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**Nanoparticles of TiO₂ and VO₂ in dielectric media:
Conditions for low optical scattering,
and comparison between effective medium and four-flux theories**

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Abstract

Spectral transmittance and reflectance in the 300 to 2500 nm solar-optical wavelength range were calculated for nanoparticles of titanium dioxide and vanadium dioxide with radii between 5 and 100 nm embedded in transparent dielectric media. Both of the materials are of large importance in green nanotechnologies: thus TiO₂ is a photocatalyst that can be applied as a porous film or a nanoparticle composite on indoor or outdoor surfaces for environmental remediation, and VO₂ is a thermochromic material with applications to energy-efficient fenestration. The optical properties, including scattering, of the nanoparticle composites were computed from the Maxwell–Garnett effective-medium theory as well as from a four-flux radiative transfer model. Predictions from these theories approach one another in the limit of small particles and in the absence of optical interference. Effects of light scattering can be modeled only by the four-flux theory, though. We found that nanoparticle radii should be less than ~20 nm in order to avoid pronounced light scattering.

Keywords: nanoparticles, optical properties, effective-medium theory, four-flux method

1. Introduction

Nanoparticles embedded in dielectric media have numerous important applications in the emerging field of green nanotechnology [1]. In this paper we consider nanoparticles of titanium dioxide and vanadium dioxide and investigate, in particular, the size ranges of interest for applications that do not permit pronounced light scattering in the $300 < \lambda < 2500$ nm wavelength range pertinent to visible light and solar radiation.

TiO₂ is a wide band gap dielectric material, and particles in porous coatings or applied as pigments in paints can have self-cleaning properties and be useful for environmental air purification when used on outdoor or indoor surfaces [2,3]. Other important applications of TiO₂ pertain to water remediation and to keeping surfaces clean and sterile [4,5]. Pigments containing TiO₂ are commonly used in white paint since they scatter visible light very efficiently provided that the particle size is of the order of a few hundred nm.

VO₂ is a thermochromic material and undergoes a metal–insulator transition at a “critical” temperature $\tau_c \approx 68$ °C [6]; this temperature can be decreased to room temperature through doping with, for example, tungsten [7,8]. The thermochromic transition affects the optical properties particularly in the near-infrared (NIR) at $700 < \lambda < 2500$ nm. Thin films of VO₂ are metallic and NIR-reflecting at $\tau > \tau_c$ and become NIR-transmitting at $\tau < \tau_c$. Thin films of VO₂ have been proposed as coatings for energy-efficient windows [1,8–11]. Furthermore, it has recently been found that nanoparticles of VO₂ can accomplish strong “nanothermochromic” modulation of solar energy transmittance through windows and lead to significantly improved performance [12–16].

Light scattering can be neglected for nanoparticles with radii much smaller than the wavelength, *i.e.*, for $r \ll \lambda$, but becomes of increasing importance as the size is increased [17]. This is of large practical significance, and it is frequently of interest to avoid visible haze by ensuring that the nanoparticles in a coating are small enough, particularly for applications on transparent substrates such as windows and glass facades in buildings. In these applications, haze is more pronounced at short wavelengths in the visible spectrum and, according to a widely used rule-of-thumb, the ratio of scattered to incident light should be below 0.01 to 0.02 in order not to affect the visible appearance of the coated glass. It follows that the size ranges that provide sufficiently small light scattering are of much interest. Electrically conducting nanoparticles with

$r \ll \lambda$ exhibit optical properties that are largely dominated by surface plasmon (SP) absorption [17]. The wavelength corresponding to the SPs can be tuned by changing the size and shape of the nanoparticles, which is important for applications to solar energy harvesting [1] as well as in other fields, including in fluorescence spectroscopy [18]. The nanoparticles' cross section for interaction with light is high, and therefore even a very small volume fraction of particles can have a pronounced effect on the optical properties of the coating. SPs are of importance for VO₂ nanoparticle composites, as will be demonstrated below.

One aim of this study is to determine the particle sizes where light scattering begins to be important in transparent coatings. As we will see, this size range lies at the border between the regions of validity for two different theoretical frameworks, and a secondary goal of the present work is to explore this transition region by comparing results given by two methods to compute optical spectra of nanoparticles embedded in dielectric coatings. The methods compared here are based on the Maxwell–Garnett effective-medium theory (EMT) [19–23] and the four-flux radiative transfer theory [24,25]. EMTs are often used for modeling optical properties of nanoparticle dispersions characterized by $r \ll \lambda$. This approach does not include light scattering, and the effective dielectric function obtained from EMTs can be used to compute transmittance and reflectance by standard thin-film theory [26,27]. The four-flux method, on the other hand, considers collimated and diffuse radiation fluxes going through the layer in the forward and backward directions [24] and obtains direct and diffuse transmittance as well as specular and diffuse reflectance from these fluxes. The fluxes depend on scattering and absorption coefficients ensuing from the interaction of light with particles as determined from Mie theory [28] or a generalization thereof. Another difference between the two theoretical approaches is that EMTs and thin-film theory consider coherent radiation and account for interference effects, while the four-flux theory is formulated in terms of light intensities so that it assumes incoherent light and no effects of interference.

Sec. 2 below outlines our computational methods, Sec. 3 presents model calculations for VO₂ and TiO₂ particles of various sizes embedded in a dielectric host medium, and Sec. 4 summarizes the main results and conclusions.

2. Theory

The structural model used in our calculations considers spherical nanoparticles of VO₂ and TiO₂ with dielectric constant ϵ_p embedded in a dielectric matrix with dielectric constant $\epsilon_m = 2.25$ (*i.e.*, the refractive index n_m is 1.5 and represents typical glasses and polymers). Fig. 1 shows a schematic picture of the relevant structure. Air is taken to be on both sides of the layer (*i.e.*, $\epsilon_s = 1$). The spectral transmittance of this structural model was computed with two approaches: the EMT and the four-flux method.

We consider EMTs based on the dipolar response of nanoparticles to an external field, which are valid roughly for $r < 0.1\lambda$. Extended EMTs that include higher multipoles are questionable for our purposes, since light scattering effects appear as absorption in these theories [29]. There are several different effective-medium formulations, all of which coincide in the dilute limit. The volume fraction is defined as $f = NV$, where N is the number of particles per unit volume and V is the volume of a single particle. We only consider a low volume fraction f , of the order 0.1 or below, in which case the Maxwell–Garnett (MG) theory [19,20] is a good approximation. The optical properties of the nanocomposite layer are then described by an effective dielectric function according to

$$\epsilon^{MG} = \epsilon_m \frac{1 + \frac{2}{3} f \alpha'}{1 - \frac{1}{3} f \alpha'} , \quad (1)$$

where the parameter α' , in the case of spherical particles, can be calculated from

$$\alpha' = 3(\epsilon_p - \epsilon_m) / (\epsilon_p + 2\epsilon_m) . \quad (2)$$

MG theory is appropriate for our case of nanoparticles dispersed in a continuous medium whereas other EMTs are adequate for different nanotopologies [30]. Spectral transmittance and reflection coefficients were then calculated using Fresnel's equations [26,27].

Our second approach is the four-flux method [24], which is a simplification of the multiple-scattering approach. It considers scattering and absorption of both direct and diffuse light beams propagating through the coating. The radiation field is modeled as consisting of four parts: collimated (c) beams going in the forward (along the incident beam, intensity I_c) and backward (intensity J_c) directions, and diffuse (d) beams going in forward (intensity I_d) and backward (intensity J_d) directions, as illustrated in Fig. 2.

Energy conservation of the radiation going through a layer containing particles leads to a set of coupled linear differential equations that can be written [24]

$$dI_c/dz = -(\alpha + \beta)I_c , \quad (3)$$

$$dJ_c/dz = (\alpha + \beta)J_c , \quad (4)$$

$$dI_d/dz = -\xi\beta I_d - \xi(1 - \sigma_d)\alpha I_d + \xi(1 - \sigma_d)\alpha J_d + \sigma_c\alpha I_c + (1 - \sigma_c)\alpha J_c , \quad (5)$$

$$dJ_d/dz = \xi\beta I_d + \xi(1 - \sigma_d)\alpha J_d - \xi(1 - \sigma_d)\alpha I_d - \sigma_c\alpha J_c - (1 - \sigma_c)\alpha I_c , \quad (6)$$

where z is the direction of the incident light and

$$\alpha = fC_{sca}/V , \quad (7)$$

$$\beta = fC_{abs}/V \quad (8)$$

are scattering and absorption coefficients per unit length, respectively, where V is the volume of a particle. Scattering and absorption cross sections, denoted C_{sca} and C_{abs} , respectively, were calculated from Mie theory [28,31] using an algorithm due to Wiscombe [32]. The average path-length parameter ξ is defined as the mean distance travelled by the diffuse fluxes as compared with those of the collimated fluxes. It has a value between unity for a perfectly collimated flux and two for an isotropic diffuse flux [24]. We tested the effect of different values of ξ on the optical transmittance and found it to be a minor one; specifically, differences in transmittance were less than 5% between the computations. We chose $\xi = 1.6$ for the computations to represent a situation of partially diffuse light in the coating. The forward scattering ratio for the collimated radiation, denoted σ_c , was calculated by a method given by Chylek [33]. As proposed by Maheu *et al.* [24], we set the forward scattering ratio for diffuse radiation equal to that of collimated light, *i.e.*,

$$\sigma_d = \sigma_c , \quad (9)$$

which is a simplification compared to the generalized form of the four-flux method as discussed by Vargas [25].

The differential equations above can be solved to obtain collimated and diffuse components of transmittance and reflectance, as shown by Maheu *et al.* [24]. The boundary conditions are then specified in terms of reflection coefficients for collimated and diffuse light. The reflection

coefficients can be obtained from Fresnel's equations [26,27], provided that an effective complex refractive index is specified for the particle-matrix composite.

In the limit of small particles, it is clear that an EMT—for example the MG formulation in Eqs. (1) and (2)—can be used in order to give consistent results in the non-scattering limit. For large and scattering particles, the calculation of the effective refractive index is a complex problem for which a general solution is lacking. For large particles well embedded in the medium, it may be appropriate to use the refractive index of the host material. In the present paper we employ an extended MG model, as in earlier work [34], in order to combine the correct small-particle limit with an approximate treatment of some effects of larger particles on the reflection coefficients. A thorough exposition of the computational method is presented elsewhere [34,35].

3. Results and discussion

Our computations employed literature data for the spectral dielectric functions of the nanoparticles. For TiO_2 , we used rutile-phase data averaged over the principal directions [36], while VO_2 was characterized by the dielectric function measured on 50-nm-thick films at $\tau > \tau_c$, and $\tau < \tau_c$ [37] (the dielectric function of VO_2 has been determined several times, and the various sets of data are in good agreement [14]). In neither case did we include any size dependence of the dielectric function; this approach is justified even for metallic VO_2 nanoparticles, since the relaxation time of the conduction electrons is exceptionally small—of the order of inter-atomic distances [38,39]—implying that surface scattering of these electrons is practically irrelevant.

We considered two layer thicknesses, 1 and 10 μm , with corresponding nanoparticle volume fractions 0.1 and 0.01, implying that the same amount of particle material was used in both cases. In the EMT, it was simply assumed that $r \ll \lambda$, while the four-flux calculations were performed for four values of r —specifically 5, 20, 50 and 100 nm—in order to explore the effects of scattering.

Fig. 3 shows transmittance and reflectance spectra of TiO_2 nanoparticles in a transparent medium. We first observe that the calculation based on EMT gives rise to prominent interference oscillations, which are absent in the four-flux calculations. The four-flux transmittance for the smaller particle sizes is in good agreement with the averaged interference-free transmittance, which is predicted to be $(T_{\min}T_{\max})^{1/2}$ according to thin-film theory [40], where $T_{\min}$ and $T_{\max}$ are

the minimum and maximum values for the oscillations in spectral transmittance, respectively. It is also clearly seen that EMT-based calculations fail to reproduce the spectral scattering behavior of TiO₂ nanoparticles with radii larger than 20 nm. Light scattering effects can be seen as a decrease in transmittance and an increase in reflectance at short wavelengths in the four-flux calculations. The scattering, and thus the reflectance, increases as the size of the nanoparticles is increased, which is apparent at the shortest wavelengths already for particles with $r = 20$ nm.

Figs. 4 and 5 present analogous data on spectral transmittance and reflectance for coatings containing VO₂ nanoparticles with semiconducting and metallic properties, *i.e.*, corresponding to $\tau < \tau_c$ and $\tau > \tau_c$, respectively. It is evident that the EMT and the four-flux method agree to within ± 0.02 , neglecting interference fringes, for small particles with radii up to 20 nm. The growing contribution of scattering for larger particles can be clearly seen as a lowering of the transmittance as the particle size is increased. This effect is prominent for semiconducting VO₂ in fig. 4. However, the reflectance displays only slight enhancements for the larger particles. The most remarkable feature in fig. 5 is the pronounced minimum for metallic VO₂ nanoparticles at $\lambda \approx 1200$ nm. The minimum is clearly caused by absorption, since the contribution from the nanoparticles to the total reflectance of the coating is only a minor one so that the reflectance curves are relatively featureless apart from the interference fringes in the EMT calculation. It is realized that the SP absorption leads to a significant modulation of solar energy transmittance around τ_c [12]. The difference between EMT and four-flux calculations is particularly large for large semiconducting VO₂ particles, while the SP absorption is dominating in the metallic case. The differences are caused by increased absorption due to the enhanced path-length of diffuse scattered light and are modeled only by four-flux theory. It is also apparent that the interference fringes inherent in the EMT-based computations are smaller than in the case of TiO₂ particles, which is a consequence of the significant absorption in VO₂.

4. Summary and conclusions

In the present work, we compared spectral transmittance and reflectance, as obtained from the Maxwell–Garnett effective-medium theory and the four-flux method, for VO₂ and TiO₂ nanoparticles embedded in a dielectric layer. Overall, we found good agreement between four-flux theory and interference-free EMT in the case of low-scattering, small nanoparticles, and

transparent coatings. Scattering is enhanced for increasing nanoparticle size and refractive index—*i.e.*, going from VO₂ to TiO₂—and the differences between the theories become more pronounced because EMT does not consider scattering. Scattering leads to lower transmittance and higher reflectance for TiO₂-pigmented coatings and to higher absorption for VO₂-based coatings. These effects start to be clearly seen for particle radii of 20 nm and are pronounced for radii in the range from 50 to 100 nm. Hence in order to avoid light scattering effects in VO₂ and TiO₂ nanoparticle coatings on window glass, for example, the particle size preferably should be around 20 nm or less. Titania nanoparticles are commercially available with a variety of sizes in this range, and nanocrystalline coatings are presently used on self-cleaning glass [3]. The size limitation may be more challenging in the case of VO₂, but the preparation of powders with average sizes below 20 nm has been reported recently [41].

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References

- [1] G.B. Smith, C. G. Granqvist, *Green Nanotechnology: Solutions for Sustainability and Energy in the Built Environment*, CRC Press, Boca Raton, USA, 2011.
- [2] K. Fujishima, K. Hashimoto, T. Watanabe, *TiO₂ Photocatalysis*, Bkc, Tokyo, Japan, 1999.
- [3] I.P. Parkin, R.G. Palgrave, Self-cleaning coatings, *Journal of Materials Chemistry* 15 (2005) 1689–1695.
- [4] A. Fujishima, X. Zhang, D.A. Tryk, TiO₂ photocatalysis and related phenomena, *Surface Science Reports* 63 (2008) 515–582.
- [5] U. Diebold, The surface science of titanium dioxide, *Surface Science Reports* 48 (2003) 53–229.
- [6] J. Morin, Oxides which show a metal-to-insulator transition at the Neel temperature, *Physical Review Letters* 3 (1959) 34–36.
- [7] J.B. Goodenough, The two components of the crystallographic transition in VO₂, *Journal of Solid State Chemistry* 3 (1971) 490–500.
- [8] S.-Y. Li, G.A. Niklasson, C.G. Granqvist, Thermochromic fenestration with VO₂-based materials: three challenges and how they can be met, *Thin Solid Films* 520 (2012) 3823–3828.
- [9] M. Saeli, C. Piccirillo, I.P. Parkin, R. Binions, I. Ridley, Energy modeling studies of thermochromic glazing, *Energy and Buildings* 42 (2010) 1666–1673.
- [10] M. Saeli, C. Piccirillo, I. P. Parkin, I. Ridley, R. Binions, Nano-composite thermochromic thin films and their application in energy-efficient glazing, *Solar Energy Materials and Solar Cells* 94 (2010) 141–151.
- [11] Y. Gao, H. Luo, Z. Zhang, L. Kang, Z. Chen, J. Du, M. Kanehira, C. Cao, Nanoceramic VO₂ thermochromic smart glass: a review on progress in solution progressing, *Nano Energy* 1 (2012) 221–246.
- [12] S.-Y. Li, G.A. Niklasson, C.G. Granqvist, Nanothermochromics: calculations for VO₂ nanoparticles in dielectric hosts show much improved luminous transmittance and solar energy transmittance modulation, *Journal of Applied Physics* 108 (2010) 063525/1–063525/8.
- [13] S.-Y. Li, G.A. Niklasson, C.G. Granqvist, Nanothermochromics with VO₂-based core-shell structures: calculated luminous and solar optical properties, *Journal of Applied Physics* 109 (2011) 113515/1–113515/5.
- [14] S.-Y. Li, K. Namura, M. Suzuki, G.A. Niklasson, C.G. Granqvist, Thermochromic VO₂ nanorods made by sputter deposition: growth conditions and optical modelling, *Journal of Applied Physics* 114 (2013) 033516/1–033516/11 [Erratum: *Journal of Applied Physics* 114 (2013) 239902/1].
- [15] S.-Y. Li, G.A. Niklasson, C.G. Granqvist, Thermochromic undoped and Mg-doped VO₂ thin films and nanoparticles: Optical properties and performance limits for energy efficient windows, *Journal of Applied Physics*, DOI: 10.1063/1.4862930 (10 pages).
- [16] Y. Zhou, A. Huang, Y. Li, S. Ji, Y. Gao, P. Jin, Surface plasmon resonance induced excellent solar control for VO₂@SiO₂ nanorods-based thermochromic foils, *Nanoscale*, to be published, DOI: 10.1039/C3NR02221H.
- [17] U. Kreibig, M. Vollmer, *Optical Properties of Metal Clusters*, Springer, Berlin, Germany, 1995.
- [18] J. Lakowicz, *Principles of Fluorescence Spectroscopy*, Springer, Berlin, Germany, 2006.
- [19] J.C. Maxwell Garnett, Colours in metal glasses and thin films, *Philosophical Transactions of the Royal Society of London, Series A*, 203 (1904) 385–420.

- [20] J.C. Maxwell Garnett, Colours in metal glasses, in metallic films, and in metallic solutions, *Philosophical Transactions of the Royal Society of London, Series A*, 205 (1906) 237–288.
- [21] G.A. Niklasson, Optical properties of inhomogeneous two-component materials, in *Materials Science for Solar Energy Conversion Systems*, edited by C.G. Granqvist, Pergamon, Oxford, UK, 1991, pp. 7–43.
- [22] R. Landauer, Electrical conductivity in inhomogeneous media, *American Institute of Physics Conference Proceedings* 40 (1978) 2–45.
- [23] D.E. Aspnes, Optical properties of thin films, *Thin Solid Films* 89 (1982) 249–262.
- [24] B. Maheu, J.N. Letoulouzan, G. Gouesbet, Four-flux models to solve the scattering transfer equation in terms of Lorenz–Mie parameters, *Applied Optics* 23 (1984) 3353–3362.
- [25] W.E. Vargas, Generalized four-flux radiative transfer model, *Applied Optics* 37 (1998) 2615–2623.
- [26] M. Born, E. Wolf, *Principles of Optics*, 7th ed., Cambridge University Press, Cambridge, UK, 1999.
- [27] R.M. Azzam, N.M. Bashara, *Ellipsometry and Polarized Light*, North-Holland, Amsterdam, The Netherlands, 1977.
- [28] G. Mie, Beiträge zur Optik trüber Medien, speziell kolloidaler Metallösungen, *Annalen der Physik (Leipzig)*, Vierte Folge, 25 (1908) 377–445.
- [29] C.F. Bohren, Applicability of effective-medium theories to problems of scattering and absorption by nonhomogeneous atmospheric particles, *Journal of Atmospheric Sciences* 43 (1986) 468–475.
- [30] G.A. Niklasson, C.G. Granqvist, O. Hunderi, Effective medium models for the optical properties of inhomogeneous materials, *Applied Optics* 20 (1981) 26–30.
- [31] C.F. Bohren, D.R. Huffman, *Absorption and Scattering of Light by Small Particles*, Wiley, New York, USA, 1983.
- [32] W.J. Wiscombe, Improved Mie scattering algorithms, *Applied Optics* 19 (1980) 1505–1509.
- [33] P. Chylek, Mie scattering into the backward hemisphere, *Journal of the Optical Society of America* 63 (1973) 1467–1471.
- [34] G.A. Niklasson, T.S. Eriksson, Radiative cooling with pigmented polyethylene foils, *Proceedings of SPIE – The International Society for Optical Engineering* 1016 (1988) 89–99.
- [35] W.E. Vargas, G.A. Niklasson, Pigment mass density and refractive index determination from optical measurements, *Journal of Physics: Condensed Matter* 9 (1997) 1661–1670.
- [36] E.D. Palik, *Handbook of Optical Constants of Solids*, Academic, New York, USA, 1985.
- [37] N.R. Mlyuka, G.A. Niklasson, C.G. Granqvist, Thermochromic VO₂-based multilayer films with enhanced luminous transmittance and solar modulation, *Physica Status Solidi A* 206 (2009) 2155–2160.
- [38] K. Okazaki, S. Sugai, Y. Muraoka, Z. Hiroi, Role of electron–electron and electron–phonon interaction effects in the optical properties of VO₂, *Physical Review B* 73 (2006) 165116/1–165116/5.
- [39] A. Gentle, A.I. Maarouf, G.B. Smith, Nanograin VO₂ in the metal phase: a plasmonic system with falling dc resistivity as temperature rises, *Nanotechnology* 18 (2007) 025202/1–025202/7.
- [40] R. Swanepoel, Determination of thickness and optical constants of amorphous silicon, *Journal of Physics E: Scientific Instruments* 16 (1983) 1214–1222.
- [41] S. Ji, F. Zhang, P. Jin, Preparation of high performance pure single phase VO₂ nanopowder by hydrothermally reducing the V₂O₅ gel, *Solar Energy Materials and Solar Cells* 95 (2011) 3520–3526.

Figure captions

Fig. 1. Structural model for a composite of spherical nanoparticles with radius r embedded in a dielectric medium. The dielectric constants of the particles, matrix, and surrounding are denoted ε_p , ε_m , and ε_s , respectively.

Fig. 2. Electromagnetic radiation fluxes inside an illuminated inhomogeneous layer. Fluxes for collimated (c) and diffuse (d) fluxes are indicated in the forward (I) and backward (J) directions.

Fig. 3. Spectral transmittance (left-hand panels) and reflectance (right-hand panels) for TiO₂ nanoparticles embedded in a dielectric medium with $n_m = 1.5$ calculated with the four-flux method for the shown nanoparticle radii r and with effective medium theory (EMT). Two combinations of layer thickness d and volume fraction of nanoparticles f are used: $d = 10 \mu\text{m}$ and $f = 0.01$ (upper panels) and $d = 1 \mu\text{m}$ and $f = 0.1$ (lower panels).

Fig. 4. Spectral transmittance (left-hand panels) and reflectance (right-hand panels) for semiconducting VO₂ nanoparticles embedded in a dielectric medium with $n_m = 1.5$ calculated with the four-flux method for the shown nanoparticle radii r and with effective medium theory (EMT). Two combinations of layer thickness d and volume fraction of nanoparticles f are used: $d = 10 \mu\text{m}$ and $f = 0.01$ (upper panels) and $d = 1 \mu\text{m}$ and $f = 0.1$ (lower panels).

Fig. 5. Spectral transmittance (left-hand panels) and reflectance (right-hand panels) for metallic VO₂ nanoparticles embedded in a dielectric medium with $n_m = 1.5$ calculated with the four-flux method for the shown nanoparticle radii r and with effective medium theory (EMT). Two combinations of layer thickness d and volume fraction of nanoparticles f are used: $d = 10 \mu\text{m}$ and $f = 0.01$ (upper panels) and $d = 1 \mu\text{m}$ and $f = 0.1$ (lower panels).

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5