core of inclusion at different value of metal fraction

The analytic analysis is carried out for the adially non-decaying the plasma vibrations in the metal part of the inclusion corresponding to the case of very small $\gamma <<< 1$.

In this case, the second terem in the denominator of Eq.(3.1.10) proportional to $\varepsilon_2''^2 \sim \gamma^2$ is very small or neglected the imaginary part of ε_2''.

The local field enhancement factor fo the dielectric core given in Eq.(3.1.10) is maximized, when the first term in the demominator of Eq.(3.1.10) is a minimum or goes to zero, the enhancement factor shows a resonant enhancement.

In this simplified analysis, the enhancement factor goes to infinity when the resonance condition is satisfied because damping effects due to reaction radiation have been neglected and this condition gives the quadratic equation with respect to the real part ε_2' is form:

$$\varepsilon_2'^2 + q\varepsilon_2{}' + \varepsilon_1\varepsilon_h = 0. \tag{3.2.1}$$

We consider the two limits of the metal fractions of the inclusion, in the first limit of the metal fracton is very small or a thin cover ($p << 1$) and a large dielectric core, with the parameter, $q = \frac{3[\varepsilon_1/2+\varepsilon_h]}{p} >> 1$.

By using the quadratic equation, we obtain the two solutions of Eq.(3.2.1) as follows:

$$\varepsilon_{21}' = -q, \tag{3.2.2}$$

$$\varepsilon_{22}' = -\varepsilon_1\varepsilon_h/q. \tag{3.2.3}$$

By subistituting the two roots of the Eq.(3.2.2) and Eq.(3.2.3) in to the Eq.(3.1.10), we obitained the two solutions of the resonant frequencies at the maximum enhancement factor as follows:

$$z_1' = \sqrt{1/(\varepsilon_\infty + q)} \approx \sqrt{1/q}, \tag{3.2.4}$$

$$z_2' = \sqrt{1/(\varepsilon_\infty + \varepsilon_1\varepsilon_h/q)} \approx \sqrt{1/\varepsilon_\infty}, \tag{3.2.5}$$

$$q >> \varepsilon_\infty, \varepsilon_1\varepsilon_h.$$

We using the $\epsilon_2'' = \gamma/z^3$, and by subistituting the two solutions of the resonant frequaency that obitained in Eq.(3.2.4) and Eq.(3.2.5) in to the Eq.(3.1.10),then the two maxima of the enhancement factors of the dielectric core of inclusion in a dielectric host matrix as the function of the frequency can be obtained as bellows:

$$\mid A(z_1') \mid^2 = \frac{81\varepsilon_h^2}{4(3[\varepsilon_1/2+\varepsilon_h])^3}\frac{p}{\gamma^2}, \tag{3.2.6}$$

$$\mid A(z_2') \mid^2 = p\frac{\varepsilon_1^2\varepsilon_h^2}{3[\varepsilon_1/2+\varepsilon_h]\varepsilon_\infty^3} \mid A(z_1) \mid^2, \tag{3.2.7}$$

$$\gamma << p.$$

It is clear that the first maxima of the enhancement factor $\mid A(z_1') \mid^2$ is much greater than that of the second one because $p << 1$. We consider the second limit, in which the metal fraction is large ($p \simeq 1$) and small dielectric cores, with the parameter, q $= \varepsilon_1/2 + 2\varepsilon_h$. In this limit, by using the quadratic equation, the two solutions of Eq.(3.2.1) can be obtained as follows;

$$\varepsilon_{21}' = -2\varepsilon_h, \tag{3.2.8}$$

$$\varepsilon_{22}' = -\varepsilon_h/2. \tag{3.2.9}$$

By subistituting these two roots in Eq.(3.2.8) and Eq.(3.2.9) in to the Eq (3.1.9),then, we obtained the following two resonant frequencies as follows:

$$z_1 = \sqrt{1/[\varepsilon_\infty + 2\varepsilon_h]}, \tag{3.2.10}$$

$$z_2 = \sqrt{1/[\varepsilon_\infty + \varepsilon_1/2}. \tag{3.2.11}$$

By using the approximation $\varepsilon_2'' = \gamma/z^3$, and by substituting the Eq.(3.2.10) and the Eq.(3.2.11) into the Eq.(3.1.10),then, two maxima of the enhancement factors as the function of the resonant frequaencies are obtained as follows:

$$\mid A(z_1) \mid^2 = \frac{81\varepsilon_h^4}{[\varepsilon_1 - 4\varepsilon_h]^2(\varepsilon_\infty + 2\varepsilon_h)^3}\frac{1}{\gamma^2}, \tag{3.2.12}$$

$$\mid A(z_2) \mid^2 = \frac{\varepsilon_1^2}{\varepsilon_h^2}\frac{(\varepsilon_\infty + 2\varepsilon_h)^3}{(\varepsilon_\infty + \varepsilon_1/2)^3}\frac{\mid A(z_1) \mid^2}{16}, \tag{3.2.13}$$

$$\varepsilon_1 \neq 4\varepsilon_h.$$

The two maxima of the enhancement factors of the Eq.(3.2.12) and Eq.(3.2.13) with the small dielectric core of the inclusion in a host matrix ($p \simeq 1$), are the same order and the large enough $\sim \frac{1}{\gamma^2}$, when we compare with the large dielectric core of the inclusion of Eq.(3.2.6) with Eq.(3.2.7), shows that, the maxima of the enhancement factor inclusion with the small cores are much largre than those of the inclusion with large cores.

The further increment of the enhancement factor happens to be posible by considering the tuned inclusion and tuning dielectric functions of the core and host matrix.

The two resonant frequencies that obitaied in Eq.(3.2.10) and Eq.(3.2.11) are coincide with the accuracy $\varepsilon_2'' << 1$ provided that:

$$\varepsilon_1 = 4\varepsilon_h. \tag{3.2.14}$$

In this case, the resonant frequencies coincide,$z_1 = z_2 = z_r = \frac{1}{\sqrt{\varepsilon_\infty + 2\varepsilon_h}}$ and the two enhancement factors merge and their maximum becomes extremely large and proportional to $\sim 1/\gamma^4$. It can be written as follws:

$$\mid A(z_r) \mid^2 = \frac{81\varepsilon_h^4}{[\varepsilon_\infty + 2\varepsilon_h]^6} \frac{1}{\gamma^4}. \tag{3.2.15}$$

The enhancement factor $\mid A_m \mid^2$ of the metal shell or at the the exterior surface of the shell can be obtained as the magnitude of the electric field ratio $|E_2/E_h|$ with $r = r_2$, $\theta = 0$, and by seting the metal fraction, $p = 1$ and making substitution $\varepsilon_1 \longrightarrow \varepsilon_2$ in to the Eq.(3.1.10), the enhancement factor of the pure silver metal can be obtained as follows:

$$\mid A_m \mid^2 = \frac{9\varepsilon_h^2}{(\varepsilon_2' + 2\varepsilon_h)^2 + \varepsilon_2''^2}. \tag{3.2.16}$$

3.3 The potential distribution in cylinderical inclusion with dielectric core in a dielectric host matrix

According to the quasi-static theory and model composites of nanoshells, the potential distribution in each shells of the nanostructure could be derived from Laplaces equation for the potential $\nabla^2\Phi_i = 0$, therefore, the electric field $E = -\nabla\Phi_i$.

The general equation for the potential distribution in each shells (i = 1, 2, 3) is the follows:

$$\Phi_i = [A_i r + B_i/r] \cos\theta, \tag{3.3.1}$$

i = 1, 2, 3 relate to the core, metal shell, and host matrix. Let us consider the composite where the coated inclusions are embedded in the linear host medium. Under the quasi-static approximation, we can readily obtain the linear electric potentials inside the core, shell and host medium [8]can be shows as follows:

$$\Phi_1 = -E_h A r \cos\theta, r < r_1, \tag{3.3.2}$$

$$\Phi_2 = -Eh(Br - C/r)\cos\theta, r_1 \leqslant r \leqslant r_2, \tag{3.3.3}$$

$$\Phi_h = -Eh(r - D/r)\cos\theta, r > r_2. \tag{3.3.4}$$

Here Φ_1 , Φ_2, and Φ_h are potentials in the dielectric core, metal, and the host medum respectively, E_h is the aplied field r and θ are the cylinderical coordinates of the observation point, (the z- axis is chosen along the cylinder axis), r_1 and r_2 are radiuses of the dielectric core and the metal shell of the inclusion respectively, and A,B,C,D, are the unknown coefficients.

From the continuity conditions of the potential given in Eq.(2.3.1) and Eq.(2.3.2), and the displacement vector on the boundaries of dielectric core-metal shell and metal- host matrix given in Eq.(2.4.1) and Eq.(2.4.2), we obitained a system of linear algebraic equations for A, B, C, D.

Then,the solution of the unknown coefficients of the system can be obitained as follows:

$$A = \frac{4\varepsilon_h\varepsilon_2}{p\Delta}, \tag{3.3.5}$$

$$B = \frac{2\varepsilon_h(\varepsilon_1 + \varepsilon_2)}{p\Delta}, \tag{3.3.6}$$

$$C = \frac{2\varepsilon_h(\varepsilon_1 - \varepsilon_2)}{p\Delta} r_1^2, \tag{3.3.7}$$

$$D = [1 - \frac{2\varepsilon_h[(2-p)\varepsilon_2 + p\varepsilon_1]}{p\Delta}] r_2^2, \tag{3.3.8}$$

$$\Delta = \varepsilon_2^2 + q\varepsilon_2 + \varepsilon_1\varepsilon_h,$$

$$q = (2/p - 1)\varepsilon_1 + (2/p - 1)\varepsilon_h,$$

were $p = 1 - (r_1/r_2)^2$ is the metal fraction for cylinderical inclusion.

The above formulas describe the electric field of the electromagnetic wave in the electrostatic approximation when the wavelength λ of the incident radiation is much larger than typical size d of the inclusion ($d << \lambda$).

3.4 Analysis the enhancement factor of cylinderical inclusions in a dielectric host matrix

We consider analytically and graphically enhancement factor of spherical and cylinderical inclusions in a dielectric host matrix as the function of metal fraction and frequency of the wave in a model when a decay of the plasma vibrations extremely small $\gamma \sim \varepsilon_2'' << 1$. And also the numerical results of the enhancement factor of the local field of spherical and cylinderical inclusions in the host matrix with ccount of the finite damping of plasma vibrations, ($\gamma << 1$).

The Amplitude of the electric field of the incident intensive radiation is enhaced in the inclusion on the frequency closed to the surface frequency of the metal, ($\omega \simeq \omega_p$).

The local field E_1 in the dielectric core of the inclusion can be obtained with the help of relation as follows:

$$E_1 = F E_h. \tag{3.4.1}$$

The enhancement factor F coincide with the coefficient A and subistituting the coefficient that given in Eq.(3.3.5) in to the Eq.(3.4.1), then the local field of the dielectri core of the inclusion obtained as follows:

$$E_1 = \frac{4\varepsilon_h \varepsilon_2}{p\Delta} E_h. \tag{3.4.2}$$

The modulus of the enhancemen factor in the dielectric core can be obitaied as belows:

$$\mid A_2 \mid = \mid E_1/E_h \mid . \tag{3.4.3}$$

And the modulus squared of the enhancement factor of Eq.(3.4.3) in dielectric core is expressied as follows:

$$\mid A_2 \mid^2 = \mid E_1/E_h \mid^2 . \tag{3.4.4}$$

By subistituting the local field of dielectric core E_1 given in Eq.(3.4.2) in to the Eq.(3.4.4), we can be obitained as follows:

$$\mid A_2 \mid^2 = \frac{16\varepsilon_h^2\varepsilon_2^2}{p^2\Delta^2}. \tag{3.4.5}$$

And by subistituting the Eq.(3.1.6) and Eq.(3.1.8) in to the Eq.(3.4.5), the enhancement factor of the local field for silver cylinderical coated nanoparticles of the dielectric core in the inclusions can be obtained as follows:

$$\mid A_2 \mid^2 = \frac{16\varepsilon_h^2}{p^2}\frac{(\varepsilon_2'^2 + \varepsilon_2''^2)}{(\varepsilon_2'^2 - \varepsilon_2''^2 + q\varepsilon_2' + \varepsilon_1\varepsilon_h)^2 + \varepsilon_2''^2(q + 2\varepsilon_2')^2}. \tag{3.4.6}$$

Here, ε_2' and ε_2'' are the real and the imaginary parts of ε_2 given in Eq.(3.1.7), respectively. For the sake of simplicity, we ignore the imaginary parts of ε_1 and ε_h.

The enhancement factor of the local field $\mid A_m \mid^2$ for a pure silver cylinderical nanopacticles can be obitained by seting the metal fraction (p = 1) and making subistitution $\varepsilon_1 \to \varepsilon_2$ into Eq.(3.4.5), then the enhancement factor for a pure cylinderical nanopacticles is the follows:

$$\mid A_m \mid^2 = \frac{2\varepsilon_h^2}{(\varepsilon_2' + \varepsilon_h)^2 + \varepsilon_2''^2}. \tag{3.4.7}$$

It coincides with the known result[13].

3.4.1 The resonant frequaencies and the local field enhancement factor in metal covered cylinderical inclusion

The analysis is carried out for the idialy non-decaying the plasma vibrations in the metal part of the inclusion corresponding to the case of very small$\gamma << 1$. In this case, the second terem in the denominator of Eq.(3.4.6) proportional to $\varepsilon_2''^2 \sim \gamma^2$ is very small. The local field enhancement factor of the dielectric core given in Eq.(3.4.6) is maximized, when the first term in the demominator of Eq.(3.4.6) is a minimum or goes to zero, the enhancement factor shows a resonant enhancement.

In this simplified analysis, the enhancement factor goes to infinity when the resonance condition is satisfied because damping effects due to reaction radiation have been very small or neglected and this condition gives the quadratic equation with respect to ε_2':

$$\varepsilon_2'^2 + q\varepsilon_2' + \varepsilon_1\varepsilon_h = 0. \tag{3.4.8}$$

We consider the two limits of the metal fractions of the inclusion, in the first limit the metal fracton is very small or a thin cover ($p << 1$) and a large dielectric core, the parameter q can be obitaied as, $q = 2[\varepsilon_1 + \varepsilon_h]/p >> 1$.

By using the quadratic equation, the two solutions of Eq.(3.5.1) is the form:

$$\varepsilon_{21}' = -q, \tag{3.4.9}$$

$$\varepsilon_{22}' = -\varepsilon_1\varepsilon_h/q. \tag{3.4.10}$$

By subistituting the two roots of the quadratic equation in Eq.(3.5.2) and Eq.(3.5.3) in to the Eq.(3.1.9) of the real part of dielectric fuction of the metal shell, we obitained the two resonant frequencies at the maximum enhancement factor.

Then, the solutions of the two resonant frequencies can be written as follows:

$$z_1' = \sqrt{1/(\varepsilon_\infty + q)} \approx \sqrt{1/q}, \tag{3.4.11}$$

$$z_2' = \sqrt{1/(\varepsilon_\infty + \varepsilon_1\varepsilon_h/q)} \approx \sqrt{1/\varepsilon_\infty}, \tag{3.4.12}$$

$$q >> \varepsilon_\infty, \varepsilon_1\varepsilon_h.$$

By using the $\epsilon_2'' = \gamma/z^3$, and by making subistituting of the two resonant frequaency solutions that obitained in Eq.(3.5.4) and Eq.(3.5.5) in the Eq.(3.4.6), the two maxima of the enhancement factor of the dielectric core as the function of the frequencies can be obtained as bellows:

$$\mid A(z_1') \mid^2 = \frac{16\varepsilon_h^2}{(2[\varepsilon_1 + \varepsilon_h])^3}\frac{p}{\gamma^2}, \tag{3.4.13}$$

$$\mid A(z_2') \mid^2 = p\frac{\varepsilon_1^2\varepsilon_h^2}{2[\varepsilon_1 + \varepsilon_h]\varepsilon_\infty^3} \mid A(z_1') \mid^2, \tag{3.4.14}$$

$$\gamma << p.$$

It is clear that, the first maxima of the enhancement factor $|A(z_1')|^2$ is much greater than that of the second one because $p << 1$. We consider the second limit, in which the metal fraction is large ($p \simeq 1$) and small dielectric cores, the parameter, $q = \varepsilon_1 + \varepsilon_h$.

By using the quadratic equation, the two solutions of Eq.(3.5.1) can be obtained as follows:

$$\varepsilon'_{21} = -\varepsilon_1, \tag{3.4.15}$$

$$\varepsilon'_{22} = -\varepsilon_h. \tag{3.4.16}$$

By subistituting these two roots of Eq.(3.5.8) and Eq.(3.5.9) in to the Eq.(3.1.9),then, we obtained the following two resonant frequencies as follows:

$$z_1 = \sqrt{1/(\varepsilon_\infty + \varepsilon_h)}, \tag{3.4.17}$$

$$z_2 = \sqrt{1/(\varepsilon_\infty + \varepsilon_1)}. \tag{3.4.18}$$

By using $\varepsilon''_2 = \gamma/z^3$, and by substituting Eq.(3.5.10) and Eq.(3.5.11) into Eq.(3.4.6), the two maxima of the enhancement factors as the function of two resonant frequaencies are obtained as follows:

$$\mid A(z_1) \mid^2 = \frac{16\varepsilon_h^4}{(\varepsilon_1 - \varepsilon_h)^2(\varepsilon_\infty + \varepsilon_h)^3}\frac{1}{\gamma^2}, \tag{3.4.19}$$

$$\mid A(z_2) \mid^2 = \frac{\varepsilon_1^2(\varepsilon_\infty + \varepsilon_h)^3}{\varepsilon_h^2(\varepsilon_\infty + \varepsilon_1)^3} \mid A(z_1) \mid^2, \tag{3.4.20}$$

$$\varepsilon_1 \neq \varepsilon_h.$$

The two maxima of the enhancement factors with small dielectric core of the inclusion in a host matrix with ($p \simeq 1$) are the same order and the large enough $\sim 1/\gamma^2$, when we compare with the large dielectric core ($p << 1$) of the inclusion, shows that the two maxima of the enhancement factors of the inclusions in the host matrix with the small dielectric cores are much largre than those of the inclusions with large dielectric cores.

The further increment of the enhancement factor happens to be posible by considering the tuned inclusion and tuning the dielectric functions of the core and host matrix from cylinderical inclusion. The two resonant frequencies that obitaied in Eq.(3.2.10) and Eq.(3.2.11) are coincide with the accuracy $\varepsilon''_2 << 1$ provided that:

$$\varepsilon_1 = \varepsilon_h. \tag{3.4.21}$$

Chapter 4

Results And Discussion

Metallic nanoshells possess optically excitable plasmon resonances that can be tuned throughout the visible and into the infrared regions of the electromagnetic spectrum by changing their aspect ratio, r_1/r_2 , where r_1 andr_2 are the dielectric core and metal shell radii of the metallic shell layer, and r_2 - r_1 is the metal shell thickness . In this case, the metal shell of the individual nanoparticle controls its plasmon resonance, thus controlling its local electromagnetic field. Thus, the metal shell thickness represents the distance over which this interaction takes place. Nanoshells exhibit unique optical properties because their interaction with the electromagnetic field is greatly intensified by a phenomenon known as localized surface plasmon resonance . This resonance effect arises from the collective oscillation of the conduction electrons in the silver shell, which efficiently couple to the incident electromagnetic field, and propagate along the surface. The quanta of these surface charge density oscillations is referred to as a surface plasmon polariton. Several distinct phenomena must be connected in order to provide a theoretical framework for understanding the interaction between silver nanoshells and optical electromagnetic fields. First, the dielectric function of silver at optical wavelengths will be introduced, after which the dielectric function of silver will be related to the resonant interaction of silver nanoshells with optical electromagnetic fields. The absorption and scattering properties of nanoshells will then be elucidated by using the quasi-static approximation. Lastly, Mie theory and plasmon hybridization will be introduced as rigorous analytical methods for calculating and understanding their optical phenomena.

The local field enhancement characters in noble metal nanoparticles depend mainly on the shape and size . Furthermore, the surrounding medium dielectric constant also plays an important role. Our studies indicated that, the influence from dielectric shell is different from homogeneous surrounding medium. The local field enhancement at the inner surface of the shell are different from that of the exterior surface. Local field enhancement effect is intense near metal particle and gets damping with increasing the distance from metal surface. Therefore, the peak intensity decreases with increasing the distance from the particle center when the core and shell radius are fixed. Similarly, the peak intensity at exterior surface decreases with increasing the shell thickness.

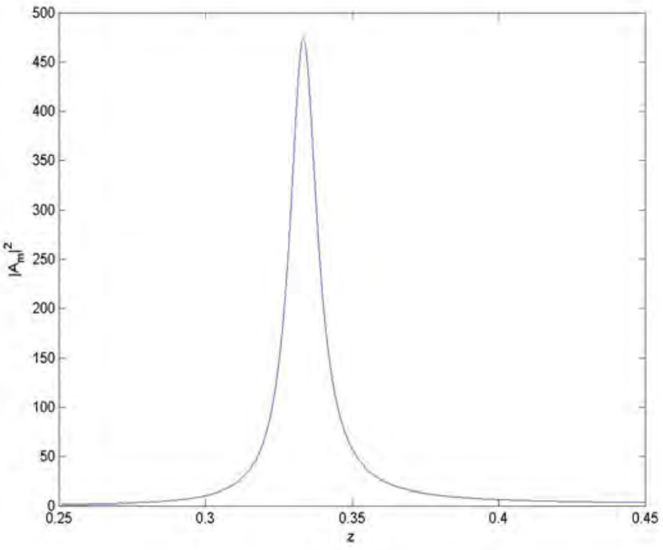

Figure 4.1: The enhancemen factor $\mid A_m \mid^2$ of a pure silver spherical nanoparticles as a function of frequaency, z. We use the following parameters of the system: $\omega_p = 1.46 \times 10^{16}$ (silver plasma frequaency),P $= 1$(metal fraction of the metal shell), $\nu = 1.68 \times 10^{14}$, $\gamma = 1.15 \times 10^{-2}$; $\varepsilon_\infty = 4.5$, $\varepsilon_h = 2.25$.

The maximum enhancement factor of the pure metal with metal fraction (p = 1) has only one resonance at some frequency ranges ($\omega \simeq 0.35\omega_p$). For pure silver nanospheres, the local field enhancemen factor $|A_m|^2$ of Eq.(3.2.16) versus frequency and for metal fraction $p = 1$ is plotted in Fig.(4.1). The local field enhancement characters in silver metal nanoparticles depend mainly on the shape and size.

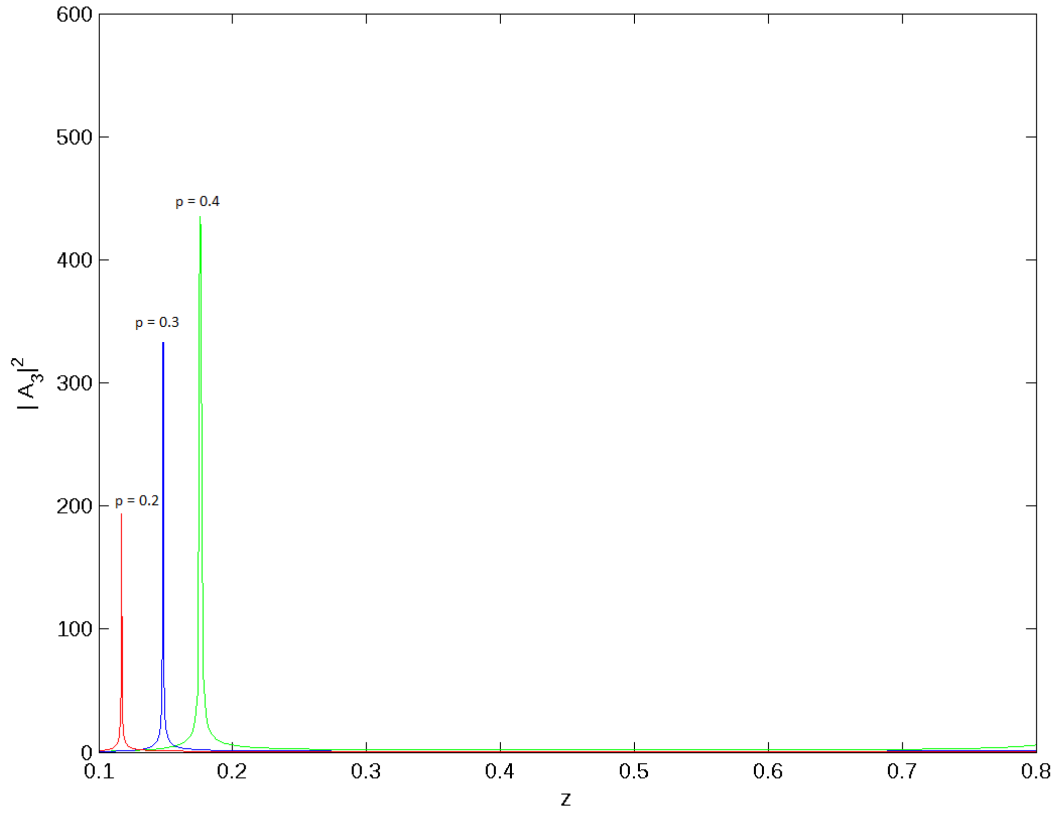

Figure 4.2: The enhancement factor $\mid A_3 \mid^2$ for silver spherical coated nanopacticle versus z at p = 0.2, 0.3, 0.4. The rest parameters are hte same as with in Fig.4.1.

At small p = 0.2, 0.3, 0.4, the second maxima is practically invisible against a background of the first one, which is about 9 times less than the maximum of $\mid A_m \mid^2$. The positions and values of the maxima strongly depend on p (at fixed the rest parameters). And generally for $p < 0.4$, the second maximum is lower than the first one and very samll. The maximum enhancement factor for each value of p has only one resonance at some frequency ranges.

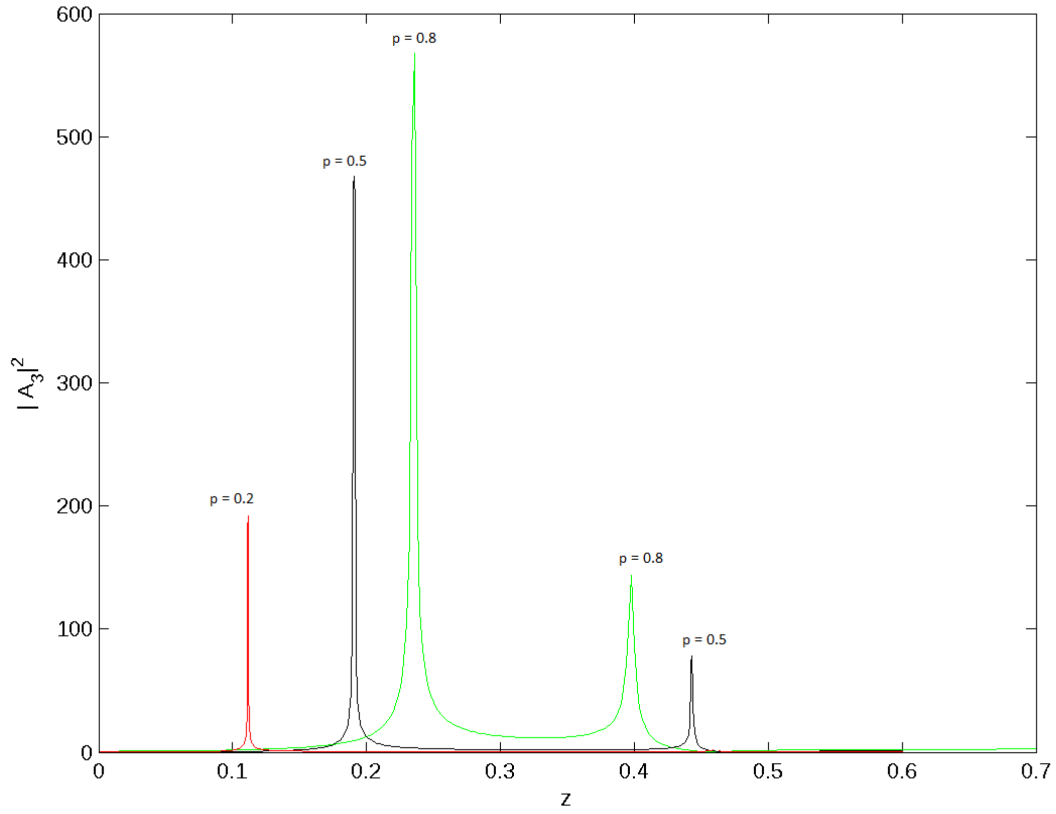

Figure 4.3: The enhancement factor $\mid A_3 \mid^2$ for silver spherical coated nanopacticle versus z at p = 0.2, 0.5, 0.8. Here further we use the following parameters: $\varepsilon_\infty = 4.5$, $\varepsilon_{1o} = 6$, $\varepsilon_h = 2.25$, $\nu = 1.68 \times 10^{14}$ and $\gamma = 1.15 \times 10^{-2}$

.

In Fig.4.3, we present $\mid A_3 \mid^2$ versus z for different p. The curves are obtained with help of Eq.(3.1.10) by neglecting the noalinear terem in ε_1 obtained in Eq.(3.1.11). The numerical parameters of the DFs of the composite are taken from Eq.(3.1.8). The most interesting features of this graph is the appearance of two peaks of the enhancemnt factor at two different frequencies. The positions and the values of the maxima strongly depend on p (at fixed the rest parameters). For metal fraction $p > 0.4$, it becomes more important and with further increment in p both maxima becoming higher and closer moving to higher frequaencies. The maxima of $\mid A_3 \mid^2$ grow very fast with p. Inparticular, at p = 0.8, the first maximum is siightly above the maximum in Fig.4.1.

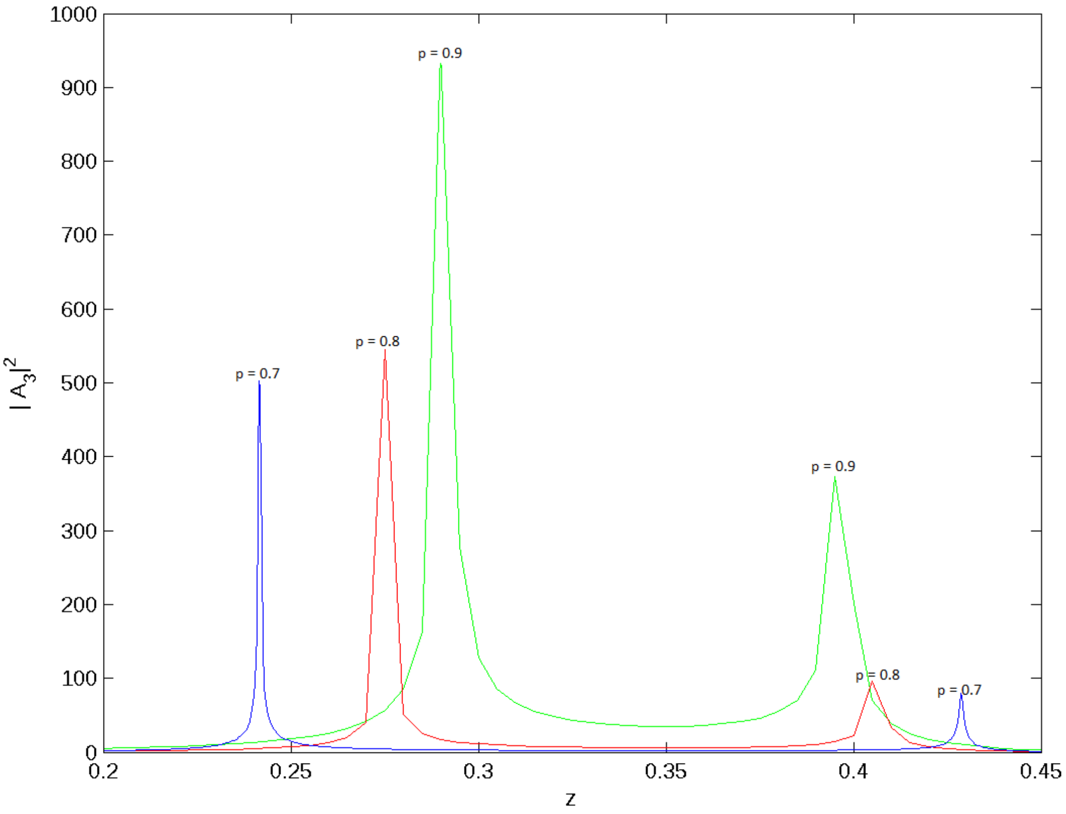

Figure 4.4: The enhancement factor $\mid A_3 \mid^2$ versus z for metal inclusion at different p; $\varepsilon_\infty = 4.5$, $\varepsilon_1 = 6$, $\varepsilon_h = 2.25$, $\omega_p = 1.46 \times 10^{16}$, (frequency of silver surfa plasmons), $\nu = 1.68 \times 10^{14}$, $\gamma = 1.15 \times 10^{-2}$.

The second maximum is practically equal to the first maximum of $\mid A_3 \mid^2$ at p = 0.5. In Fig.4.4, we present $\mid A_3 \mid^2$ versus z for different p.The curves are obtained with help of Eq.(3.1.10) by neglecting the noalinear terem in ε_1 obtained in Eq.(3.1.11). The numerical parameters of the DFs of the composite are taken from Eq.(3.1.8). The most interesting features of this graph is the appearance of two peaks of the enhancemnt factor at two different frequencies. The positions and the values of the maxima strongly depend on p (at fixed the rest parameters). For metal fraction $p \geq 0.7$.

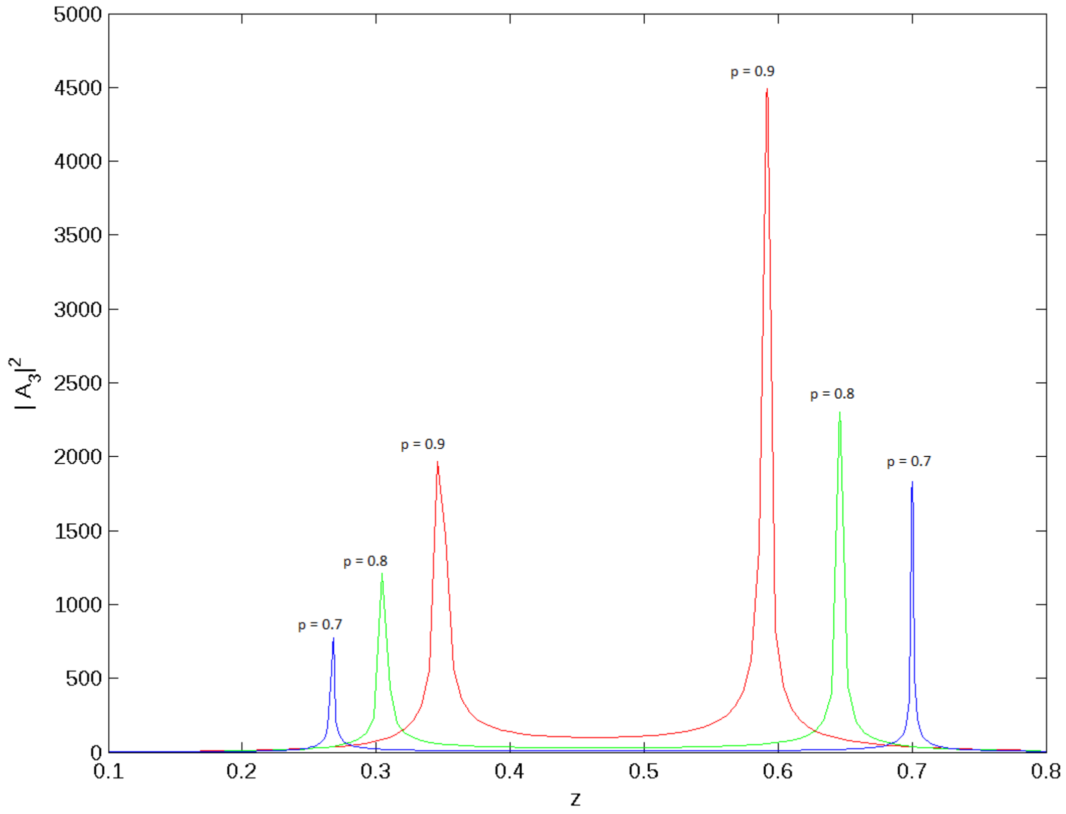

Figure 4.5: The enhancement factor $| A_3 |^2$ versus z for a silver nanparticle at different p; $\varepsilon_\infty = 1$. The rest parameters are the same as in Fig.4.4.

It is interesting to note that at the above parameters of the composites the second (smaller) maximum is blue shifted with respect to the first one. One more parameter that strongly affects the enhancement factors is ε_∞. But choosing $\varepsilon_\infty = 1$ along with the same rest DFs, we obitained that the second maximum $| A_3 |^2$ becomes higher than that of the first one. At the same time, the maxima getting narrower and the corresponding resonance frequencies show a blue shift.

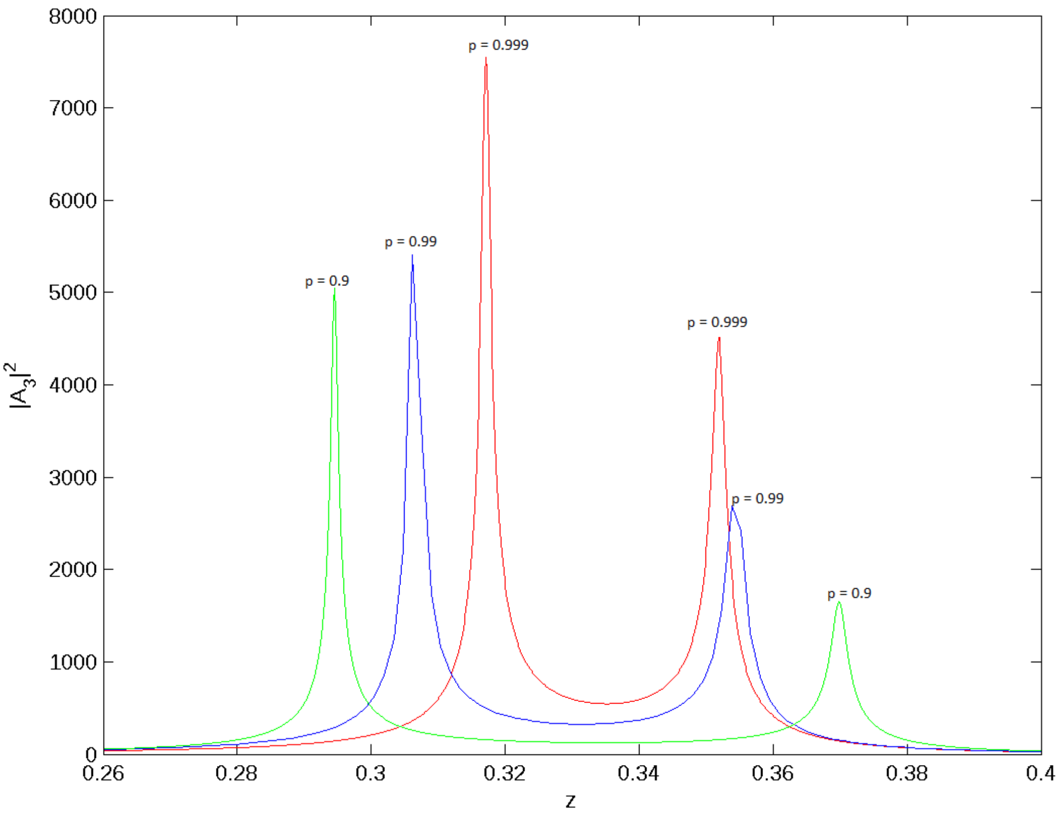

Figure 4.6: The enhancement factor $\mid A_3 \mid^2$ for a silver spherical coated naonparticle versus z at "large" p $=$ 0.9, 0.99, 0.999,or $p \simeq 1$.The rest parameters are the same as in Fig.4.4.

The most interesting inclusions with $0.9 \leq p \leq 0.999$, where absolute values of both maxima considerably large.The enhancement factor of cylinderical inclusions of metal fraction p shows the same behavior. The resonant frequencies are particularlly the same for the spherical and cylinderical inclusions but the absolute values of maxima of cylinderical enhancement factor $\mid A_2 \mid^2$ are 2-4 times less than of the maxima of spherical inclusion $\mid A_3 \mid^2$.The Fig.4.6 plotted the further increment in metal fraction p practically does not change the resonant frequaencies and the absolute values of the maxima of the enhancement factor.

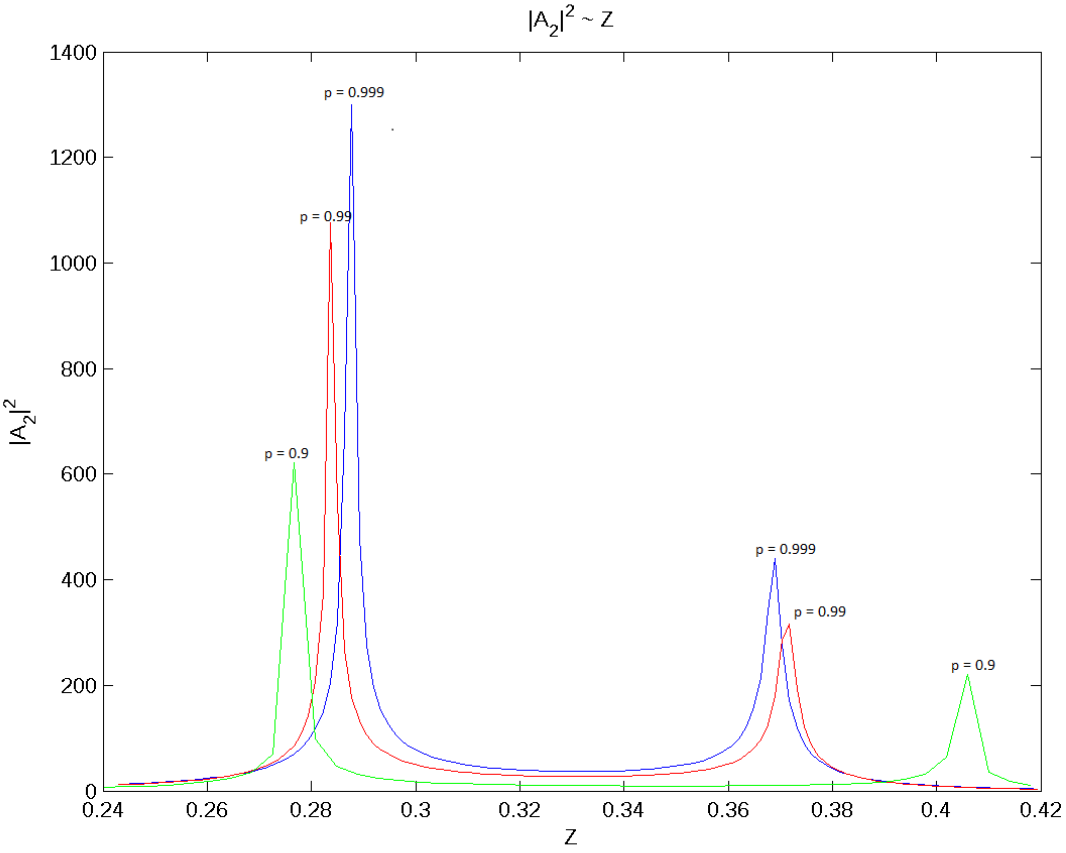

Figure 4.7: The enhancement factor $\mid A_2 \mid^2$ for a silver cylinderical coated nanoparticles versus z for "large" p = 0.9, 0.99, 0.999 or $p \simeq 1$. The rest parameters are the same as in Fig.4.4.

The Figure of 4.7 plotted by using the Eq.3.4.6 of the cylinderical local enhancement factor $\mid A_2 \mid^2$ versus frequaency z. The further increment in metal fraction p practically does not change the resonant frequaencies and the absolute values of the maxima of the enhancement factor. The numerical values shows that at $p = 0.999$, two maxima of the enhancement factor of spherical and cylinderical inclusions merge into one peak.

Its absolute values is about of one order higher than the maxima of the tuned inclusions with p = 0.999 (Figs.4.6,4.7). The maximum value of $\mid A_3 \mid^2$ and the resonant frequecy z_r for some tuned ε_1 and ε_h at $p = 0.999$.

According to the above presented analysis, the further increment in the enhancement factor is possible if we use the "tuned" inclusions when the condition in Eq.(3.2.14) holds true. It gives $\varepsilon_1 = 4\varepsilon_h$ and $\varepsilon_1 = \varepsilon_h$ for spherical and cylinderical inclusions,respectively. It was obitaied for very small decay constant γ and p very close to unit. Furthermore, the surrounding medium dielectric constant ε_h also plays an important role. Local field enhancement features at the core surface of the shell are different from that of the metal shell surface.

The theoretical calculated of the local field enhancement factor $\mid A_3 \mid^2$ of Eq.(3.1.10) for a silver spherical coated nanopacticles versus frequency are plotted in Figs.(4.2-4.6) with different metal fraction ($p = 1-(r_1/r_2)^3$) and constant the rest parameters of the system. The numerical calculations shows that at p = 0.999, two maxima of the enhancemen factor of spherical and cylinderical inclusions merge into one peak. Its absolute value is about of one order higher than the maxima of untuned inclusion with Fig.2.1 . For the nanoinclusion with small dielectric cores one has to check is it possible to use the bulk dielectric prmttivity .Taking inclusions of a radius $\sim$ 20nm and using the relation $r_1 = (1-p)^1/nr_2$ that connected the radiuses of the core and the inclusion we get $r_1 = 2nm$ and $r_1 = 0.06nm$ for the pherical and cylinderical inclusions at the metal fraction ($p = 0.999$), respectively. The existence of two maxima of the enhancement factor of the local filed in metal inclusion with small dielectric cores seems to be a resalt of two interfaces dielectric-metal and metal -host. In particular, for the small dielectri cores, the two resonant frequencies depend only on the ε_∞ and ε_1 or ε_h in Eq.(2.3.4). The two maxima of $\mid A_3 \mid^2$ or two minima in $\mid \Delta \mid^2$, which enters into the polarazibility of the inclusion of Eq.(2.5.4),results in some peculiarties in the frequency dependence of the optical parameters of composites.

Chapter 5

CONCLUSION

Our theoretical calculations of the local filed show that the local field enhancement factor maximum increasing with increasing the metal fraction. And the position and the value of the two maxima strongly depend on the metal fraction p (at fixed rest parameters). The existence of two maxima of the enhancement factor of the local field in the metal inclusions with dielectric cores seems to be a result of the two interfrances, dielectric − metal and metal − host. The resonant frequencies and the enhancement factors of the local filed depend on the composite parameters, ε_∞, ε_1 and ε_h.

In conclusion, we note that the availability of two maxima of the enhancement factors of the local field in metal inclusions with small dielectric core at two diffrerent resonant frequencies and comparatively large amplitudes must result on the small nonlinear properties of the composites of these inclusions. We show that the enhancement factor of the local field in spherical and cylinderical inclusions with dielectric core in a linear host matrix has two maxima at two diffrerent frequencies.

The second maximum corresponding to the higher resonant frequency can be specified and becomes important in inclusions with a large metal fraction p $\geq$0.9.

References

[1] R.W. Boyd, Nonlinear Optics, Academic press, New York, 1992.

[2] A. Szoke, V. Daneu, J. Goldhar,N.A.Kurnit, Appl.Phys.Lett.**15**(1969)376.

[3] H.M. Gibbs, S.L. McCall, T.N.C. Venkatesan, Phys.Rev.Lett. **36**(1976)113.

[4] N. Kalyaniwalla, J.W. Haus, R. Inguva, M.H. Birnboin, Phys. Rev. A **42**(1990) 5613

[5] A.E. Neeves, M.H. Birnboim, J. Opt. Soc. Am. B **43**(1989)787

[6] K.M. Leung, Phys. Rev. A **33**(1986)2461.

[7] O.A. Buryi, l.G. Grechko, V.N. Mal'nev, Sisay Shewamare, Ukr. J. Phys. **56**(2011)311,Sisay shewamare, V.N.Mal'nev, Physical B, v.**401**, pp. 4837-4842(2012)

[8] G.A. Korn and T.M. Korn, *Mathematical Hanabook for Scientists and Engineers* (McGraw-Hill, New York,1968),John Jackson,19999.

[9] C.Kittel, *Introduction to Solid state Phys. Eigth Edition*,(John Wilely Sons,2005).

[10] J.W. Hausa, H.S. Zhou, S. Takami, M. Hirasawa, I. Honma, H. Komiyama, J. Appl. Phys. **73** (1993) 1043.

[11] C.F. Boren and D.R.Huffman. *Absorption and Secattering of Light by small* ‘particles, Willey, N.Y, 1983.

[12] R.W. Boyd, Nonlinear optics, academic prass,N.Y.(1992)

[13] L.D. Landau and E.M. Lifshits, *Electrodynamics of Continuous Media*, Pergamon press,1960.

[14] Tserkezis, C., G. Gantzounis, and N. Stefanou, *Collective plasmonic modes in ordered assemblies of metallic nanoshells*,

J. Phys.: Condens. Matter, Vol. **20**, 075232, 2008.

[15] Chau, Y.-F., *Surface plasmon effects excited by the dielectric hole in a silver-shell nanospherical pair, Plasmonics,* Vol. **4**, 253, 2009.

[16] Okamoto, T., *Near-field Optics and Surface Plasmon Polaritons*, **99**, S. Kawata (Ed.), Springer, Berlin, 2001.

[17] Bohren, C. F. and D. R. Huffman, *Absorption and Scattering of Light by Small Particles*, Wiley, New York, 1983.

[18] Yannopapas, V., *Non-local optical response of two-dimensional arrays of metallic nanoparticles*, J. Phys.: Condens. Matter, Vol. **20**, 325211, 2008.

[19] Mark I. Stockman. Opt. Exp. 2011;19(22): 22029-22106.

[20] Y. Wang, E. W. Plummer and K. Kempa. Advances in Physics 2011;**60(5)**: 799-898.

DECLARATION

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

Name: ***Seyoum Eyasu***

Signature:– – – – – – – – – – – – –

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

Name:***Pro. Maln'v***

Signature:– – – – – – – – – – – –

Place and time of submission:

Department of Physics
Addis Ababa University
June 2013