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Enhancement of Light Absorption Using Nanoparticles Embedded Double Layer Anti-Reflection Coating

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Abstract. The formation of nanoparticles embedded composite materials is a means for engineering the electrical and optical properties of thin films. This paper proposes the use of gold and silver nanoparticles (NPs) to fine tune the effective index of two dielectric layers towards anti-reflection coating applications. Here, the upper layer is magnesium fluoride (MgF_2) and the lower layer is Polymethyl methacrylate (PMMA). The effect of embedding the nanoparticles in the PMMA layer on the transmission spectrum is studied while tuning the concentration (volume fraction) and size of particles. With the increase in the size of the nanoparticles there is a red shift in the transmission dip. On varying the nanoparticle size the peak absorption can be shifted to the desired near ultra-violet radiation region, hence prevent transmission through the thin film arrangement. Considering 30 nm silver nanoparticle with the volume fraction ranging from 0.001 to 0.05, maximum transmission of light is observed under lower wavelength range of visible spectrum across the novel structure

Keywords: Anti-reflection coating, T-matrix, effective medium theory, nanoparticles, SERS.

1. Introduction

In order to increase the transmission of light through a subject, anti-reflection coating (ARCs) are widely used in diverse range of industrial and ophthalmic application [1-4]. The development of efficient low-cost ARCs is an active area of scientific research as a marginal change in the efficiency will be reflected on cost sensitive industries such as photovoltaic. In the recent years, there has been improvement in the efficiency of solar cells (SCs). This enhancement in the power output is mainly confined to the material composition and structural arrangement of the photo voltaic cell [5-6], however the overall performance has not reached its peak potential as 30% of the incident light on the solar panel is reflected back to the atmosphere [7-8]. There are various antireflection techniques to augment the percentage of transmitted light. Sub-wavelength structures [9-10], plasmonic surface [11], surface passivation, single quarter wavelength and multilayer coating are such techniques which suppress the reflection by enhancing the transmission or absorption. In this paper we are trying to inculcate the quarter wavelength property, multilayer coating (precisely double layer coating) and plasmonic properties of metal nanoparticles to the advantage of high performing antireflection coating under visible light spectrum. In order to obtain the thin film effect, the thickness of the layer has to be a quarter or half the wavelength of the light which results in suppression of reflection. On considering reflection minima under wide range of visible light spectrum (350nm-800nm) the use of multiple layer AR coating materials can improve performance [12]. The design of such multilayer coating is a complex process, henceforth lesser number of coating layers should be considered to reduce the fabrication complexity. A two-layer coating reduces the reflection of usable sunlight to 4%. Film materials with specific refractive index are desired to obtain efficient anti-reflection properties which are rare in nature. Therefore, an effective medium has to be engineered for the required dielectric constant. In this work, we simulated and investigated the proposed structure, which consist of metal NPs in one of the layers of thin film structures. Here we have considered gold and silver spherical nanoparticles of specific size (5nm) in-order to avail the property of localize surface Plasmon resonance properties under the wavelength of 380nm-420nm. This helps in the absorption of ultraviolet radiation (UV-A) and near blue light and hence prevent its transmission through the proposed structure. Based on the simulation results it has been observed that the proposed structure prevents the transmission of harmful ultraviolet radiation while simultaneously exhibiting ultralow reflectivity within the visible light spectrum.

In the next section, a new composite material with a specific effective medium is desired to obtain the quarter wavelength property for antireflection. In the effective medium the host is considered to a dielectric (PMMA) where nanoparticles are embedded to regulate the overall property of the material. In the third section the property of the metal nanoparticles will be analysed and the overall effect on the composite thin film is observed. The refractive index of the effective medium with the adherent plasmonic property of the metal nanoparticles is then utilized in the fourth section. In the fourth section, the scattering matrix is considered as computation electromagnetic method for deriving the intensity of transmitted and absorbed light through the composite material. Following this section thin films of two layers will be arrange in a manner to attain antireflection property for broadband transmission maxima along the range of visible light spectrum.

2. Effective Medium

The performance of an optical coating is dependent upon the number of layers, the thickness of the individual layers, and the refractive index of the individual layers and difference at the layer interfaces. However, the main difficulty is finding material with desired refractive indices for the design of interest. Hence, this work proposes embedding Ag/Au nanoparticles in the lower layer for effective index tuning. Maxwell Garnett mixing formula is preferred as it gives the permittivity of the effective medium in terms of the permittivities and volume fraction of the individual constituent of the complex medium. However, if a composite contains particles whose dimensions are beyond the range of the Rayleigh size limit, then the Maxwell Garnett mixing formula does not support the actual effect of the inclusion in the host medium. Maxwell-Garnett [13] formulation is only applicable if the size of the inclusion is diminutive as compared to the wavelength of the light source and hence does not address the particles size beyond the range of Rayleigh size. Dynamic Maxwell-Garnett theory is valid if the medium contains the inclusion whose dimension is within the range of Rayleigh size. In order to analyze the size effect in the Maxwell-Garnett theory the screening parameter has to be considered. Particle shape determines the screening parameter k, which is observe as number '2' in the Maxwell-Garnett equation:

$$\left(\frac{\varepsilon_{MG} - \varepsilon_h}{\varepsilon_{MG} + 2\varepsilon_h}\right) = f_m \left(\frac{\varepsilon_i - \varepsilon_h}{\varepsilon_i + 2\varepsilon_h}\right) \tag{1}$$

However, the size effect is considered within the screening parameter, k which seems to have just the effect of the shape.

And, hence, the inclusion shape and size will be evident in the composite optical properties [14] where ε_{MG} the effective permittivity of the composite material is. In 1983, Meier and Wokaun [15], in an effort to predict metal particle size effects in surface enhance Raman scattering (SERS), derived an a expression for effective

depolarization factor q_{eff}, to understand the size effect which is related to nano-scale sphere radius "a" via

$$q_{eff} = \frac{1}{3} - \frac{1}{3}k^2a^2 - \frac{2}{9}ik^3a^3 \qquad (2)$$

where the first term is the Lorentz depolarizing factor (q = l/3) for spheres. The second term involves dynamic depolarization and accounts for particle size which experience different phases of the incident field as size increases. The third term accounts for damping of the induced dipole due to radiation emission. And $k = 2\pi n_h / \lambda$, n_h is the refractive index of the host material.

An effective screening factor k_{eff} can be calculated using the following equation:

$$k_{eff} = (1/q_{eff}) - 1 \qquad (3)$$

Using the approach of replacing 2 over k_{eff} in the Maxwell Garnett equation, leads to the following equation:

$$\left(\frac{\varepsilon_{MG} - \varepsilon_h}{\varepsilon_{MG} + k_{eff}\varepsilon_h} \right) = f_m \left(\frac{\varepsilon_i - \varepsilon_h}{\varepsilon_i + k_{eff}\varepsilon_h} \right) \qquad (4)$$

With the obtained effective permittivity of the desired medium, the corresponding refractive index acquired using the relation:

$$\varepsilon_{MG} = \varepsilon' + i\varepsilon'' = (n + ik)^2 \qquad (5)$$

where, ε' and ε'' are the real and imaginary part of the complex permittivity; n and k are the real and imaginary part of the refractive index. Using the complex refractive index in the T-Matrix, the reflectivity and transmissivity of the thin film anti-reflection coating is obtained.

3. Metal Nanoparticles

On considering the two layers of thin film acting as an antireflection coating it is possible to obtain the high transmission. Metal nanoparticles are considered as an inclusion in one of the layer in the thin film to obtain the desired refractive index. However, there is the property of surface plasmons in the metal nanoparticles which can be harnessed to absorb the specific band of spectrum and prevent the transmission.

These surface plasmons trap the energy by the means of strong enhancement of the local electromagnetic field compared to the exciting electromagnetic field. Surface plasmons exist for metals like Au, Ag, Al, and Cu, depending on the dielectric function of both, the metal and the neighbouring dielectric. Metal nanoparticles behaviour under plasmon condition is defined by the electron's effective mass and charge, the electron density, size and the geometry of the particle. Here, we have considered the effect of the size of spherical particles.

Its has been studied[16] that with the increase in the size of the spherical nano-particles there is an absorption of the light under higher wavelength. Its desired in our proposed condition to absorb the wavelength near UV light by regulating the size of the metal nanoparticles. This would prevent the transmission of UV-B (290-315 nm) or UV-A (315-380 nm) which causes ocular health effects. From the previous studies[17] it is conformed that Ag, Au, and Cu nanoparticles exhibit intense surface-enhanced Raman scattering (SERS) spectra in the visible region of the spectrum, with intensities on Au and Cu reduces at a faster rate than on Ag as frequency is increased. Here, Ag is considered in our simulation as its gives a sharp dip in the transmission light under a specified wavelength under near UV. The slab of layers in Fig. 1 gives a schematic diagram of the arrangement of two thin layers where the size of the nanoparticles in the lower layer is varied and the response of the transmissivity is observed.
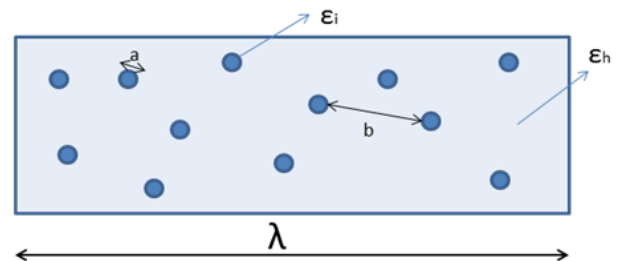

Fig. 1. Schematic of effective medium with metal nanoparticles as inclution of premitivity ε_i having radius 'a' with average spacing of distance 'b' embedded in the host medium of premitivity ε_h . b $<<$ λ.

4. Light Interference in Effective Medium

T-matrix approach is considered to obtain the reflectance and transmittance of the layer arrangement which relates the electric and magnetic field at both interfaces of the two-layer thin film via a characteristic matrix [18]. In this paper semi-analytical method is applied as there are multiple layers of thin film which are uniform along the longitudinal direction. Semi-analytical method is adapted to simulate this novel structure as it is highly efficient when the analytical direction is large and cumbersome. It has established itself as a potent tool in electro-magnetic computation and provides highly structured analysis of layered structure. An overall scattering matrix is obtained by using the Redheffer star product [19] on combining scattering matrices calculated for each layers that depicts the propagation along the longitudinal direction through the entire device. As

shown in Fig. 3, a double layer anti-reflection coating over a silicon substrate is considered where the ray of light has to get through three boundary conditions in order to reach the silicon substrate.

The parameters in the scattering matrix actually quantify the physical interpretation of the light behaviour in the device. The S_{11} parameter depicts the quantity of light reflected from the concern layer with L1 representing as the top layer of the antireflection coating and L2 representing as the bottom layer of the antireflection coating. S_{21} depicts the quantity of light transmitted through the device. For symmetric devices, we can consider, $S_{11} = S_{22}$ and $S_{21} = S_{12}$.

Considering the scattering matrix of the top layer S(T) that is to be followed by the scattering matrix of bottom layer S(B), the global scattering matrix can be calculated using the Redheffer Star Product $S(G) = S(T) \otimes S(B)$ as follows:

$$\begin{bmatrix} S_{11}^G & S_{12}^G \\ S_{21}^G & S_{22}^G \end{bmatrix} = \begin{bmatrix} S_{11}^{L1} & S_{12}^{L1} \\ S_{21}^{L1} & S_{22}^{L1} \end{bmatrix} \otimes \begin{bmatrix} S_{11}^{L2} & S_{12}^{L2} \\ S_{21}^{L2} & S_{22}^{L2} \end{bmatrix} \quad (6)$$

The global scattering matrix acts as a connecting matrix between the mode coefficient of the reflected (c_{ref}) and transmitted wave (c_{trn}) to the mode coefficient of the incident wave (c_{inc}) as shown:

$$\begin{bmatrix} c_{ref} \\ c_{trn} \end{bmatrix} = \begin{bmatrix} S_{11}^G & S_{12}^G \\ S_{21}^G & S_{22}^G \end{bmatrix} \begin{bmatrix} c_{inc} \\ 0 \end{bmatrix} \quad (7)$$

On relating the mode coefficient with the fields, reflected and transmitted wave is attained for the double layer coating. The simulation of effective medium for a desired refractive index for antireflection property along with the property of the metal nanoparticles has been utilize in the scattering matrix computational technique to obtain the intensity of the transmitted light. However, low reflectivity is required under a broader spectrum of visible light wave. In the present section, the thickness of the two layers has not been discussed which will lead to the property of quarter wavelength for the antireflection. In the next section, various techniques to achieve broadband antireflection properties using thickness regulation will be discussed.
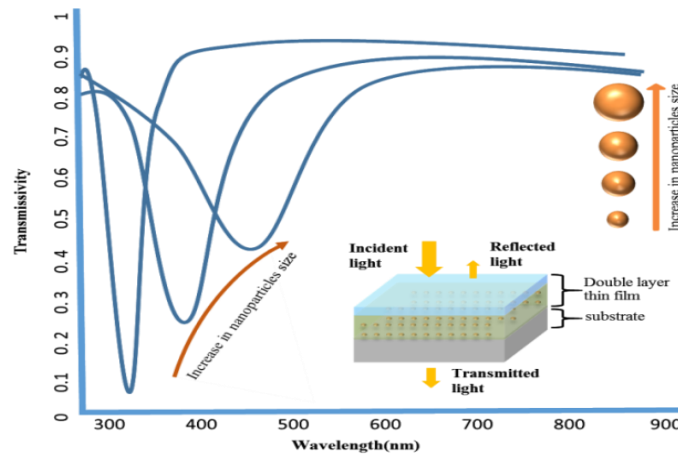

Fig. 2. Transmission response of the thin film arrangement on the variation of the size of nano-particles located on the lower layer of the thin film.
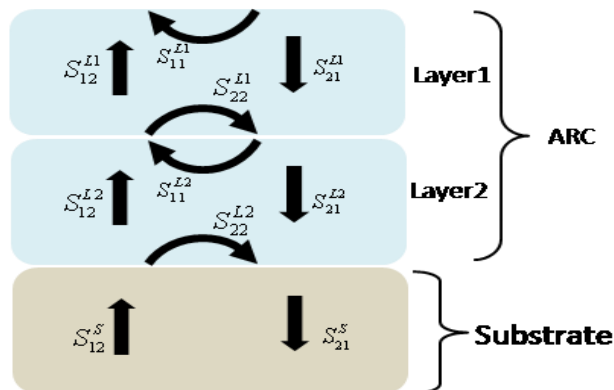

Fig. 3. Combination of conventional scattering parameters matrices of two layers acting as ARC over a substrate.

5. Antireflection Coating

In order to improve the performance of the thin layer as an anti-reflection coating several layers of films are stacked together to perform as multilayer antireflection coatings. However, the problem of controlling the thickness of the multiple coating in production must be taken in consideration. Hence it is advantageous to consider lesser number of layers under fabrication point of view realising adequate anti-reflection property. On varying the thickness of the antireflection coating subjected to a specific wavelength a change in the intensity of the transmitted light through it can be realised. In general, V-coats are the type of thin layer coating designed to gain the property of AR. However, this type of coating is designed to increase the transmission over a very narrow waveband with reference to a specific wavelength where maximum peak transmission will be observed. Narrow band reflection reduction is accomplished by V-coating for double layer antireflection coating where two layers of quarter wavelength thickness layered over a substrate. Broadening of reflection reduction is observed by W-coating [20]. In order to optimize the double layer coating using w-type coating a half wave film is layered between a quarter wave single layer antireflection coating and the substrate [21] as shown in Fig. 4.

In order to analyse the effect of the mentioned two type of coating a double layer coating is considered with 510nm as the specified design wavelength. In Fig. 5(a) the substrate is considered to have refractive index of 1.5. The refractive index of the top layer is considered to be 1.38(n1) as per MgF$_2$ and the bottom layer as 1.7(n2) as per PMMA under the specified wavelength of 510nm. For the V-type coating the thickness of top layer is 92.4nm ($d1 = 510/(4 \times n1)$) and the bottom layer is 75nm ($d2 = 510/(2 \times n2)$). Considering the same condition with just a change in the thickness of the bottom layer to 150nm ($d2 = 510/(4 \times n2)$) a change in the reflectance is observed as shown in Fig. 5(a) which is termed as W-type coating. Similarly, another condition is considered (under Fig. 5(b)) where the refractive index of the substrate is 1.6 and the refractive index of the bottom layer is 1.8. Under both the condition the W-type coating gives reflectance minimum under a broad spectrum.

As for our application, minimum reflectance is required for a broader spectrum of light ranging from 300nm to 800nm. Based on the simulation results it has been observed that W-type coating yields a broader reflectance minimum as compared to the V-type coating.

Hence W-type coating design is considered over V-type coating.

6. Results and Discussion

On considering the double layer antireflection coating with upper layer MgF$_2$ (92 nm thickness) and lower layer PMMA (184 nm thickness) as host with metal nanoparticles inclusion over the silicon substrate as shown in Fig. 6, it has been observe that as the concentration and the size of the metal nanoparticles changes the transmitted light gets affected through the proposed structure.

As the property of quarter wavelength has to be utilized for attaining the antireflection properties, thickness can be considered within the range of 100nm to 200nm as the source wavelength is of the range of visible light spectrum (consider mid range of 500 nm). Acknowledging the permittivity of the thin film, we have considered the thickness of the top layer as 92nm and the lower layer is double the thickness (184 nm). The thickness and the refractive index of the thin film layers has to be tuned in order to attain the quarter wavelength properties which in this case has given a satisfactory results in terms of antireflection property.

The effective extinction cross section of a silver nanoparticle can be very large as compared to the physical cross section. As compared to the gold nanoparticles the effective extinction cross section of silver nano-particle is higher. The scattering loss under the near UV and blue light range by a spherical silver nano-particle is due to SPR. The SPR peak wavelength can be tuned from 380nm to 600nm by varying the size of nanoparticles ranging from 5nm to 70nm. This property can be utilized to block the near UV light on considering the size less than 5nm. Moreover, the host (PMMA) which acts as a medium within which the nanoparticles resides plays a vital role in determining the shift of SPR peak wavelength. From Fig. 7, as the size of the nanoparticles increases there is a red shift in the resonance peak of the transmitted light intensity along its wavelength. The sharp and tapered dip of the transmitted light intensity of the silver nanoparticles under the size of 5nm is considered to assist in blocking the light under near UV light and the double layer arrangement aids in high transmission for the remaining spectrum of the visible light. Moreover, based on Fig. 7(a) and (b) it is evident that there is a gradual dip in the intensity of the transmitted light along the wavelength with the increase in size of Silver nanoparticles which is not featured in Gold nanoparticles.
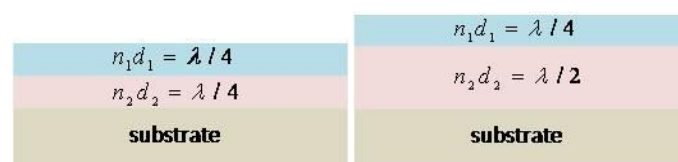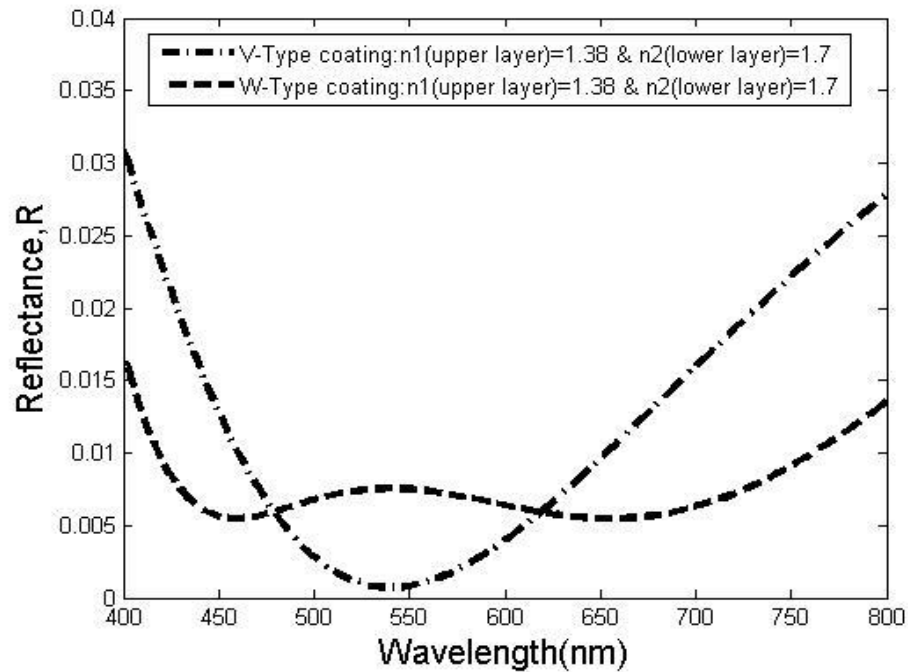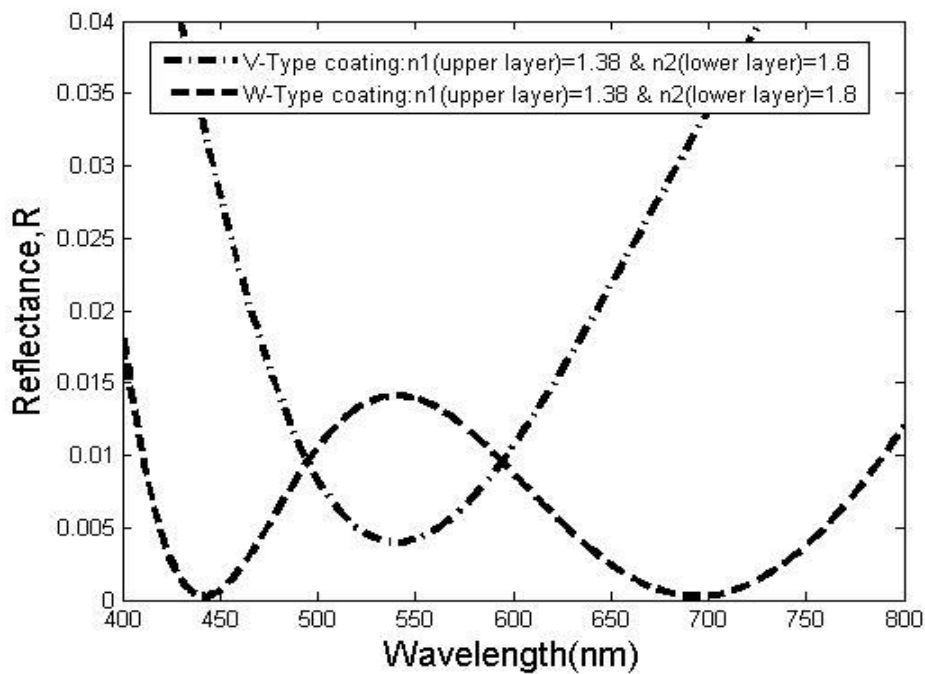

Fig. 4. Condition for double layer w-type antireflection coating.

(a)

(b)

Fig. 5. Comparison of reflectance between the V-types and W-type two-layer coating on a substrate with (a) substrate index (ns)=1.5 and medium index(no)=1.0(air); 4n1d1=510nm (top layer); 4n2d2= 1020 nm (bottom layer under W-type coating) and 4n2d2= 510nm (bottom layer under V-type coating); (b) substrate index (ns)=1.6 and medium index(no)=1.0(air); 4n1d1=510nm (top layer); 4n2d2= 1020 nm (bottom layer under W-type coating) and 4n2d2= 510nm (bottom layer under V-type coating).
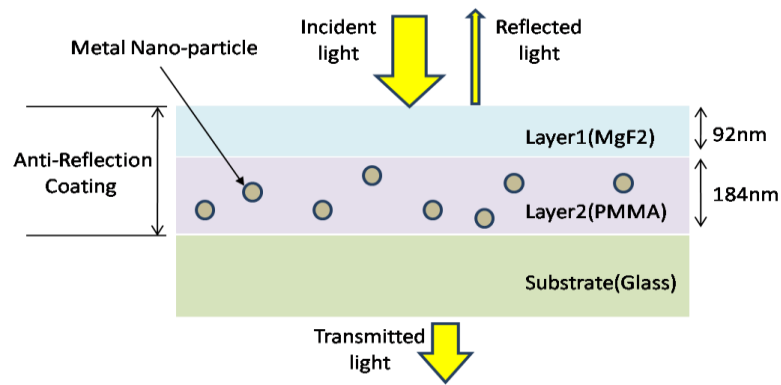
Fig. 6.   Schematic diagram of the proposed structure.
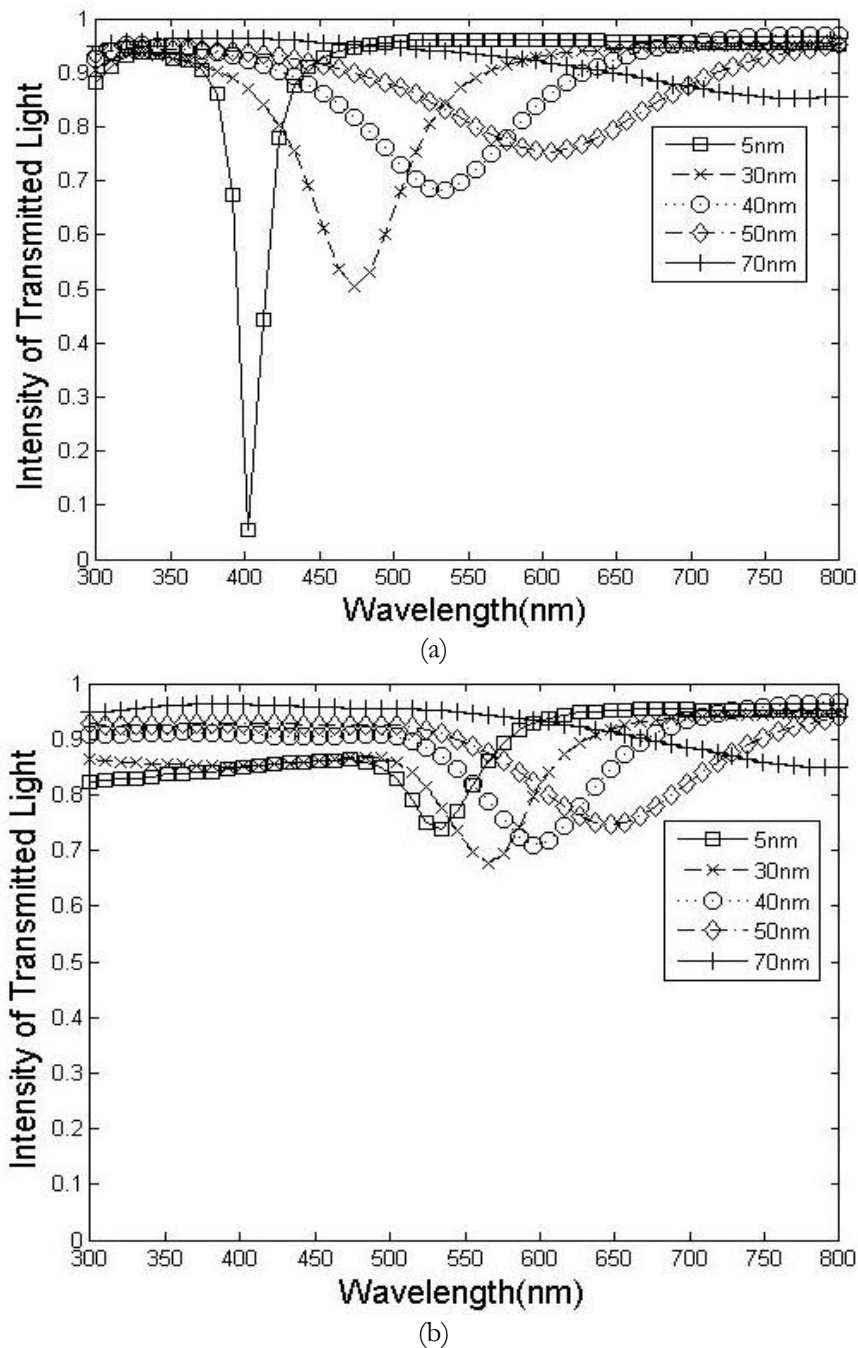

(a)

(b)

Fig. 7.   Size variation effect of nanoparticles over the transmitted light through the antireflection coating; (a) inclusion: Silver; (b) inclusion: Gold; Host: PMMA; Volume Fraction :0.01.
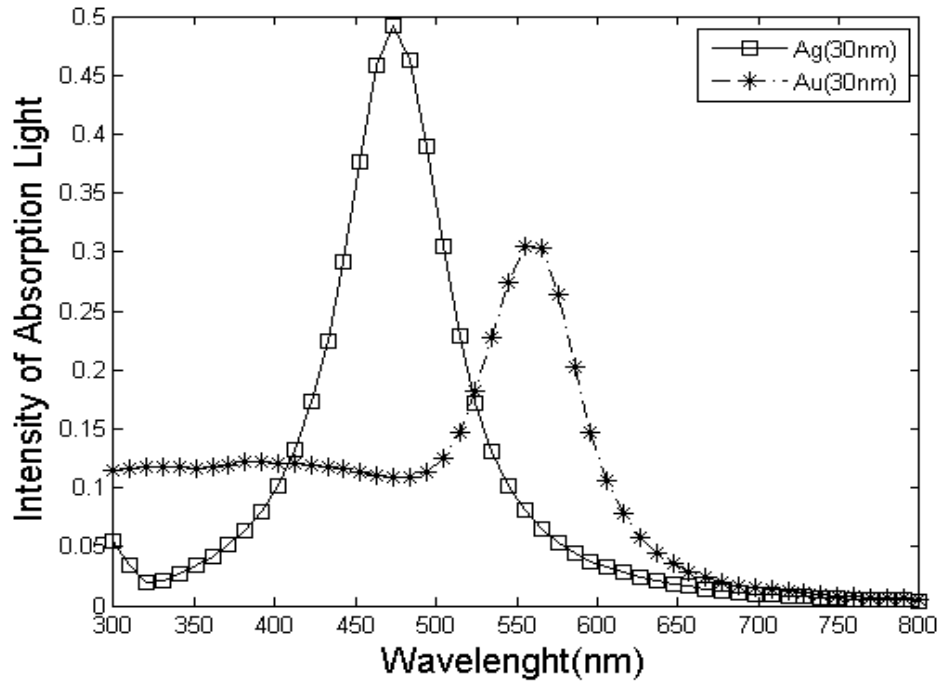
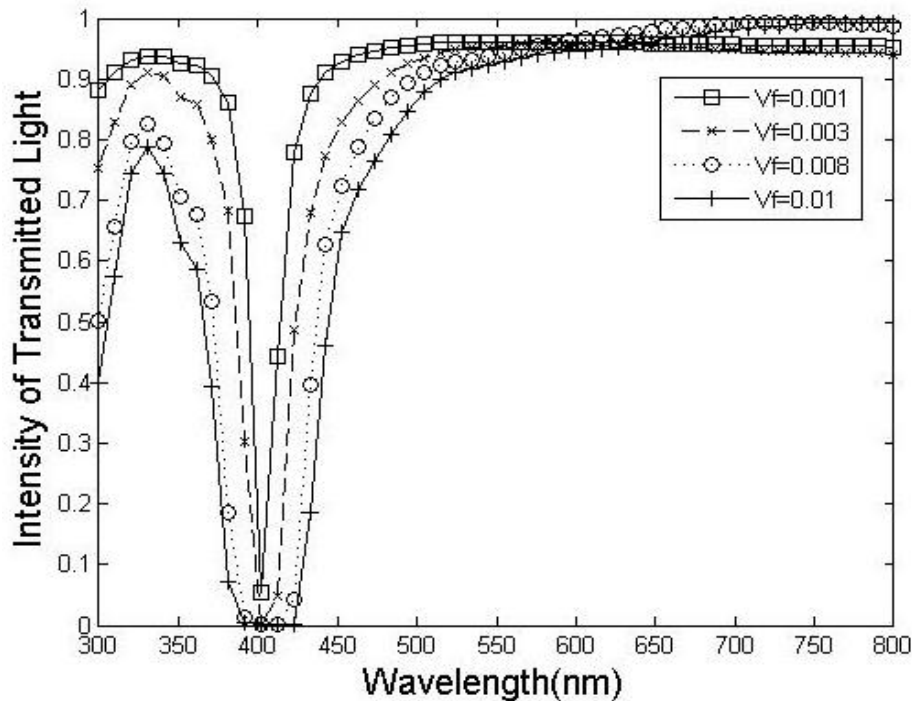Fig. 8. Change in the absorption peak intensity with the types of metal (Gold and Silver) nanoparticles.

Fig. 9. Effect in intensity of the Reflected light with varying silver nanoparticles embedded in PMMA under MgF$_2$ layer

The gold nanoparticle of 30nm size has lower peak absorption as compared to that of silver nanoparticles of the same size as seen from Fig. 8. Furthermore, the peak absorption is carried out at different frequency for the two metal nanoparticles (near 470nm for silver and near 560nm for gold). It has been observed that with different metal nanoparticles the response to light interference varies. These variations are subjected to the extinction cross section which represents the loss of energy from the incident light. In the Rayleigh regime, the total scattering cross section and absorption cross section are defined as [16]

$$\sigma_{ext} = \sigma_{sca} + \sigma_{abs} \qquad (8)$$

$$\sigma_{ext} = \frac{2\lambda^2}{3\pi}\alpha^6 \left|\frac{m^2-1}{m^2+2}\right|^2 + \frac{-\lambda^2}{\pi}\alpha^3 \, \text{Im}\left\{\frac{m^2-1}{m^2+2}\right\} \qquad (9)$$

The extinction cross section is a function of the particles size, 'a' (30nm in radius), the property of the material, 'm'(refractive index) and the property of the material, 'm_0'(refractive index of the host). Here, $\alpha = \frac{2\pi a}{\lambda}$, a dimensionless unit parameter and $\lambda = \frac{\lambda_0}{m_0}$ where λ_0 is the incident wavelength in vacuum, and m_0 represents the refractive index of the host medium(PMMA). As the material parameter (inclusion and host medium) is considered within the expression of extinction cross section the absorption behaviour of the gold and silver particles under a specific wavelength differs. For a dielectric particle the imaginary part of the refractive index (k) is zero, resulting in the $\sigma_{ext} = \sigma_{sca}$ as the contribution of the absorption cross-section is zero. Therefore metal nanoparticles are considered for the absorption of the specific light intensity. It is observed that in silver nanoparticles the absorption of light is carried out at lower wavelength. This would support the blocking of the light of higher frequency and prevent the light to transmit through the layers of thin film. The particle sizes cannot be too large as this leads to multipole oscillations which tend to decrease the scattering efficiency of the nanoparticles [22].

With the change in the volume fraction of the metal nanoparticles inclusion in the lower layer of the antireflection coating there is a change in the width of the transmission dip. As shown in Fig. 9, with the increase in the volume fraction of the gold nanoparticles (5nm in radius) the dip in the intensity of the transmitted light is broadened. Hence, tuning the volume fraction of nanoparticles can regulate the range of wavelength (here, 350nm-450nm for the size of 5nm) that can be restrain through the proposed coating. The broadening of the transmission dip is due to the increase in the concentration of the metal nanoparticles which leads to the absorption of light in the larger range of wavelength. The imaginary part (k_{eff}) of the effective refractive index gradually increases with the increase in the volume fraction of the inclusion under a specific wavelength resulting in the increase in the absorption.

On analysing Fig. 10 it has been observed that the transmitted light intensity is higher at the lower wavelength (ranging from approximately 350nm-500nm) when the volume fraction is as low as 0.001 to 0.05, whereas there is a decrease in the intensity as the volume fraction increase under higher wavelength. Hence, there is a depressing effect on the intensity of the transmitted light with the increase in the concentration of the metal nanoparticles, which can be compromised with the effect of light absorption near ultra violet light.

7. Conclusion

As per the above discussion based on the simulation results of the proposed structure it is apparent that the size of the nanoparticles within the range of 5nm prevents the transmission of light under near UV light. Moreover, the double layers thin film arrangement over the substrate acts as an antireflection coating over a wide range of the visible light spectra ranging from 450nm to 800nm. Moreover, here exists an effect of the concentration of the nanoparticles in the form of volume fraction in the effective medium. The increased in the concentration of the nanoparticles leads to the broadening of the transmitted light intensity dip. Hence, apart from the regulation of the refractive index of the effective medium there can be a possibility of tuning the blocking range of the wavelength through the substrate. On regulating the volume fraction of the effective medium and size of nanoparticles we can fine tune the optical property leading to an effective anti-reflection coating. In order to have maximum transmission under the prescribed condition the volume fraction of the effective medium should be in the range of 0.001-0.003 and size of the silver nanoparticles should be as less than 30nm. Moreover, here is a possibility of blue shift of the SPR peak wavelength towards the infrared region of electromagnetic spectrum by considering the change in shape (prolate spheroid) of the silver nanoparticles. Finally the entire disposition of the system makes it an antireflection coating over the range of visible light spectrum with near ultra-violet (UV-A) blocking property.
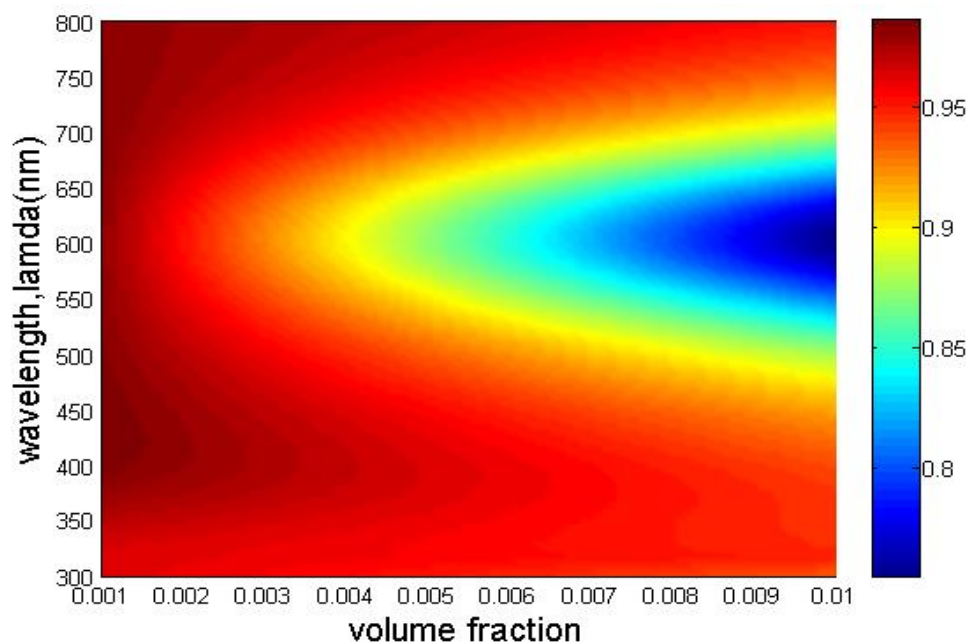
Acknowledgements

Fig.10. Effect of the transmitted light intensity with the change in the volume fraction of the inclusion (Ag) in the PMMA host.

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Sunil Deka, photograph and biography not available at the time of publication.

Waleed Mohammed, photograph and biography not available at the time of publication.