Optical Properties of Coated Nanospheres in Visible Wavelength Range

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Abstract: In this paper we have studied polymeric structures which possess coated spherical nanoparticles in visible wavelength range. Medium with metallic cores and silicon (Si) shells and structures composed by Si cores and metallic shells have been studied. Since, the size of particles is very small related to the incident wavelength, semi-static approximation and Clausius-Mossotti formula have been used in order to calculate the effective permittivity. Resonance wavelength of the structure depends on size and filling fraction of the guest nanoparticles. Resonance wavelength has been obtained by the mathematical relations and simulation results. Studied structures are applicable in invisibility.

1 INTRODUCTION

Mediums composed by core-shell nanoparticles in terms of application in photovoltaic cells, nanoantenna, optical switches and building blocks of metamaterial are important (Tang, Huo, Brittman, Gao, and Yang, 2011, Li, Engheta, 2007, Li, Salandrino, and Engheta, 2007, Rostami, Shahabadi, AfzaliKusha, and Rostami, 2012, Paniagua-Dominguez, Lopez-Tejeira, Marques, and Sanchez-Gil, 2011).

Also, sensors composed by coated spherical particles which possess the metal (Ag) cores and dielectric shells (SiO_2) or crystalline core-shell particles are of interest to the researches (Aslan, Wu, Lakowicz, and Geddes, 2007, Choi, Park, and Kim, 2009). Biosensors based on Localised Surface Plasmon Resonance (LSPR) constructed by core-shell nanoparticles have been studied in (Hao, Sonnefraud, Dorpe, Maier, Halas, and Nordlander, 2008, Endo, Kerman, Nagatani, Hiepa, Kim, Yonezawa, Nakano, and Tamiya, 2006) .

Moreover, medium with random distribution of core-shell nanoparticles in terms of simplicity in construction are particularly important. These polymeric mediums have electric resonances managed by filling fraction and size of particles, therefore these structures can be applicable in optical cloaking.

In this paper first we have studied a polymeric media with random distribution of spherical coated nanoparticles in visible wavelength range. First a polymeric medium possesses spherical nanoparticles with a metallic cores (Ag) and Si shells has been studied. Since metallic cores are surrounded only by Si, the effective medium has only one resonance wavelength in the visible range. For cylindrical cloaking excited by a transverse magnetic (TM) plane wave polarization state permittivity from 0 to 1 is required, so design of a cloak based on these investigated structures is possible in visible wavelength range (Cai, and Shalaev, 2010).

Polymeric medium which possesses core-shell nanoparticles with Si cores and metallic shells (Ag) has been studied in the next step. In this structure metals are surrounded by two medias (Si cores and polymeric host) therefore the structure has two resonance wavelengths. The resonances have been occurred in the visible and ultraviolet bands. Therefore as has been noted for first structure, cloak operated at two wavelengths in visible and ultraviolet bands can be designed by desired filling factor and size of particles. Resonance conditions of both effective medium have been obtained by mathematical relations and simulation results. It has been shown that the resonance condition strongly depends on the size and filling fraction of guest nanoparticles.

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Since the size of nanoparticals is very small related to the incident wavelength, semi-static approximation and Clausius-Mossotti formula in order to calculate the effective permittivity have been used.

2 THEORETICAL AND SIMULATION RESULTS

A polymeric media with random distribution of spherical nanoparticles containing metallic cores (Ag) with radius a_1 and Si shells with radius a_2 has been studied. Dielectric constants of silver cores and Si shells are ε_1 and ε_2 respectively. Effective permittivity of media has been obtained by Clausius-Mossotti formula as (Paniagua-Dominguez, Lopez-Tejeira, Marques, and Sanchez-Gil, 2011) :

$$\frac{\varepsilon_{eff} - \varepsilon_h}{\varepsilon_{eff} + 2\varepsilon_h} = f\frac{\alpha_E}{4\pi a_2^3} \qquad (1)$$

Where, α_E, f and ε_h are electric polarizability, filling fraction of the spherical nanoparticles and permittivity of host media respectively. A schematic of the structure has been shown in Figure 1:

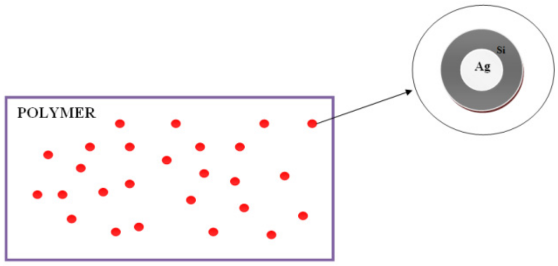


Figure 1: First structure has nanoparticles with metallic cores and Si shells.

Resonance wavelength of effective medium depends on size and filling fraction of the spherical particles as has been discussed in the following.

Since, the size of the particle is very small related to the incident wavelength, the electric polarizability of a core-shell can be expressed by semi-static approximation as equation (2) (Bohren, and Huffman, 1983):

$$\alpha = 4\pi a_2^3 \frac{(\varepsilon_2 - \varepsilon_h)(\varepsilon_1 + 2\varepsilon_2) + \tau(\varepsilon_1 - \varepsilon_2)(\varepsilon_h + 2\varepsilon_2)}{(\varepsilon_2 + 2\varepsilon_h)(\varepsilon_1 + 2\varepsilon_2) + \tau(2\varepsilon_2 - 2\varepsilon_h)(\varepsilon_1 - \varepsilon_2)} \qquad (2)$$

Where, $\tau = (a_1/a_2)^3$. The resonance condition for a particle can be obtained by setting the denominator of equation (2) to zero, but for the effective medium the resonance condition is different, because we are dealing with a set of particles. The effective permittivity can be expressed versus electric polarizability as:

$$\varepsilon_{eff} = \frac{(1 + \frac{2f\alpha}{4\pi a_2^3})\varepsilon_h}{(1 - \frac{f\alpha}{4\pi a_2^3})} \qquad (3)$$

By inserting (2) in (3) the effective permittivity can be discussed as:

$$\varepsilon_{eff} = \frac{(\varepsilon_2+2\varepsilon_h)(\varepsilon_1+2\varepsilon_2)+\tau(2\varepsilon_2-2\varepsilon_h)(\varepsilon_1-\varepsilon_2)+2f(\varepsilon_2-\varepsilon_h)(\varepsilon_1+2\varepsilon_2)+2f\tau(\varepsilon_1-\varepsilon_2)(\varepsilon_h+2\varepsilon_2)}{(\varepsilon_2+2\varepsilon_h)(\varepsilon_1+2\varepsilon_2)+\tau(2\varepsilon_2-2\varepsilon_h)(\varepsilon_1-\varepsilon_2)-f(\varepsilon_2-\varepsilon_h)(\varepsilon_1+2\varepsilon_2)-f\tau(\varepsilon_1-\varepsilon_2)(\varepsilon_h+2\varepsilon_2)}\varepsilon_h \qquad (4)$$

By setting the denominator of (4) to zero and solving the equation versus ε_1, resonance conditions will be expressed as:

$$\varepsilon_1^{res} = \frac{\varepsilon_2(2\varepsilon_2 + 4\varepsilon_h - 2\tau\varepsilon_2 + 2\tau\varepsilon_h - 2f\varepsilon_2 + 2f\varepsilon_h + f\tau\varepsilon_h + 2f\tau\varepsilon_2)}{-\varepsilon_2 - 2\varepsilon_h - 2\tau\varepsilon_2 + 2\tau\varepsilon_h + f\varepsilon_2 - f\varepsilon_h + f\tau\varepsilon_h + 2f\tau\varepsilon_2} \qquad (5)$$

By setting, dielectric constants of shells and host as ε_2=12.1 and ε_h=2.1904 equation (5) can be discussed by:

$$\varepsilon_1^{res} = \frac{12.1(32.9 - 19.8\tau - 19.8f + 26.3f\tau)}{-16.4 - 19.8\tau + 9.9f + 26.3f\tau} \qquad (6)$$

Dielectric constant of the metallic core has been assumed as Drude model (Cai, and Shalaev, 2010) :

$$\varepsilon_1(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega\gamma} \qquad (7)$$

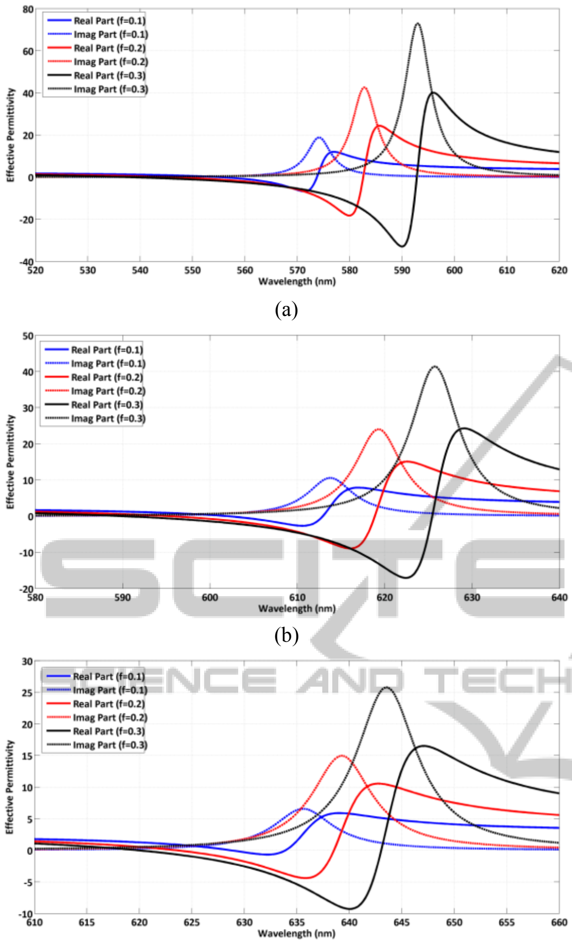
Where, ω_p=14*10^{15}and γ=0.032*10^{15} are the plasma frequency and collision frequency of silver respectively (Cai, and Shalaev, 2010). Therefore the resonance frequency is obtained as equation (8):

$$1 - \frac{(14*10^{15})^2}{\omega_{res}^2 + i*0.032*10^{15}*\omega_{res}} - \frac{12.1(32.9 - 19.8\tau - 19.8f + 26.3f\tau)}{-16.4 - 19.8\tau + 9.9f + 26.3f\tau} = 0 \qquad (8)$$

The roots of the above equation are complex; the frequency which has the positive real part is the desired answer. Real part of this value is resonant frequency of the effective media.

The roots depend on τ and f. The effective permittivity of the polymeric medium by fixing the inner radius and changing the outer radius of the guest particles has been shown in Figure 2.

As can be seen in Figure 2 the real part of the effective permittivity is negative in visible wavelength range, so these structures can be useful in construction of building blocks for metamaterials. The wavelength band with negative permittivity becomes wider by



(a)

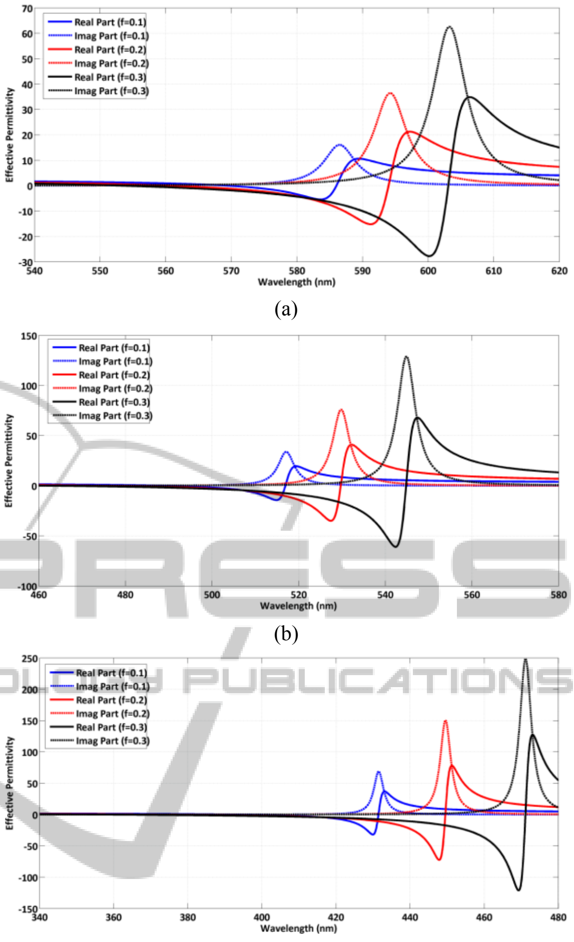
(b)

(c)

Figure 2: Real part of effective permittivity with different fill factors in visible region for (a) R_{in}=15nm, R=25nm, (b) Rin=15nm, R=30nm, (c) R_{in}=15nm, R=35nm, the host medium is polymeric with refractive index of 1.48.

increasing the filling fraction of nanoparticles, because the number of the guest particles increases. The particle can be assumed as a cavity, so by increasing the outer radius of the cavity resonance wavelength shifts to higher values. The effective permittivity by fixing the outer radial and changing the inner radial of the particles has been shown in Figure 3.

By increasing the inner radius of the particles the resonance wavelength is shifted to the lower wavelength values. Because by increasing the inner radial of the particles and decreasing the thickness of the shells, the particles can be assumed in the polymeric medium without any shells. Therefore the resonance conditions for the particles ($\varepsilon_{\text{particles}}=-2\varepsilon_h$) shifts to lower wavelength.



(a)

(b)

(c)

Figure 3: Real part of effective permittivity with different fill factors in visible region for (a) R_{in}=20nm, R=35nm, (b) R_{in}=25nm, R=35nm, (c) R_{in}=30nm, R=35nm, the host medium is polymeric with refractive index of 1.48.

As has been shown in Figure 2 and 3 the resonance wavelength depends on the filling fraction. The resonance wavelength which has been obtained by mathematical relations and diagrams has been given in the Table 1.

Table 1: Resonance wavelength obtained by mathematical relations and diagrams for different size and filling fraction of guest particles.

f	τ	λ(nm)by (8)	λ(nm)by Diagrams
0.1	$(15/25)^3$	574.7	574.1
0.2	$(15/30)^3$	620	619.3
0.3	$(15/35)^3$	644.5	643.5
0.3	$(20/35)^3$	603.9	603.3
0.2	(25/35)3	530.1	529.9
0.1	(30/35)3	431.4	431.6

Also a polymeric medium with random distribution of spherical nanoparticles compose by Si cores with radius a_1 and metallic shells (Ag) with radius a_2 has been studied. The structure which has been considered has been shown in Figure 4:

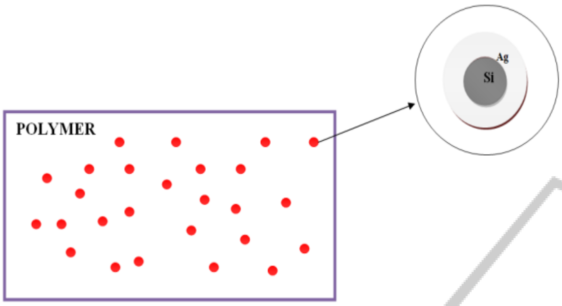
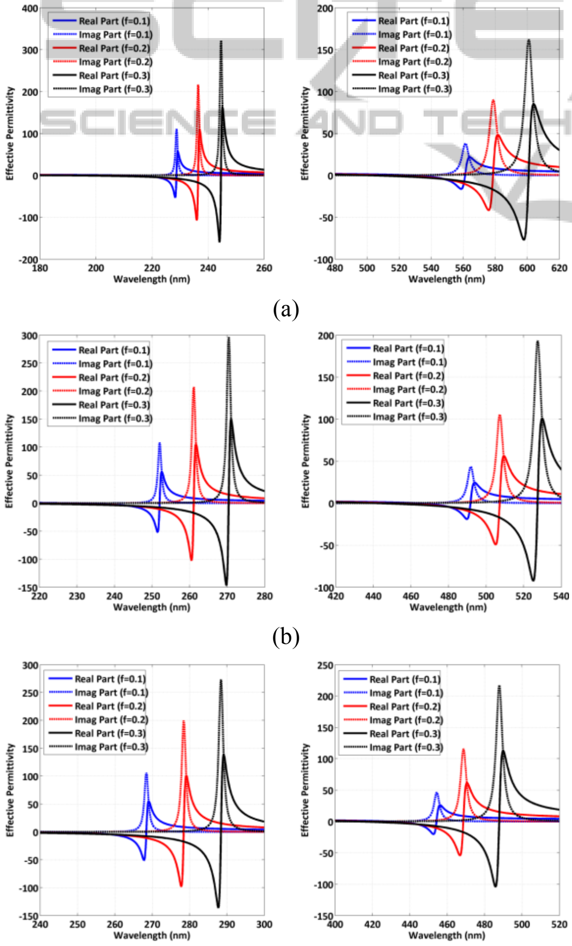


Figure 4: nanoparticles with Si core and metallic shell.



(a)

(b)

(c)

Figure 5: Real part of effective permittivity with different fill factors in visible region for (a) Rin=15nm, R=25nm, (b) R_{in}=15nm, R=30nm, (c) R_{in}=15nm, R=35nm, the host medium is polymeric with refractive index of 1.48.

By similar analysis which have been accomplished for the first polymeric media, the resonance wavelength can be obtained as the following.

The resonance conditions is achieved by setting the denominator of (4) to zero and solving it for ε_2.

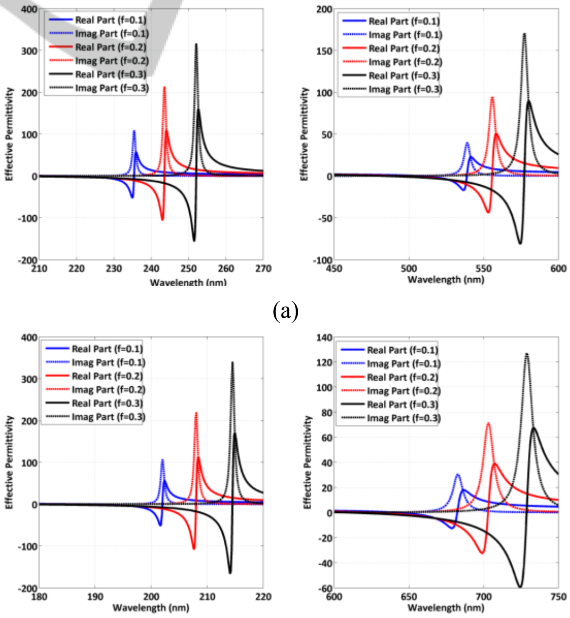
As has been obtained in (9), the equation is a binominal equation versus ε_2. In this polymeric media there are two resonance wavelengths.

One of the wavelengths is in ultraviolet wavelength range and the other one is in visible band. Two resonances are due to existence of the silvers surrounded by two mediums, shells and host polymeric media. Each mediums causes to resonance separately. The resonance in visible range is due to the Si cores and another one in ultraviolet band is due to the polymeric host media.

$$\begin{aligned}&(\varepsilon_2+2\varepsilon_h)(\varepsilon_1+2\varepsilon_2)+\tau(2\varepsilon_2-2\varepsilon_h)(\varepsilon_1-\varepsilon_2)\\&-f(\varepsilon_2-\varepsilon_h)(\varepsilon_1+2\varepsilon_2)-f\tau(\varepsilon_1-\varepsilon_2)(\varepsilon_h+2\varepsilon_2)=0\end{aligned} \qquad (9)$$

The effective permittivity of the polymeric medium by fixing the inner radius and changing the outer radius of the guest particles has been shown in Figure 5.

Also effective permittivity by fixing the outer radius and changing the inner radius has been indicated in Figure 6.



(a)

(b)

Figure 6: Real part of effective permittivity with different fill factors in visible region for (a) R_{in}=20nm, R=35nm (b) R_{in}=25nm, R=35nm, the host is polymeric medium with refractive index of 1.48.

The resonance which has been obtained by mathematical relations and diagrams has been given in the Table 2 and Table 3 for the visible and ultraviolet band respectively. Since the effective medium is studied, the resonance wavelength depends on the filling fraction as has been discussed previously. This polymeric structure can be useful in construction of building blocks for metamaterials in two wavelength bands.

Table 2: Resonance wavelength in the visible range has been obtained by mathematical relations and diagrams for different size and filling fraction of guest particles.

f	τ	λ by Equation (nm)	λ by Diagrams (nm)
0.1	$(15/25)^3$	561.39	561.4
0.2	$(15/30)^3$	507.26	507.4
0.3	$(15/35)^3$	487.86	487.9
0.3	$(20/35)^3$	577.22	577.2
0.2	$(25/35)^3$	703.12	703.2

Table 3: Resonance wavelength in the ultraviolet range has been obtained by mathematical relations and diagrams for different size and filling fraction of guest particles.

f	τ	$\lambda$by Equation (nm)	$\lambda$by Diagrams (nm)
0.1	$(15/25)^3$	228.76	228.8
0.2	$(15/30)^3$	261.13	261.1
0.3	$(15/35)^3$	288.43	288.4
0.3	$(20/35)^3$	252.06	252.1
0.2	$(25/35)^3$	208.01	208

As has been observed in Table 3 in visible wavelength range, by increasing the thickness of the metal the resonance wavelength shifts to lower amounts of wavelength. This effect can be related to the plasmon resonance energy. Since the electric charge increases the plasmon resonance energy is greater, therefore the resonance wavelength shifts to the smaller amounts of the wavelength.

In the ultraviolet band according to the Table 4, the noted argument can be used for fixed outer radius too. In ultraviolet wavelength range by increasing the size of particles the resonance wavelength shifts to the higher amounts of wavelength. Similar to the argument which has been expressed for the first structure which the particles can be considered as a cavity.

4 CONCLUSIONS

We studied two polymeric structures with random distribution of core-shell nanoparticles. In the first structure the coated spheres with metallic cores (Ag) and Si shells was studied. Resonance wavelength was obtained by mathematical relations and simulation results. In this structure, the medium possess only one resonance wavelength because the metallic particles are surrounded by one shell (Si). The similar analysis were accomplished in a polymeric media with Si cores and metallic shells. Since the metals are surrounded by two medias (Si cores and polymeric host) effective medium has two resonance wavelengths. The resonance conditions were studied by mathematical relations and simulation results. It was shown that the resonance wavelength in both structures depends on size and filling fraction of spherical nanoparticles. In order to calculate the effective permittivity semi-static approximation and Clausius-Mossotti formula used. Both studied structures are applicable in optical cloaking with metamaterials.

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