THEORETICAL APPROACH FOR OPTICAL PROPERTIES OF NANOMATERIALS

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ABSTRACT

Optical properties of nanoparticles have a long and interesting history in material science due to plethora of applicationsassociated with them. In present paper we investigated Green's function based theoretical approach for optical efficiencies of silver and silicon nanomaterial under study from optical spectrum measured experimentally, which provide us beneficial physical information to develop efficient photonic devices in renewable energy technologies, optical switches, modulators , detector, sensing techniques etc. Also our attempt is to analyse the behaviour of these properties with alteration in shape, size as well as surrounding parameters. The numerical simulation outcomes indicate that contribution in the optical efficiencies of both metal and semiconductor nanoparticles are mainly by surface plasmon resonance, intra band transition and dielectric constant.

Keywords: Green Function, Light Scattering, Optical Efficiencies, Silver and Silicon Nanomaterial, Surface Plasmon Resonance.

I. INTRODUCTION

Nowadays scientist are attempt to build the materials with small size, high efficient and lightweight. In this direction they are reached to nanometric dimensions. Generally nanomaterials having at least one external dimension in the size range from 1 to 100 nm. Due to their reduced dimension their physical properties gradually change from their bulk counterparts. At this size range quantum size effect and increase surface-to-volume ratio play important role. These factors can change or enhance properties such as reactivity, strength, electrical, optical and magnetic behavior of materials[1- 2]. Among many important physical properties, optical properties of nanomaterial have always been given special attention. The unique optical properties like absorption, emission, scattering, photoluminescence and electroluminescence properties of nanomaterials result from their discrete electronic energy levels.

Study of light scattering by small particles is an important problem for modern applications in plasmonics and nanotechnologies field. There are lot of literaturefor optical property of small particles in the form of review, articles, books, which contain information from Gustav Mie theory1908 to latest work[3-4,9].Almost all of these theories and method based on electromagnetic behaviour of light as an electromagnetic wave, Maxwell equation are an introductory step for deriving these numerous methods from them.Maxwell introduced four equations on electromagnetism which defining electric field, magnetism, Faraday's law and ampere law respectively. As the light intensity changes periodically with the time, Maxwell's equations are usually used in time harmonic form [15]. The response of the particlematerial to the electromagnetic fields ***E*** and ***H*** of the refracted wave generally consists of four contributions: a polarization ***P***, a magnetization ***M***, a current ***J***, and a displacement current $\partial \boldsymbol{D} / \partial t$.

At any position*r* and time *t*, physical field *E(r, t)* is definite by the complex amplitude as

$\boldsymbol{E}(\boldsymbol{r}, t) = Re\{\boldsymbol{E}(r)\, e^{-i\omega t}\}$(1)

Similarequation holds for the magnetic field $\boldsymbol{H}(\boldsymbol{r}, t)$.

When a light propagation in homogeneous medium is conveniently expressed in terms of electromagnetic plane wave which is time-harmonic field. For a plane wave, the complex amplitude of the electric field is

$\boldsymbol{E}_{pw}(\boldsymbol{r}) = \boldsymbol{E}_0 e^{ik.r}$(2)

Where E_0 is a constant vector and the wave vector k points in the direction of propagation. The length of the wave vector, $k = \omega\sqrt{\varepsilon_h}\,\mu_h$, is called the wave number. Due to deviation of permittivity of a nanoparticle from that of the surroundings, the electromagnetic field, which satisfies time-harmonic Maxwell equations, can be written as a superposition of the scattered field and incident plane wave

$\boldsymbol{E}(r) = \boldsymbol{E}_{pw}(r) + \boldsymbol{E}_{sca}(r)$ (3)

$\boldsymbol{H}(r) = \boldsymbol{H}_{pw}(r) + \boldsymbol{H}_{sca}(r)$(4)

Far away from the nanoparticle, the scattered electric field can always be expressed in spherical coordinates in the following form [16]

$$E_{sca}(r) = \frac{e^{ikr}}{r} F(\theta, \phi) \quad (5)$$

The scattering mechanism can be categorised in two types based on the frequency. The polarisation plane of the incident beam may change light scattering phenomenon. If the frequency or wavelength of scattered radiation is identical respect to the incident light then it is known as elastic scatteringand Mie scattering. This type of scattering takes place when the absolute temperature of particles is near to zero; otherwise the scattered light is emitted in all frequencies which form thermal scattering.

When the light scattering is dependent on the wavelength or frequency of incident beam is called inelastic scattering like Raman scattering, Brillouin scattering, X-rayscattering and Compton scattering[17]. The relation between polarisationP and electric field E of incident light according to Maxwell equationis given by the equation

$$P = \sum \varepsilon_0 \chi E \quad (6)$$

Whereχ is the susceptibility of the matter which depend on $\boldsymbol{E}$ that is,$\chi(\boldsymbol{E})$.

In the paper, we examine Green's function centred theoretical analysis of optical properties nanoparticles which provide a reference for manufacturing and characterisation of novel nanomaterials. Until the optical efficiencies of both metal (sliver) and semiconductor (silicon) nanoparticles certainly not studied earlier together. Ourobjective is to correlate the optical spectra of these nanoparticles to shape and size parameters as well as their material properties. We hope that this paper can be help to determine and optimize nanoparticle physical properties during and after growth. Scattering formalisms based on Green's tensor with free space dyadic Green's function (Cartesian tensor) and dipole approximation employed for numerical analysis and simulation which is used to understand the optical efficiencies of nanoparticles.

II. THEORY AND FORMALISM

If we consider a media that is transparent and homogeneous the only scattering entity is then expected to be the particles. A single scattering is appeared when a single particle interacts with the incident light independently and nearby particles do not affect the scattering efficiency of other ones. In a cluster of single scattering particles

the total scattering efficiency is obtained by the sum of scattering of individual particle, cluster are treated as a single unit and the total efficiency is achieved by the summation of the efficiencies of each cluster[12]. The electromagnetic field of such an arbitrary finite particle changes as that would exist in an unbounded homogeneous space. The total electromagnetic field is equal to the vector sum of the *incident* (original) field and the *scattered* field. The polarization and angular distribution of the scattered field depend on the directional and polarization characteristics of the incident field as well as on such properties of the scatterer as its size relative to the wavelength and its shape, composition and orientation. Therefore, in practice one usually must solve the scattering problem with a new theoretical approach[13].

The incident light is scattered and absorbed by each particle or aggregate in the volume to a certain extent, for the transmitted light is diminished along the propagation direction of the incident light by these absorption and scattering processes. With the extinction and scattering cross - sections, integral properties of the single particle are defined which are a measure of the ability to scatter and absorb light.

The scattering cross sections C_{sca}obtain by total power scattered by the particle divided by the intensity of the incident wave.

$$C_{sca} = \frac{W_{sca}}{I_i} \text{ Or } \qquad W_{sca} = C_{sca} I_i \text{ (7a)}$$

Where I_i is beam intensity and W_{sca} total power scattered by this particle. It can thereby be interpreted as the area, from which the scatterer takes energy off from the incident light and converts it into scattered light. In analogy, the absorption cross section C_{abs} describes the area from which optical energy is absorbed.

$$W_{abs} = C_{abs} I_i \text{ (7b)}$$

Particles absorb as well as scatter electromagnetic radiation.

The third cross section, is extinction cross section C_{ext}, describes the total effect of both scattering and absorption

$$C_{ext} = C_{abs} + C_{scat} \text{ (8)}$$

If we want to obtain cross section efficiencies for above mention individual part then we must divide it by area of scattering particle *S* then extinction, absorption and scattering efficienciesrespectively given by

$Q_{ext} = C_{ext}/S \quad ; \quad Q_{abs} = C_{abs}/S \quad ; \quad Q_{sca} = C_{sca}/S$

In case of single spherical particle optical efficiencies of calculated by Mie theory the following equations.

$$Q_{ext} = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n+1) \Re(a_n + b_n) \text{ (9a)}$$

$$Q_{scat} = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n+1) \Re(|a_n| + |b_n|) \text{ (9b)}$$

$$x^2 = \frac{2\pi r}{\lambda}$$

Where, a_n and b_n Mie scattering coefficient which is calculated numerically,x^2 is size parameter depend on wavelength λ and particle radius *r*, *n* is the refractive index of the particle. In case of clusters the above calculations may need some modification [12-14].

Moreover, the spatial dispersion of the scattered energy by the particles isrelativelyattracting due to its utility as optical nano-antennas [18]. Generally differential scattering efficiency Q_{diff} is defined the angular dependence of this energy

$$Q_{diff} = \frac{1}{x^2}\left\{\left|\sum_n \frac{(2n+1)}{n(n+1)}(a_n\pi_n + b_n\tau_n)\right|^2 + \left|\sum_n \frac{(2n+1)}{n(n+1)}(a_n\tau_n + b_n\pi_n)\right|^2\right\} \quad (10)$$

Here $\boldsymbol{\pi}_n$ and $\tau_n$lead the angular dependency over the Legendre functions of first kind. In the special cases the forward and backward directions the differential scattering efficiencies are represented by the forward scattering Q_{FS} and the radar backscattering Q_{RBS} efficiencies, as [19]:

$$Q_{FS} = \frac{1}{x^2}\left|\sum_n (2n+1)\cdot(a_n + b_n)\right|^2 \quad (11a)$$

$$Q_{RBS} = \frac{1}{x^2}\left|\sum_n (2n+1)(-1)^n(a_n - b_n)\right|^2 \quad (11b)$$

The early two electric and magnetic terms (a_1, a_2 and b_1, b_2) can be used to approximate the multipolar expansion.In this approximation, earlier equation for extinction efficiency of the nanoparticle Q_{ext} change as follows:

$$Q_{ext} = \frac{2}{x^2}\left[3\,\mathrm{Re}(a_1 + b_1) + 5\,\mathrm{Re}(a_2 + b_2)\right] \quad (12)$$

While $Q_{diff}$is given by

$$Q_{diff} = \left\{\left|\frac{3}{2}(a_1 + b_1\cos\theta) + \frac{5}{6}(3a_2\cos\theta + 6b_2\cos^2\theta - 3b_2)\right|^2 + \left|\frac{3}{2}(a_1\cos\theta + b_1) + \frac{5}{6}(6a_2\cos^2\theta - 3a_2 + 3b_2\cos^2\theta)\right|^2\right\} \quad (13)$$

And forward scattering $Q_{FS}$and radar backscattering Q_{RBS} efficiencies, for such small particle expressed as[12]

$$Q_{RBS} = \frac{1}{x^2}\left|-3(a_1 - b_1) + 5(a_2 - b_2)\right|^2 \;;\; Q_{FS} = \frac{1}{x^2}\left|3(a_1 + b_1) + 5(a_2 + b_2)\right|^2 \quad (14a, b)$$

The local electric field can be given as when the scattering object is irradiated by an incident electromagnetic field

$$E_{loc}(r_i, \omega) = E_{inc}(r_i, \omega) - \sum_{i \neq j} g_0(r_i, r_j, \omega)P_j \quad (15)$$

In the Discrete Dipole Approximation method, it is given by

$\boldsymbol{E}_{i,\,local} = \boldsymbol{E}i_{,\,inc} + \boldsymbol{E}i_{,\,dip} = \boldsymbol{E}_0 e^{i\mathbf{k}.\mathbf{r}} - \sum_{ij}\boldsymbol{A}_{ij}\boldsymbol{P}_j$ (16)

Where P is polarization and $E_{inc}(r,\omega)$, $E_{loc}(r,\omega)$ are the incident and local field respectively[11] ;

$P_j = \alpha_j \cdot E_{loc}$; $\alpha_j = \dfrac{\left[\varepsilon_s(r_j,\omega) - \varepsilon_a(r_j,\omega)\right]}{\left[\varepsilon_s(r_j,\omega) + 2\varepsilon_a(r_j,\omega)\right]}\dfrac{3V_j}{4\pi}$ and V_j is the volume of the jth dipole scattering particle,$\boldsymbol{A}ij$is the dipole-dipole interaction matrix. Once we have solved the 3N-coupling complex linear equations given by $\boldsymbol{P}_i = \alpha_i \boldsymbol{E}_{i\,loc}$, have each dipole moment [22]. $g_0(r,r,\omega)$ show the Green's tensor for an infinitely homogeneous surrounding media. If we consider surface effects then the Green's tensor represented by

$g\,(r, r', \omega) = g_0(r,r',\omega) + g_s(r, r',\omega)$. Here $g_s\,(r, r', \omega)$ denotes Green's tensor corresponding to the surface effects [20]. The surface effects play important role to determine the optical nature of metal nanoparticle.

When a light (electromagnetic) wave striking on the metal surfaces of smaller penetration depth, then freeelectrons on the surface are most significant and which leads an induced secondary electromagnetic flow on the surface of particle their collective oscillations are known as surface plasmon and surface plasmon resonance

intensify optical absorption and scattering efficiencies due to the movement of free electrons at the surface.The extinction cross section C_{ext}, which is composed of absorption and scattering, is given by

$$C_{ext} = \frac{24\pi^2 R^3 \varepsilon_m^{\frac{3}{2}}}{\lambda} \frac{\varepsilon''}{\left(\varepsilon' + 2\varepsilon''\right)^2 + \varepsilon''^2} \quad (17)$$

Where R is the radius of the particle, $\lambda$the wavelength ofthe incident electromagnetic radiation, $\varepsilon'$and $\varepsilon''$are thereal and imaginary parts of the dielectric constant of the particles respectively, and e_m is the dielectric constant ofthe embedding medium

Extinction would be maximum when $\varepsilon' + 2\varepsilon'' = 0$,which gives rise to a surface plasmon resonance (SPR)band. The position of the SPR band is dependent on thesize and shape of the particle and dielectric constant ofthe medium in which the particles are dispersed [21].

For knowingthe entire optical characteristics of metal particles their absorption, scattering and extinction efficiencies necessarily considered.The optical efficiencies for a target given in terms of incident field and polarisation are expressed as follows[12].

$$C_{ext} = \frac{4\pi k}{|E_{inc}|^2} \sum_{i=1}^{N} \mathrm{Im}\left(E^*_{inc,i} \cdot P_i^*\right) \quad (18a)$$

$$C_{abs} = \frac{4\pi k}{|E_{inc}|^2} \sum_{i=1}^{N} \left\{ \mathrm{Im}\left[P_i \cdot \left(\alpha^{-1}\right)^* P_i^*\right] - \frac{2}{3} k^3 |P_i|^2 \right\} \quad (18b)$$

Here * denote the complex conjugate.

The scattering cross section can be found by the following relation, $C_{sca} = C_{ext} - C_{abs}$.

Let us suppose a finite scattering body in the form of a single object or a fixed aggregate embedded in an infinite, homogeneous, linear, isotropic, and non-absorbent medium; to obtained the scattering field $\mathbf{E}_{sca}(\mathbf{r})$, we launch a free space dyadic Green's $G(\mathbf{r}, \mathbf{r}')$ function as a dyadic Cartesian tensor satisfying the differential equation

$$\nabla \times \nabla \times \overleftrightarrow{G}(r, r') - k_1^2 \overleftrightarrow{G}(r, r') = \overleftrightarrow{I}\delta(r, r') \quad (19)$$

Where $\overleftrightarrow{I}$ is the identity dyadic and $\delta(\mathbf{r} - \mathbf{r}') = \delta(x - x')\,\delta(y - y')\,\delta(z - z')$ is the 3-D Dirac delta function. This operation may be thought of as a 3×3 matrix representing the dyadic multiplying a column matrix consisting of the initial vector components, thereby producing another column matrix consisting of the resulting vector components. The components of both vectors mustbespecified in the same coordinate system. By the mathematical calculation we get *scattered field* Esca(r) in the terms of free space dyadic Green's G(r, r′) function and the forcing function j(r).

$$E_{sca}(r) = \int dr' \overleftrightarrow{G}(r, r') \cdot j(r') \quad (20)$$

While complete solutionfor total field given by

$$E_{total}(r) = E_{inc}(r) + E_{sca}(r) = E_{inc}(r) + \int dr' \overleftrightarrow{G}(r, r') \cdot j(r') \quad (21)$$

To find the free space dyadic Green's function $G(\mathbf{r}, \mathbf{r}')$,we first express it in terms of a scalar Green's function $g(\mathbf{r}, \mathbf{r}')$ as follows:

$$\overleftrightarrow{G}(r, r') = \left(\overleftrightarrow{I} + \frac{1}{k^2} \nabla \otimes \nabla\right) \cdot g(r, r') \quad (22)$$

After calculation Equation (21) expresses the total electric field everywhere (interior or exterior of scattering body) in space in form of the incident field and the total field inside the scattering body [13].

$$E_{total}(r) = E_{inc}(r) + k_1^2 \int dr' \overleftrightarrow{G}(r,r') \cdot E(r') \left[m^2(r') - 1\right]$$

$$= E_{inc}(r) + k_1^2 \left(\overleftrightarrow{I} + \frac{1}{k^2} \nabla \otimes \nabla \right) \int dr' \left[m^2(r') - 1\right] E(r') \frac{e^{ik_1|r-r'|}}{4\pi|r-r'|} \quad (23)$$

The total electric field computed by the Rayleigh-Gans approximation,for improvement approximation can be continued until the total field converges within a given numerical accuracy and after final analysis it can be expressed in terms of the incident field as follows;

$$E_{total}(r) = E_{inc}(r) + \int dr' \overleftrightarrow{G}(r,r') \cdot \int dr'' \overleftrightarrow{T}(r',r'') \cdot E_{inc}(r'') \quad (24)$$

Where dyadictransition operator $\overleftrightarrow{T}$ (Tsang *et al*. 1985) obtained in the integral form by solving equation (21), (23) and (24)

$$\overleftrightarrow{T}(r,r'') = k_1^2 \left[m^2(r) - 1\right] \delta(r-r') \overleftrightarrow{I} + k_1^2 \left[m^2(r) - 1\right] \int dr'' \overleftrightarrow{G}(r.r'') \cdot \overleftrightarrow{T}(r'',r') \quad (25)$$

This kind of expressions appears in the quantum theory of scattering, are called Lippmann-Schwinger equations.

III. RESULT AND DISCUSSION

Optical efficiencies (scattering efficiency, absorption efficiency and extinction efficiencies) of metal and semiconductor nanoparticles are studied as a function ofλ with different geometries like spheres, cylinder and different sizes. By altering the shapes and sizes of nanoparticles, we can derive new optical characteristics, and produce new nano-materials to serve the requirements of humanity.

Figure 1 shows that the optical efficiency of silver sphere with radius ~ 40 nm, we can observe thatat about 320 nm all the *Qext* spectra have a local minimum that corresponds to the wavelength at which intra-band electron transitions on silver start because of both the real and imaginary parts of the Ag dielectric parameter almost reach zero at that wavelength.

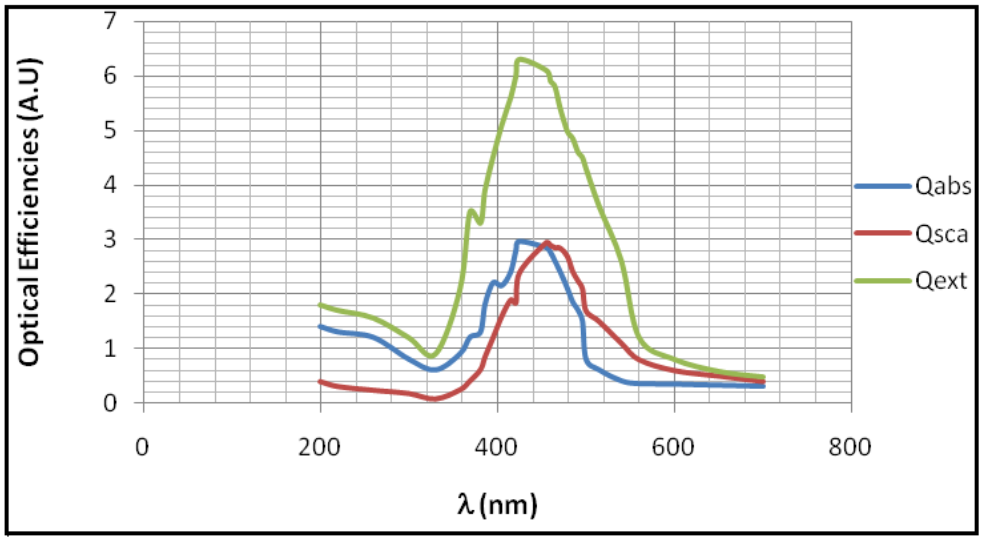


Fig.-1 Optical Efficiency of A Silver Nanosphere Radius (40nm) Vs Wavelength

As seen clearly from Fig. 1 for range 300nm <λ<520 nm the contribution of both the scattering and absorption efficiencies given to the extinction efficiency.

The absorption spectrum shows some peaks for λbetween 320nm and 400 nm, these peaks are due to the SPR excited in the nanoparticles, which are associated with the plasmon resonances and inherent to the nanosphere geometries.The peaks location and intensity depend on the particular geometries of each nanoparticle. There is a strong and large peak at about 440nm wavelength, this is because both the scattering and absorption efficiencies are strong, and the metallic surface is ex-cited by surface plasmon resonance when the silver nanoparticles are irradiated by light. However, for larger wavelength, the spectrum for the nanoparticle decays very quicklyas the increase of their size, while the spectraof the other nanoparticles do not show the same behaviour.

Spectral feature of optical efficiency versus wavelength of silver nano cylinder displays Fig. 2as seen; we observe that the surface plasmon resonance is complicated with its sizes and geometries and this characteristic can used to achieve surface enhancement Raman scattering(SERS) for studying molecule structure[5,6].

In SERS mechanism, surface plasmon resonance plays important role. Metal surface contributes the required condition to produce optical surface coupling, electrons in the metal surface are excited to higher bands and are resonant with the electromagnetic fields of the light, and enhance electrical fields near the metal surface to result in enhanced Raman scattering.

Fig.2.show that theextinction efficiency is independent from the scattering efficiency when wavelength below 440 nm, while in the range 440nm <λ< 700 nmscattering efficiency has a long tail which is sign of its contribution to the extinction efficiency. However the absorbing efficiency is the primary contributed to the scattering efficiency as a cylinder in Fig. 2. If we study only the extinction efficiency, it is impossible to observe the properties of absorbing and scattering efficiency, because they are hidden.

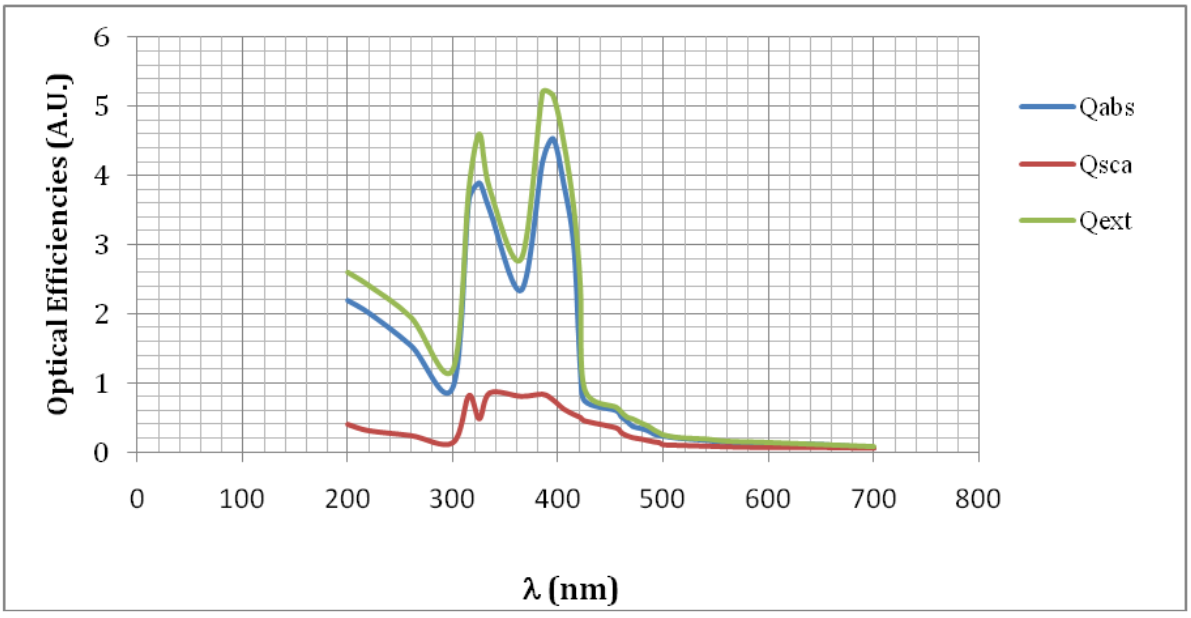
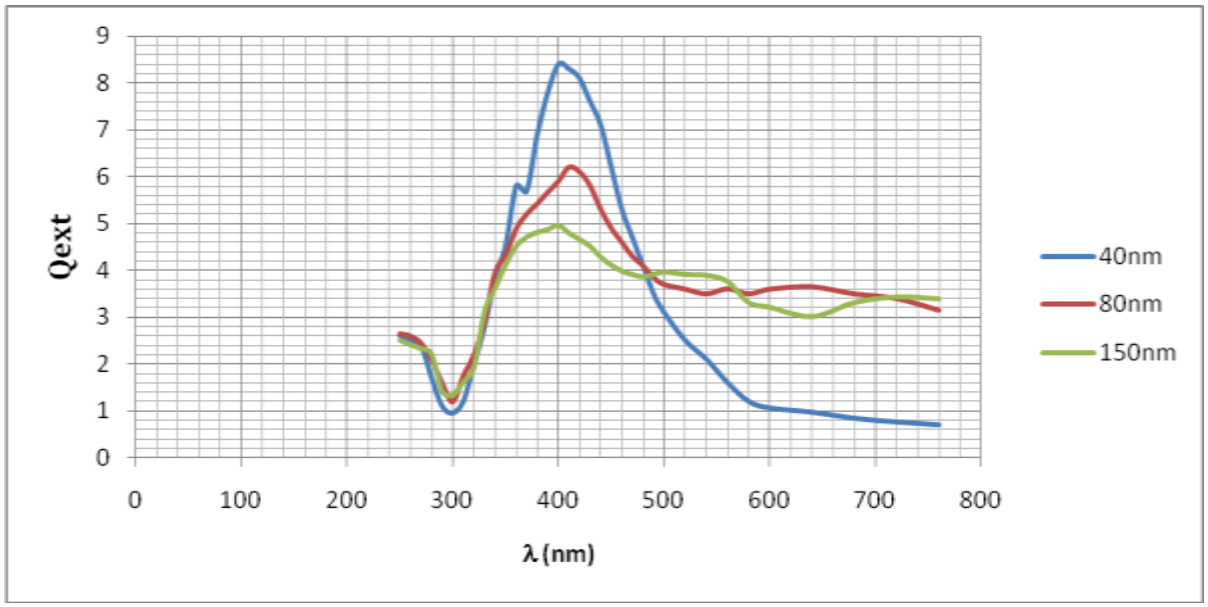


Fig. 2Optical Efficiency As A Function of Wavelength of Silver Nanocylinder

Fig.3 shows *Qext* as a function of λ for silver spheres of 40 nm, 80 nm and 150 nm with 65,000 dipoles. We can observe at ~300 nm all *Qext* spectra have minima that due to the wavelength at which intra-band electron transitions on silver start as earlier mention also these transitions give rise to maxima at about ~ 400 nm as clearly seen in fig.3.



Furthermore many experimental calculation express as increment in the number of dipoles dramatically from few thousands to lakh, peaks appears in plot is gradually eliminated. It is also reduces withsize of the nano-particle increases or with decreasing in wavelength.

Intensity of light scattering is less for small radius nanospheres, it is according to our expectation. On the basis of these considerations, we can explain the *Qext* spectra for wavelength greater than 350nm as light scattering effects for the spheres larger than 40 nm.

Optical efficiency of semiconductor nanoparticles (mostly germanium and silicon) having radius below 500nm range, has attractive and complicated spectral nature compare to plasmonic nanoparticles because of displaying together electric and magnetic resonances [7-8]. Although, similarity is both depend on the size, shape of particles and the refractive index of the neighbouring medium. Thus scattering properties of semiconductor nanoparticles can be tuned by use of active mediums (such as liquid crystals).

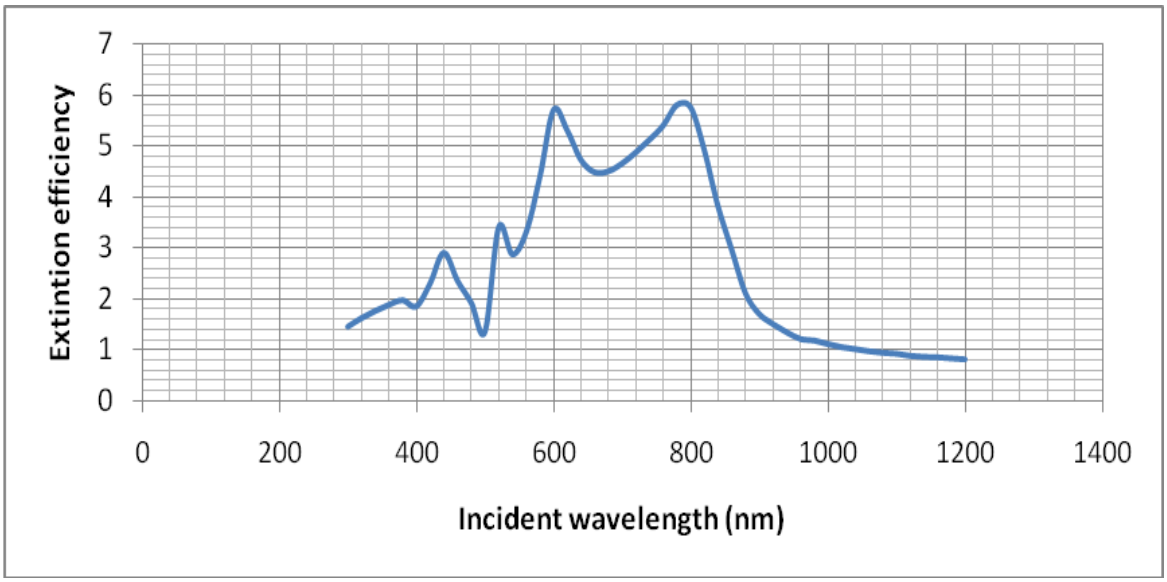


Figure 4 Shows the Extinction Efficiency Qext, of Silicon Nanoparticle with Radius of 100nm Embedded in a Nematic Liquid Crystal

In above figure the extinction efficiency Q_{ext}, of Si nanoparticle is consideredwhere Liquid Crystal is taken in ordinary state.As plot shown thevarious resonant peaks which are mostly presented in light scattering, unlike metals whose resonances are mainly related to the absorption[10].

The contribution of electric (a_1, a_2) and magnetic term(b_1 and b_2) to extinction pattern of semiconductor nanoparticles(i.e. silicon) plays dominant role and produces a slight modification from the ordinary to the extraordinary state with change in refractive index.It is investigated; due to interferential phenomena betweenboth components create anisotropy in the angular distribution of the scattered radiationwhile in light scattering of semiconductor nanoparticles it permits the appearance of coherent effects.

IV. CONCLUSION

By using Green's tensor the important optical characteristics of metal (silver) and semiconductor (silicon) nanoparticles have been studied.We investigated the special features of spectra of optical efficiencies forboth semiconductor and metal nanoparticles with varying sizes, shapes, wavelength and its surrounding medium which provide valuable informationin the particles growth and characterization.

Noble metal nanoparticles taken as special attention throughout the world recent years because of it commenced new areas for research in materials science; likein medical diagnosis for imaging cancer cells, therapeutic and treatment utilities, ecological investigation, Raman scattering and optics. Although, progress in this field is still in the initial phase and for utilizing these particles, smart and thorough studies are required. In this study we revealed the role of surface plasmon resonance, in optical efficiencies of metal and semiconductor nanoparticles up to its contribution in Raman scattering. We examinedthe variation in directional behaviours of light scattering of semiconductor nanoparticles embedded in a Liquid crystal. The resonant behaviours of semiconductor nanoparticle during light scattering are significant stimulus for Plasmonics.

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