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A THEORETICAL STUDY OF LOCALIZED PLASMON RESONANCES IN METAL NANOPARTICLES AND DISPERSION RELATION OF SPP (SURFACE-PLASMON-POLARITONS) BOTH AT INTERFACE AND MDM WAVEGUIDE

SACHIDANAND SINGH[2], L. K. MISHRA[1]

1. Department of Physics, Magadh University, Bodh-Gaya-824234 (Bihar).
2. Professor in charge, Mahila Mahavidalaya, Daudnagar (Aurangabad-824143 (Bihar).

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Abstract: - Using the theoretical formalism of **Maier and Atwater (J. Appl. Phys, 2005)**, we have theoretically studied the localized Plasmon resonances in metal nanoparticles. We have evaluated the absorbance (ab. Units) as a function of energy for thin gold film, 30nm Au nanoparticle. We have also presented the evaluated results of light scattering spectra as a function of photon energy (eV) of Au nanocrystal 100nm. This study gives the possibility of localization of electromagnetic energy in one, two and three dimensions. This can also be applied in sensing and wave guiding for fundamental photonic devices. Our obtained results of dispersion relation of SPPs both at interface and MDM waveguide are in satisfactorily agreement with the experimental data and also with other theoretical workers.

Keywords: Surface-plasmon polariton, Surface enhanced spectroscopy, Coherent electron oscillations, Surface enhanced Raman spectroscopy, Metal nanostructure, Nanoparticle, Nanocrystals, Localization and guiding of electromagnetic energy, Time resolved pump-probe measurements, Second harmonic generation, Light scattering spectra, Far-field dipole interactions, Noval metamaterials.

Corresponding Author: MR. SACHIDANAND SINGH

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1. INTRODUCTION

Optical fibers now span the globe, guiding light signals that convey voluminous streams of voice communications and vast amount of data. This capacity has led some researchers to look about that photonic device which channel and manipulate visible light and other electromagnetic waves to replace electronic circuits in microprocessors and other computer chips. Unfortunately the size and performance of photonic devices are constrained by the diffraction limit. This is because of interference between closely spaced light wave the width of the optical fibre carrying them must be at least half the light wavelength inside the material. Scientists have been working on a new technique for transmitting optical signals through minuscule nanostructure material[1,2]. It has been confirmed experimentally that directing light waves at the interface between a metal and dielectric (a nonconductive material such as air and glass) can, under the right circumstance, induce a resonant interaction between the wave and mobile electrons at the surface of the metal. In other words the oscillations of the electrons at the surface match with those of the electromagnetic field outside the metal. The results is the generation of surface Plasmons-density wave of electrons that propagate inside the interface like the ripples that spread across the surface of a pound after one throw a stone inside the water.

Over the past decade investigators have found that by actively designing metal-dielectric interface they can generate surface plasmons with the same frequency as the outside electromagnetic waves with much shorter wavelength. This phenomenon could allow the plasmons to travel along nanoscale wires called interconnects, carrying information from one part of the microprocessor to another. Plasmonic interconnects would be a great boon for chip designer who have been able to develop ever smaller and faster transistors. It may be possible to employ plasmonic component in a wide varieties of instruments which can be used to improve resolution of microscope, the efficiency of light emitting diodes (LEDs) and the efficiency of chemical and biological detectors. Scientists are also considering medical applications, designing tiny particles that can use plasmonic resonance absorption to kill cancerous tissues. It was also theorized that certain plasmonic materials can alter the electromagnetic field around an object to such an extent that it would become invisible. In this field it was discovered that one can use Raman spectroscopy for observing the scattering of laser light off a sample to determine its structure from molecular vibrations. This field got another boost with the discovery of noval metamaterials. This new materials are capable of having electron oscillation which can result optical properties. This has made possible to build up and test ultra small plasmonic devices and connects. At first glance, the use of metallic

structures to transmit light signals seems impractical because metals are known for high optical losses. The electrons oscillating in the electromagnetic field collide with the surrounding lattice of atoms rapidly dissipating the field energy. But the Plasmon losses are lower at the interface between a thin metal film and a dielectric than inside bulk of the metal.

The electromagnetic properties of metal/dielectric interface have attracted a vast amount of research effort ever since the work of Mie[3] and Ritchei[4] for small particles and flat interfaces respectively. The ability of such structure to sustain coherent electron oscillations is known as surface-Plasmon polaritons (SPPs). This leads to electromagnetic confinement on the metallic surfaces. This field has been intensively investigated[3,4] both in the light of fundamental physics involved and for applications such as surface enhanced spectroscopy and enhancement of nonlinear light generation. After initial studies of the physics of these excitations in 1980, SPPs started to attract the attention of chemists, as the electric field enhancement around metal nanostructures was found to be crucial for surface-enhanced Raman spectroscopy. Recently, the development of nanofabrication techniques such as electron-beam lithography, ion-beam milling with modern nanocharacterization technique has lead to a resurgence of interest in this field[5]. This is because of potential applications for creating sub wavelength optical devices. The unifying physical processes enabling light localization and guiding in such structures are SPPs excitations[6].

The strong interaction of microscopic metal particle of dimensions below 1μm with visible light has been studied. The optical properties of metal nanoparticles particularly those of noble metals Au, Ag and Cu show differences relative to their bulk or thin film optical responses. The resonant electromagnetic behavior of noble-metal nanoparticles is due to the small particle volume. For a particle with diameter $d<<\lambda$, the conduction electrons inside the particle move all in phase upon plane-wave excitation with radiation of wavelength λ. This leads to the buildup of polarization charge on the particle surface. These charges act as an effective restoring force allowing for a resonance to occur at a specific frequency- the particle dipole Plasmon frequency. In general, the spectral position, damping and strength of the dipole as well as the higher order Plasmon resonance of single metal nanoparticles depend on the particle material, size, geometry and the dielectric function of the surrounding host. One can study interacting particle ensembles as a basis for applications of metal nanoparticles in optical devices. In such type of study there is an enhancement of local fields around metal nanoparticles structure. This can be used for sensing and nonlinear applications[7-14].

One study interface Plasmon polaritons at metal/dielectric boundaries. In a metal/air boundary plasma loss for electrons occur at a frequency $\frac{\omega_p}{\sqrt{2}}$. Here ω_p is the characteristic bulk-Plasmon energy of respective metal. This lowering of the plasma resonance is due to depolarizing effect of the flat surface, analogous to the case of localized Plasmons in metallic nanoparticle. However for excitations with fast electrons plasma waves at flat interface do not propagate (group velocity v_g=0) SPPs at lower energies exhibit a significant dispersion with wave vector **k** due to retardation effect[15].

In this paper, we have theoretically studied the localized Plasmon resonances in metal nanoparticles. We have evaluated the absorbance (arbitrary units) as a function of photon energy for thin gold film, 30nm Au nanoparticle. We have also presented the evaluated results of light scattering spectra as a function of photon energy (eV) of Au nanocrystal 100nm. In another work, we have shown the measured extinction spectra (abs. units) of 2D array of Au nanoparticle as a function of wavelength (nm) for different d. d is the distance between adjacent nanoparticles. Our evaluated theoretical results are in good agreement with other theoretical workers[16-18].

2. MATERIALS AND METHODS

For a spherical metal nanoparticle of radius a<<λ embedded in a non-absorbing surrounding medium of dielectric constant ε_m then we have expression for particle polarizability α

$$\alpha = 4\pi a^3 \frac{[\varepsilon - \varepsilon_m]}{[\varepsilon + 2\varepsilon_m]} \qquad (1)$$

Where the complex ε=ε(ω) is the dispersive dielectric response of the metal. For ellipsoidal particle with principal axes a, b and c, an analogous expression can be obtained in the quasi-static approximation via introducing geometrical depolarization factor L_i these axis

$$\alpha = \frac{4}{3}\pi abc \frac{[\varepsilon - \varepsilon_m]}{[\varepsilon + L_i(\varepsilon - \varepsilon_m)]} \qquad 2(a)$$

$$\Sigma L_i = 1 \qquad 2(b)$$

For spherical particle, L_1=L_2=L_3=1/3. For spheroidal particle (L_1=L_2). The Plasmon resonance splits into two modes, (i) Long axis mode (Polarization parallel to the long axis) (ii) Short axis

mode (Polarization perpendicular to the long axis). For non linear applications and surface enhanced Raman sensing the local field E_{local} close to the metal surface should be maximized. The local field enhancement factor L is given by

$$L=E_{local}/E_0 \qquad (3)$$

Where E_o is the amplitude of the incoming field. For single nanoparticle it is the product of two functions

$$L= L_{SP}(\omega)L_{LR} \qquad (4)$$

Here L_{SP} is the surface Plasmon resonance of the whole particle and L_{LR} is the lighting rod effect. For a perfect spherical particle surface Plasmon resonance contributes to the enhanced process

$$L \alpha Q \alpha T_2 \qquad (5)$$

This is the absorption dominated regime. T_2 and Q are rephrasing time. T_2 is determined both using time-resolved pump-probe measurements[19-22] and higher harmonic generation[23]. Now the total enhancement of second-harmonic generation on rough silver surface scale upon resonance

$$\neg\ L(\omega)^4\ L(\omega)^2 \qquad (6)$$

$$\neg\ L(\omega_{exi})^2\ L(\omega_{RS})^2 \qquad (7)$$

Where ω_{exi} is the excitation frequency and ω_{RS} is Raman Stoke frequency. One calculates the dispersion relation of surface Plasmon polariton propagating at a flat interface using a simple boundary condition analysis for electromagnetic surface waves[3]

$$K_x = \frac{\omega}{c}\left[\frac{[\varepsilon(\omega)\varepsilon_2]}{(\varepsilon(\omega)+\varepsilon_2)}\right] \qquad (8)$$

Here $\varepsilon(\omega)$ is the (complex) dielectric function of the metal and ε_2 is the dielectric constant of the adjacent dielectric half space. We have frequency of the SPPs as

$$\omega_{sp} = \omega_p(1+\varepsilon_2)^{\frac{1}{2}} \qquad (9)$$

for large wave vector. Here ω_p is the bulk Plasmon energy[24,25].

3. RESULTS AND DISCUSSION

In this paper, we have studied the localized the Plasmon resonances in metal nanoparticles. The evaluation has been performed with the help of theoretical formalism of S. A. Maier and H. A. Atwater[26]. In table T1, we have shown the evaluated results of absorbance (arbitrary units) as a function of energy (eV) for thin gold film, 30nm Au nanoparticle in water using classical electromagnetic theory[3]. Results were verified by an aqueous solution of 30nm Au collides. These results show a striking difference between the optical response of the thin film and nanoparticles. The film absorbs light throughout the near- infrared and visible regions due to free electron absorption but for nanoparticle this process is strongly quenched for energies lower than 2eV. In Table T2, we have presented the evaluated results of light scattering spectra (Scattering intensity in abs. unit) as a function of photon energy (eV) of Au nanocrystal 100nm. These results indicate direct visualization of spectral properties of single particle. In Table T3, we have shown the measured extinction spectrum (abs. unit) of 2D arrays of Au nanoparticle as a function of wavelength (nm) for different d.'d' is the space between adjacent nanoparticles. The results were calculated for d=500nm, 600nm, 700nm and 800nm. The measured results show that as the wavelength (nm) increases the magnitude of the extinction spectra increases with increasing d. It has a peak for each d at a particular value of λ. The peak is highest for d=500nm and lowest for d=850nm. In table T4, we have presented the measured result of decay time (fs) of the collective dipole Plasmon mode as a function of grating constant (nm) due to far-field dipole interactions. The decay time was measured on the grating constant d for a regular square array of 150nm diameter Au nanoparticles. Both the variation of the spectral position and width of the resonances can be explained by assuming far-field dipole interactions. Here the ensemble acts effectively as a grating reading to increase radiation damping of the collective resonances[27]. In Table T5, we have shown the evaluated results of dispersion of surface Plasmon-polaritons propagating at a Ag/air, Ag/glass and Ag/Si interface. This calculation is performed using a simple boundary condition analysis for electromagnetic surface waves[3] with equation (8). Coherent electron oscillations lead to enhanced local fields at this surface of metallic structure and also at flat interfaces. As is well known that the interface between a metallic film and a dielectric can sustain SPPs in the form of coherent longitudinal charge oscillations of the conduction electrons, this leads to a surface waves confined within one dimension perpendicular to surface[3]. For lower wave vectors, surface Plasmon –polaritons can be excited by TM-polarized light providing that the retarded-induced momentum mismatch is compensated. The main techniques for achieving this momentum matching are prism coupling, coupling via surface grating or roughness (defects) and using focused optical excitation. In Table T6, We have shown the evaluated results of dispersion relation of coupled

Plasmon mode on a thin Ag-film embedded in glass. S_b is odd bound mode and a_b is even bound mode. S_b is high frequency odd mode which shows an asymmetric distribution of the component of the electric field in the propagation direction (longitudinal component associated with collective electron oscillation). a_b is low frequency even mode [28]. Both these modes exhibit marked differences with decreasing film thickness. This is due to different symmetries of their spatial field distributions. The odd S_b mode shows a mode of the longitudinal electric field component in the symmetry plane of the metal film. On the other hand the bound even mode a_b shows the opposite behavior with respect to both propagation constant and attenuation. Its field becomes more and more confined to the metal with decreasing film thickness. Since a mode with an asymmetric transverse electric field component cannot exist in a homogeneous background material. Both bound modes do not show cutoff thickness for a symmetric environment.

In Table T7, we have presented an evaluated result of attenuation distance of odd mode S_b and even mode a_b as a function of wavelength for different thickness of thin Ag-film embedded in glass. The low attenuation of the S_b mode for small thickness allows propagation distance for surface Plasmons one to two orders of magnitude longer than those of thick metal film[29]. This can be used as surface sensing. S_b mode is the mode of decreasing attenuation with film thickness. There is some recent calculation[30-42] which also reveals the same facts.

4. CONCLUSION

From our theoretical investigations and findings, we have come across the following conclusions;

(i) We have studied the physics of surface-plasmon excitations at metal/dielectric interfaces. There is possibility of using such excitations for the localization of electromagnetic energy in one, two and three dimensions.

(ii) This physics can be applied in sensing and wave guiding for fundamental plasmonic devices.

(iii) plasmonic structures and optically active media. Prominent applications will be mid infrared quantum cascade laser where SPPs are confined to the boundary between a gain medium and a thin metallic layer.

Table T1: An evaluated result of absorbance (Arbitrary Units) as a function of energy (eV) for thin gold film, 30nm Au nanoparticles in water using classical electromagnetic theory Results were verified by an aqueous solution of 30nm Au colloids

Energy (eV)	<----Absorbance (abs. Unit)-------→		
	Thin film	30nm Au nanoparticle (Mie Theory)	Measured value of an aqueous soln of 30nm Au colloids
0.5	0.85	0.027	0.008
1.0	0.72	0.058	0.052
1.5	0.64	0.098	0.187
2.0	0.57	0.876	0.809
2.5	0.48	1.102	1.128
3.0	0.53	0.947	1.106
3.5	0.67	0.823	0.802
4.0	0.76	0.727	0.938
4.5	0.87	0.686	0.967
5.0	0.98	0.897	1.023

Table T2: An evaluated result of light scattering spectra (Scattering intensity in abs. unit) as a function of photon energy (eV) of Au nanocrystal (100nm)

Photon Energy (eV)	Scattering intensity (abs. unit)
1.4	0.258
1.5	0.362
1.6	0.789

1.7	0.726
1.8	0.689
1.9	0.587
2.0	0.432
2.1	0.368
2.2	0.276
2.3	0.216
2.5	0.208

TableT3: A measured extinction spectrum (abs. unit) of 2D Square arrays of Au nanoparticles as a function of wavelength (nm) for different d (d is the space between adjacent nanoparticles

Wavelength	**<-----Excitation spectrum (abs. unit)---→**			
(nm)	d=500nm	d=600nm	d=700nm	d=850nm
600	0.027	0.016	0.208	0.217
620	0.136	0.158	0.262	0.222
650	0.242	0.226	0.367	0.359
700	0.678	0.365	0.402	0.432
720	0.885	0.423	0.487	0.516
750	0.967	0.489	0.507	0.628
800	0.864	0.527	0.628	0.723
820	0.723	0.486	0.587	0.642
850	0.678	0.424	0.462	0.508

900	0.532	0.375	0.355	0.427
920	0.427	0.308	0.282	0.348
950	0.368	0.225	0.235	0.287
1000	0.216	0.178	0.198	0.208

Table T4: A measured value of Plasmon decay time (fs) of the collective dipolar Plasmon mode as a function of grating constant (nm) due to far-field dipole interactions

Grating constant (nm)	Plasmon decay time (fs)
350	5.22
370	6.27
400	7.17
420	8.23
450	9.25
500	9.78
520	6.24
550	4.62
600	3.35
620	4.56
650	5.78
700	6.36

Table T5: An evaluated result of dispersion of surface Plasmon-polaritons propagating at Ag/air, Ag/glass and Ag/Si interface

Wave vector	<------- Energy (eV)----------→		
K (10^6 cm^{-1})	Ag/Air	Ag/glass	Ag/Si
0	0.052	0.067	0.072
5	0.654	0.732	0.254
10	1.247	1.167	0.467
15	2.369	1.864	0.672
20	2.743	2.087	0.746
25	3.686	2.856	1.087
30	3.753	2.967	1.268
35	3.854	3.086	1.309
40	3.895	3.158	1.332
45	3.923	3.267	1.406
50	3.946	3.332	1.528

TableT6: An evaluated result of dispersion relation of coupled surface Plasmon modes on a thin Ag-film embedded in glass. S_b is odd bound mode and a_b is even bound mode.

Wave vector $(nm)^{-1}$	<-------Energy (eV)---------→					
	S_b			a_b		
	100nm	50nm	35nm	100nm	50nm	35nm
0.00	0.067	0.075	0.082	0.047	0.038	0.029

0.02	2,363	2.438	.453	1.867	1.764	1.708
0.04	2.425	2.543	2.554	2.325	2308	2.287
0.06	2.586	2.634	2.650	2.428	2.404	2.386
0.08	2.623	2.864	2.872	2.476	2.455	2.422
0.10	2.665	2.948	2.950	2.555	2.536	2.558
0.12	2.687	3.084	3.102	2.678	2.654	2.637
0.14	2.696	3.126	3.165	2.692	2.678	2.678
0.15	2.723	3.208	3.217	2.704	2.699	2.653
0.17	2.746	3.247	3.258	2.716	2.704	2.688
0.19	2.752	3.278	3.293	2.725	2.715	2.696
0.20	2.768	3.325	3.337	2.738	2.722	2.708

TableT7: An evaluated result of attenuation distance (propagation distance m) of odd modes S_b and even modes a_b as a function of wavelength (nm) for different thin Ag film embedded in glass.

Wavelength	**<------ Propagation distance (m)---------→**					
(nm)	Even a_b bound modes			Odd S_b bound modes		
	100nm	50nm	35nm	100nm	50nm	35nm
500	$2.6x10^{-5}$	$1.8x10^{-6}$	$2.6x10^{-7}$	$1.2x10^{-6}$	$2.2x10^{-5}$	$1.5x10^{-4}$
600	$3.8x10^{-5}$	$3.7x10^{-6}$	$4.5x10^{-7}$	$3.8x10^{-6}$	$4.6x10^{-5}$	$3.3x10^{-4}$
700	$4.7x10^{-5}$	$5.0x10^{-6}$	$6.2x10^{-7}$	$5.5x10^{-6}$	$6.7x10^{-5}$	$5.9x10^{-4}$
800	$8.8x10^{-5}$	$6.7x10^{-6}$	$8.4x10^{-7}$	$8.9x10^{-6}$	$9.5x10^{-5}$	$8.7x10^{-4}$
1000	$9.7x10^{-5}$	$8.8x10^{-6}$	$10.8x10^{-7}$	$13.2x10^{-6}$	$14.7x10^{-5}$	$12.6x10^{-4}$

1200	$11.8x10^{-5}$	$12.7x10^{-6}$	$14.2x10^{-7}$	$22.6x10^{-6}$	$23.8x10^{-5}$	$17.3x10^{-4}$
1500	$22.6x10^{-5}$	$18.6x10^{-6}$	$18.3x10^{-7}$	$32.8x10^{-6}$	$34.6x10^{-5}$	$27.6x10^{-4}$
1600	$30.7x10^{-5}$	$29.2x10^{-6}$	$24.6x10^{-7}$	$42.7x10^{-6}$	$45.8x10^{-5}$	$38.2x10^{-4}$
1700	$37.8x10^{-5}$	$32.7x10^{-6}$	$35.8x10^{-7}$	$58.5x10^{-6}$	$60.2x10^{-5}$	$45.9x10^{-4}$
1800	$48.6x10^{-5}$	$40.5x10^{-6}$	$45.5x10^{-7}$	$67.5x10^{-6}$	$69.9x10^{-5}$	$57.3x10^{-4}$
1900	$55.7x10^{-5}$	$52.9x10^{-6}$	$56.2x10^{-7}$	$75.6x10^{-6}$	$78.8x10^{-5}$	$65.5x10^{-4}$
2000	$67.8x10^{-5}$	$62.0x10^{-6}$	$67.8x10^{-7}$	$80.2x10^{-6}$	$82.6x10^{-5}$	$74.4x10^{-4}$

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