On the Scattering and Absorption by the SiO$_2$-VO$_2$ Core-Shell Nanoparticles Under Different Thermal Conditions

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ABSTRACT The core-shell kinds of nanoparticles are greatly useful in biomedical applications inclined to targeted drug delivery, sensing, etc. The electromagnetic interaction characteristics of silicon dioxide (SiO$_2$)-vanadium dioxide (VO$_2$) core-shell nanoparticle of deep subwavelength size are discussed. The assumptions allow the use of Rayleigh scattering theory for analytical approach. Because of VO$_2$ having inherent temperature-dependent constitutive characteristics, the light-matter interaction properties are taken into account in terms of normalized polarizability, scattering- and absorption cross-sections, considering the different values of size parameters as well as ambient temperature. It is found that the dimensional properties of chosen nanoparticles can be exploited to tune the spectral features, and also, the usefulness of these becomes obvious in temperature sensing. Moreover, the effect of interchanging the core/shell mediums (of the nanoparticles) on the scattering and absorption characteristics are also obtained. The results reveal the loss of resonance in this case.

INDEX TERMS Core-shell nanoparticle, scattering, polarizability.

I. INTRODUCTION Scattering of lightwaves due to micron- to nano-sized particles has been of great interest due to many possible technological advantages of such light-matter interactions [1]. Recently, the investigations pivoted to scattering and absorption of waves by specially designed core-shell nanoparticles have become of great attraction [2]–[4]. In general, the use of nanostructured mediums exhibit intriguing changes in the optical, magnetic, electronic, and biomedical properties for different dimensionality conditions, thereby allowing these to be prudent in biosensing, catalytic reactions, biology, material chemistry etc., [5]–[12]. Within the context, their use in bio-imaging and targeted drug delivery, gene delivery etc., [13]–[17] would be of special mention.

One of the important applications of core-shell nanoparticles would be the detection of cancer cells [18].

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References [19], [20] illustrate the use of such nanoparticles in tissue engineering. These are also useful for microwave absorption. It has been demonstrated that the Fe/Fe$_3$C–C core-shell nanoparticle has tunable high absorptivity in the microwave regime [21]. The volume of relevant research makes evident the use of core-shell nanoparticles in enhancing the solar cell efficiency [22], [23]. Such applications are more inclined to plasmonic photocatalysis that experienced considerable developments as an efficient technique for the conversion of solar energy [24]–[26], thereby proving to be prudent alternative to conventional (less efficient) semiconductor catalysis [25], [26]. Apart from these, the core-shell nanoparticles-based nanoantennas have been reported to yield improved directivity, and therefore, have potentials in the development of nanoscale laser and photovoltaic devices [27], [28].

The present work deals with the investigation of temperature dependent features of silicon dioxide (SiO$_2$)-vanadium dioxide (VO$_2$) core-shell nanoparticle. It is noteworthy
that the VO₂ medium has temperature dependent dielectric permittivity, thereby altering the optical properties under varying thermal conditions. We exploit this characteristics to study the polarizability, scattering and absorption cross-sections of the stated kind of core-shell nanoparticle in the wavelength span of 400–1600 nm, considering different values of the ambient temperature. Apart from this, the effect of dimensional characteristic of the SiO₂-VO₂ core-shell nanoparticle is also taken into account.

II. MODEL AND THEORY

Mie theory provides the exact solution to the scattering of waves due to a particle having its size comparable to the operating wavelength of the incidence radiation. However, if the particle size is very small as compared to the wavelength λ (i.e., ka ≪ 1; k and a, respectively, being the wave number and the radius of particle), the scattering object is considered to be a point source radiating like a Hertzian dipole. In such a case, the analytical treatment is made by exploiting the Rayleigh scattering theory. With this view point in mind, we consider the SiO₂-VO₂ core-shell nanoparticle, as shown in fig. 1. Here the core is comprised of SiO₂ medium, which is covered with a shell layer of VO₂ having its thickness as d. As such, the SiO₂ and VO₂ spherical mediums have the respective radii as rc and rs so that rs - rc = d. The medium around the VO₂ shell is free-space. Upon the interaction of core-shell nanoparticle with the incidence electromagnetic waves, the potentials due to the core, shell and the surrounding region of nanoparticle satisfy the boundary conditions [29], [30]. The Laplace equation provides approximately constant fields close to the origin in static limit.

Let us consider a time-harmonic plane wave of amplitude E₀, polarized along the z-direction, propagates in the x-direction, in order to excite the nanoparticle structure of fig. 1. Suppressing the time harmonic e⁻jωt factor, the incident wave can be written as

\[ \vec{E}_{inc} = \hat{z}E_0e^{jkx} \]  (1)

where \( k = \omega \sqrt{\epsilon_f} \) is the wave number; \( \epsilon_f \) and \( \mu_f \) being the free-space permittivity and free-space permeability, respectively.

We consider the mediums to be non-magnetic in nature, thereby allowing \( \mu_f = 1 \). For such light-matter interaction, the field solutions can be written [23]. Now, the corresponding equations for potential in each region of space, which satisfy boundary conditions, can be expressed as (2)–(4), shown at the bottom of this page. In these equations, \( \epsilon_c \) and \( \epsilon_s \) are the relative permittivity values of the core and shell mediums, respectively. Also, \( \varphi_c \), \( \varphi_s \) and \( \varphi_f \) are the potentials due to the core, shell and the surrounding medium, respectively, at a distance \( r \) from the observation point.

As it has been stated before that the dimensional size of nanoparticle is very small as compared to the incident wavelength, the polarizability of particle can be deduced from the amplitude of potential \( \varphi_f \) for the surrounding medium, as shown in eq. (4). With this view point in mind, the polarizability \( P_0 \) is proportional to the dipole moment [30], [31], i.e.,

\[ P_0 = 4\pi \epsilon_f r_s^3 \left( \frac{\epsilon_s-\epsilon_f}{\epsilon_c+2\epsilon_s} + \left( \frac{\epsilon_f}{r_s^3} \right)^3 \frac{\epsilon_c-\epsilon_s}{\epsilon_f+2\epsilon_s} \right) \]

By using eq. (5), the magnitude of dipole moment can be written as [31]

\[ II = -i \left( \frac{4\pi k r_s^3}{\eta_f} \right) \times \left( \frac{\epsilon_s-\epsilon_f}{\epsilon_c+2\epsilon_s} + \left( \frac{\epsilon_f}{r_s^3} \right)^3 \frac{\epsilon_c-\epsilon_s}{\epsilon_f+2\epsilon_s} \right) E_0 \]

where \( \eta_f \) is the impedance of free-space.
Now, by using eq. (5), the scattering cross-section $\sigma_{sca}$ can be derived by taking the square of the magnitude of polarizability; this ultimately yields

$$\sigma_{sca} = \frac{1}{6\pi \varepsilon_f} k^4 |P_0|^2$$

(7)

Also, the absorption cross-section $\sigma_{abs}$ can be calculated as

$$\sigma_{abs} = \frac{k}{\varepsilon_f} \text{Im}(P_0)$$

(8)

It is noteworthy that the resonance condition of core-shell nanoparticle is achieved when the real part of denominator in eq. (5) corresponds to a vanishing value. Furthermore, such a resonance may be achieved through properly choosing the dimensional values (of core/shell), because the denominator in eq. (5) consists of the ratio $(r_c/r_s)$ – the fact that can be utilized for the tuning of resonance condition. However, in the present investigation, we demonstrate the effects of core/shell dimensions and the ambient temperature on the polarizability, scattering cross-section and absorption cross-section characteristics of the nanoparticle structure under consideration.

III. RESULTS AND DISCUSSION

We now investigate the interaction of light with the SiO$_2$-VO$_2$ core-shell nanoparticle, considering different values of core-shell radii and ambient temperatures. For the purpose of analyses, we take the SiO$_2$ core permittivity $\varepsilon_c$ to be 2.25, whereas the VO$_2$ shell exhibits the temperature- and wavelength-dependent permittivity $\varepsilon_f$-values [32]–[34]. Within the context, refs. [33], [34] show the temperature-dependent phase transition of VO$_2$. However, small changes in the VO$_2$ phase transition temperature can be observed in different reports [35], [36]. For example, ref. [35] determines that VO$_2$ undergoes a structural transition from a dielectric monoclinic phase to a metallic rutile phase at 68 °C, whereas the refs. [33], [34] claim that phase transition to take place at 70 °C – the value that we consider in the present investigation. Considering the mediums to be non-magnetic in nature, we take the permeability of core and shell as unity in the optical regime.

We first investigate the polarizability of the proposed core-shell nanoparticle by taking into account the different values of shell radius and ambient temperature. At this point, one may wish to look at the effect of thermal hysteresis of VO$_2$, which depends more on the VO$_2$ deposition conditions [34]. This feature is, however, not considered in the present work. As such, we avoid the transition of VO$_2$ medium between the monoclinic and rutile phases, and consider only those temperatures in which the material remains only in one phase.

In order to study the polarizability, which is a complex quantity, we take the illustrative values of the ambient temperature to be 30 °C, 70 °C and 85 °C. Figure 2 exhibits the wavelength dependence of the real and imaginary parts of the normalized polarizability $P(\lambda)$ at the chosen temperatures corresponding to the dimensional values of core/shell as $r_c = 50$ nm and $r_s = 100$ nm; the span of operating wavelength is taken to be 400–1600 nm. It is noteworthy that VO$_2$ remains in the dielectric phase at 30 °C; as the temperature is increased, it attains a metallic phase. At 85 °C, it becomes completely metallic, whereas the phase transition starts happening at temperature $\sim$70 °C.

Figure 2 exhibits that, corresponding to all the illustrative values of temperature, the real part of polarizability $\Re(P)$ remains significantly larger than the imaginary component $\Im(P)$ of it. Further, corresponding to the dielectric phase (i.e., at 30 °C), the polarizability remains the least dependent on the variation in wavelength. However, in the 400–500 nm span of the visible regime, the $\Re(P)$ and $\Im(P)$ components exhibit ripples, thereby making the normalized polarizability $\Re(P)$ to be a little higher ($\sim$0.75) as compared to the remaining wavelength span, which exhibits $\Re(P) \sim 0.72$. At the same time, the value of $\Im(P)$ shows variation in the range of $\sim$0.02–0.10 corresponding to the used wavelength span.

With the increase in temperature, we observe in fig. 2 that the $\Re(P)$ and $\Im(P)$ parts of polarizability become highly wavelength dependent, thereby making the core-shell nanoparticles to be more dispersive in nature. The maximum variation in polarizability components appears corresponding to the metallic phase of VO$_2$ attained at 85 °C, which is well-justified. We notice that the normalized $\Re(P)$ reaches $\sim$1 at 85 °C corresponding to wavelength $\sim$1600 nm. In general, $\Re(P)$ goes on increasing as the wavelength is increased beyond $\sim$800 nm, and corresponding to 85 °C, the polarizability becomes maximum for large operating wavelengths. As the ambient temperature alters, it affects the permittivity value of VO$_2$, and therefore, the distribution of charged particles at the core-shell interface is reshaped, thereby modifying the polarizability of nanoparticle, as becomes obvious in fig. 2.

In order to observe the effect of nanoparticle dimensions on the polarizability characteristic, we plot figs. 3a, 3b and 3c...
Further, as the VO₂ has little effect on both the real and imaginary components of the core-shell nanoparticle. The wavelength dependence of the real and imaginary parts of polarizability was partially depicted in fig. 2 as well. Upon impact on the polarizability characteristic. This nature of polarizability depends on the size of nanoparticle as well, hence, the property of polarizability. In phase changing media like VO₂, the dipole moment essentially depends on the ambient temperature. As the polarizability is proportional to the dipole moment, it varies with temperature. Figure 3c – the case of metallic phase of VO₂ – exhibits the maximum dependence of polarizability on the ambient temperature. Though the polarizability depends on the size of nanoparticle as well, the impact of temperature remains more significant.

We now investigate the effect of shell radius and ambient temperature on the normalized scattering cross-section σ_{sca}. Within the context, we use the same parametric values of core-shell nanoparticle and operating conditions to determine the wavelength dependence of σ_{sca}; figs. 4a, 4b and 4c depict those corresponding to the temperatures 30 °C, 70 °C and 85 °C, respectively. Comparing these figures, the impacts of altering the temperature and dimensional parameters remain obvious.

In the case of metallic phase of VO₂ (fig. 4c), the scattering cross-section becomes more dependent on wavelength, and the least dependence is observed corresponding to the dielectric phase (fig. 4a). Apart from this, all these figures exhibit a slow increase. As such, a dip in the polarization patterns related to the σ_{P} components is observed at ~800 nm wavelength.

Upon further increasing the ambient temperature to 85 °C, the dispersive characteristic of nanoparticle becomes more prominent as the shell medium remains in the metallic phase. This becomes very clear from fig. 3c which exhibits the plots of the normalized σ_{P} and σ_{I} parts against wavelength, considering a fixed value of the SiO₂ core radius as 50 nm and varying values of the VO₂ shell radius as 70 nm, 80 nm, 90 nm and 100 nm. With the increase in shell dimension, the σ_{P} component of polarizability increases upon increasing the operating wavelength in the IR regime. An opposite trend can be seen in the visible regime along with a stronger dip in the normalized polarizability σ_{P} patterns at ~800 nm, as compared to that observed in fig. 3b; the dip becomes sharper with the increase in shell radius. As such, the effect of shell dimension on polarizability due to changing temperature becomes quite obvious.

It can thus be said that the geometrical factors leave very small impact on the resonance frequency. In fact, the ambient temperature remains the more dominant parameter to affect the resonance condition as it introduces phase change, and hence, the property of polarizability. In phase changing media like VO₂, the dipole moment essentially depends on the ambient temperature. As the polarizability is proportional to the dipole moment, it varies with temperature. Figure 3c – the case of metallic phase of VO₂ – exhibits the maximum dependence of polarizability on the ambient temperature. Though the polarizability depends on the size of nanoparticle as well, the impact of temperature remains more significant.

Corresponding to the aforementioned values of ambient temperature 30 °C, 70 °C and 85 °C, respectively. However, we vary the radius of shell, and keep the core radius r_c fixed. In such an attempt, we take the value of r_s to be 50 nm, and use four different values of shell radius r_s, namely 70 nm, 80 nm, 90 nm and 100 nm. These figures illustrate the wavelength dependence of the real and imaginary parts of polarizability of the core-shell nanoparticle.

Figure 3a clearly indicates that the increase in shell radius has little effect on both the real and imaginary components. Further, as the VO₂ medium remains in the dielectric phase at 30 °C, the effect of shell dimension hardly leaves significant impact on the polarizability characteristic. This nature of polarizability was partially depicted in fig. 2 as well. Upon increasing the ambient temperature to 70 °C, since the phase of shell (i.e., VO₂) tends to alter (toward the metallic phase), the normalized σ_{P} and σ_{I} parts experience the effects due to nanoparticle dimensions, though it is not highly pronounced. This is clear from fig. 3b, wherein we observe the alteration in the real and imaginary parts of polarizability with increasing wavelength. Indeed, a gradual transfer to the metallic phase (of the shell) makes the medium to assume more dispersive nature. Figure 3b depicts that the σ_{P} components of normalized polarizability decrease with increasing wavelength in the visible regime, and further increase in wavelength to the infrared (IR) regime causes σ_{P} to get a slow increase. As such, a dip in the polarization patterns related to the σ_{P} components is observed at ~800 nm wavelength.

FIGURE 3. Plots of the real and imaginary parts of normalized polarizability of nanoparticle against wavelength corresponding to (a) 30 °C, (b) 70 °C and (c) 85 °C ambient temperatures, and with r_c as 50 nm and r_s as 70 nm, 80 nm, 90 nm and 100 nm.
that the increase in shell radius results in relatively larger scattering, which is well-justified owing to the dimensional features. Furthermore, the scattering is significantly enhanced corresponding to the larger operating wavelengths – the characteristic which is also justified due to the ratio of light-matter interaction parameters. This is more pronounced in the metallic phase of VO$_2$ (fig. 4c), where the value of normalized $\sigma_{\text{sca}}$ becomes $\sim 1$ corresponding to the wavelength $\sim 1250$ nm and the shell radius of 100 nm. As stated before, the dielectric phase of VO$_2$ exhibits the normalized $\sigma_{\text{sca}}$ to be minimum dependent on wavelength (fig. 4a), except for a visible wavelength span of 400–500 nm where $\sigma_{\text{sca}}$ becomes a little higher, as compared with the remaining wavelength range. The scattering cross-section peaks correspond to the situation wherein the resonance is attained due to alteration of free-electrons on the VO$_2$ surface. In the metallic phase of VO$_2$, the density of free-electrons remains higher, and therefore, the resonance conditions become more prominent (fig. 4c), as compared to the situation when the VO$_2$ medium remains in the dielectric phase (fig. 4a).

We now move to investigate the absorption property of the SiO$_2$-VO$_2$ core-shell nanoparticle, considering the effects of altering the ambient temperature and dimensional features. All the operating and parametric conditions are kept as used before. Figures 5a, 5b and 5c exhibit the plots of the normalized absorption cross-section $\sigma_{\text{abs}}$ against wavelength in the 400–1600 nm span corresponding to the temperatures 30 °C, 70 °C and 85 °C, respectively.

We observe in fig. 5a that, at 30 °C ambient temperature, the normalized $\sigma_{\text{abs}}$ shows a little higher value in the visible regime of $\sim 400–550$ nm, and it decreases with the further increase in $\lambda$. As such, the visible wavelengths are more absorbed than the near-IR and IR wavelengths in the dielectric phase of shell (of nanoparticle). We also find that larger shell dimensions generally contribute to higher absorption of the incidence radiation. It becomes clear from fig. 5a that the use of 100 nm VO$_2$ shell radius results in the $\sigma_{\text{abs}}$-value $\sim 0.5$ at 400 nm wavelength, whereas reducing the shell radius...
to 70 nm causes the normalized $\sigma_{\text{abs}}$ to be $\sim 0.1$ at the same wavelength. The difference in $\sigma_{\text{abs}}$ values obtained for various shell dimensions keeps on reducing with the increase in wavelength, and it becomes nearly vanishing at a wavelength of 1600 nm. Therefore, the absorption of waves may not be achieved well corresponding to higher wavelengths, as long as the VO$_2$ medium of shell remains in the dielectric phase.

Upon increasing the temperature, the normalized $\sigma_{\text{abs}}$ values are significantly increased – this is well-reflected in fig. 5b, which corresponds to 70 °C. We also find that, at this transition temperature for the dielectric (to metallic) phase, the VO$_2$ shells of larger dimension exhibit higher absorption, which remains significant in the visible span of 400–550 nm wavelength – the feature observed in fig. 5a as well for the dielectric phase of VO$_2$ (i.e., at 30 °C). However, the noticeable fact remains that, apart from the significant increase of absorption in this phase, the difference in the normalized $\sigma_{\text{abs}}$ values corresponding to the nanoparticles of different dimensions is also increased with the increase in wavelength. Furthermore, the absorption is not more vanishing at large wavelength values, which is in contrast to what we observed before in the case of dielectric phase (fig. 5a).

We also find in fig. 5b that $\sigma_{\text{abs}}$ exhibits a little increase in the near IR wavelength of $\sim 1150$ nm, and then it decreases monotonically; the more the shell radius is, the higher the absorption becomes.

Corresponding to further increase in temperature to 85 °C, when the VO$_2$ shell medium attains a metallic phase, we observe the wavelength dependence of the normalized $\sigma_{\text{abs}}$ in fig. 5c. The features of absorption are greatly affected in this case, wherein we find that the $\sigma_{\text{abs}}$ values are largely increased, and also, two $\sigma_{\text{abs}}$ maxima are attained at $\sim 850$ nm and $\sim 950$ nm of the near-IR regime, which become stronger with the increase in VO$_2$ shell dimension. Corresponding to 100 nm shell radius, $\sigma_{\text{abs}}$ becomes $\sim 1$ at 400 nm and $\sim 950$ nm wavelengths. As such, we find that the absorption cross-section of the core-shell nanoparticle is greatly affected due to both the shell thickness and ambient temperature.

It must be stated at this point that the peaks corresponding to the scattering and absorption cross-sections relate to the resonance conditions in the core-shell nanoparticle. Additionally, these resonance peaks are prominent in the metallic phase of VO$_2$, as figs. 4c and 5c exhibit. These essentially attribute to the surface plasmon resonance condition [1] achieved at certain wavelengths – the phenomenon that yields the characteristics of absorption and scattering of waves.

Further to the present investigation, one would be curious to look at the spectral characteristics upon interchanging the materials used to form the core-shell nanoparticle, i.e., if the core and shell regions are formed by the VO$_2$ and SiO$_2$ mediums, respectively. With this viewpoint in mind, we present the obtained results of the wavelength dependence of the normalized scattering and absorption cross-sections in figs. 6 and 7, respectively, corresponding to the dielectric and metallic phases of VO$_2$ – the respective states that are attained at the ambient temperatures 70 °C and 85 °C. In these figures, the symbols $R_c$ and $R_s$ represent the radius of core and shell, respectively, of the nanoparticle.
Comparing fig. 6 with fig. 4 (a,c), and fig. 7 with fig. 5 (a,c), we observe that the interchange of mediums, which the nanoparticle is comprised of, essentially lifts up the resonance condition for both the scattering and absorption cross-sections. Figure 6 shows that the scattering is much reduced in this case (as compared to that observed in fig. 4), and the same happens for absorption spectra as well without significant resonance (fig. 7). As such, this kind of interchange (of mediums) in the nanoparticle would not be of enough potential for technological applications.

IV. CONCLUSION

The aforementioned results indicate the usefulness of SiO$_2$-VO$_2$ core-shell nanoparticle. The scattering and absorption properties essentially depend on the dimensional characteristics of nanoparticle, and also, the thermal condition of ambience. This is primarily because the VO$_2$ shell medium undergoes phase change with altering temperature. The shell becomes more dispersive in the metallic state – the fact that greatly affects the polarizability characteristics of nanoparticle. The similar behavior can be observed for the scattering and absorption cross-sections as well; the higher the temperature is, the more are the scattering and absorption of waves. It has been found that the scattering becomes large corresponding to longer wavelengths in the near-IR regime. Also, with the increase in shell dimension, the scattering cross-section increases. As to the absorption cross-section, its values become significantly large at ~900 nm wavelength in the metallic state, and increase as the shell dimension is increased. The dimensional features of core-shell nanoparticle can be used for the tuning of resonance condition, and the temperature-dependence of shell can be exploited for sensing applications in the biomedical arena. It is further observed that the interchange of core/shell mediums (of the nanoparticle) results into the loss of resonance.

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