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OPTICAL PROPERTIES OF MONOLAYER OF SPHERICAL PARTICLES IN ABSORBING HOST MEDIUM UNDER NORMAL ILLUMINATION

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Abstract

The method is developed to describe optical properties of a normally illuminated monolayer (2D array) of spherical particles embedded in a homogeneous absorbing host medium (matrix). It is based on multipole expansion of electromagnetic (EM) fields and dyadic Green's function in terms of vector spherical waves functions and takes into account multiple scattering of waves in the quasicrystalline approximation. The method is applied to describe the scattering and absorbing properties of partially ordered monolayers of spherical voids in silver (Ag) matrix.

1 Introduction

The particulate layers are object of intensive investigations in the last decades. The interest is caused by unique possibilities which are opened by usage of these layers in optics, optoelectronics, photonics and nanophotonics, medical applications, etc. The majority of work devoted to these structures imply that absorbing or nonabsorbing particles are located in a nonabsorbing host medium. However, some applications require consideration of particulate layers in absorbing environment, e.g. solar cells, photonic crystals, ocean optics, etc. There are publications where optical properties of single particles [1] or sparse ensembles of particles [2,3] in absorbing host medium are considered. But we are not aware of publications in which densely packed monolayers of particles in an absorbing medium would be studied.

We present a brief summary of the method we are developing for determining the optical properties of a monolayer of spherical homogeneous particles in an absorbing host medium (matrix). This method is used to describe the optical characteristics of a monolayer of identical densely packed partially ordered spherical voids in a silver plate.

2 Theory

We consider the monolayer of N identical spherical particles centered at the points $\mathbf{R}_1$, $\mathbf{R}_2$, …, $\mathbf{R}_N$ in the monolayer plane (x,y). The coordinate origin O is in the center of arbitrarily chosen particle (Figure. 1). The monolayer stands in an absorbing matrix and is normally illuminated by the plane EM wave with electric vector $\mathbf{E}_0$ and unit polarization vector $\hat{\boldsymbol{\varepsilon}}_0 = \varepsilon_x \hat{\mathbf{x}} + \varepsilon_y \hat{\mathbf{y}}$:

$$\mathbf{E}_0 = \hat{\boldsymbol{\varepsilon}}_0 E_0 e^{i k_h z}, \tag{1}$$

where $k_h=2\pi m_h/\lambda$ is complex wavenumber, $m_h=n_h+i\kappa_h$, n_h and κ_h are refractive and absorption indices of host medium, λ is the wavelength of incident wave in vacuum, E_0 is the amplitude of incident wave in the monolayer plane (z=0).

The field $\mathbf{E}(\mathbf{r})$ in some point $\mathbf{r}$ is the sum of fields of incident wave and waves scattered by the given configuration of particulate ensemble:

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + \sum_{j=1}^{N} \mathbf{F}_j(\mathbf{r}, \mathbf{R}_j) \cdot \tag{2}$$

Here

$$\mathbf{F}_j(\mathbf{r}, \mathbf{R}_j) = \frac{(k_p{}^2 - k_h{}^2)}{4\pi} \int_{V_p} \mathrm{d}\mathbf{r}' \overleftrightarrow{\mathrm{G}}(\mathbf{r}, \mathbf{R}_j + \mathbf{r}') \cdot \mathbf{E}(\mathbf{R}_j + \mathbf{r}') \tag{3}$$

is the field created in point $\mathbf{r}$ by the particle centred at point $\mathbf{R}_j$, $k_p=2\pi m_p/\lambda$ is complex wavenumber, $m_p=n_p+i\kappa_p$, n_p and κ_p are refractive and absorption indices of particle, V_p is the particle volume, $0 \le |\mathbf{r}'| \le D/2$, D is the particle diameter, $\mathbf{E}(\mathbf{R}_j+\mathbf{r}')$ is the field in point $\mathbf{R}_j+\mathbf{r}'$ within the particle (internal field), $\overleftrightarrow{\mathrm{G}}$ is the dyadic (tensor) Green's function:

$$\overleftrightarrow{\mathrm{G}}(\mathbf{r}, \mathbf{r}_0) = \left[\overleftrightarrow{\mathrm{I}} + \frac{1}{k_h^2} \nabla \otimes \nabla \right] \frac{e^{i k_h |\mathbf{r} - \mathbf{r}_0|}}{|\mathbf{r} - \mathbf{r}_0|}, \tag{4}$$

$\overleftrightarrow{\mathrm{I}}$ is identity dyadic.

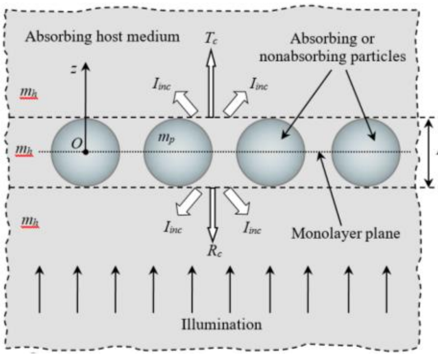

***Figure 1** Schematic view (along the monolayer plane) of normally illuminated monolayer of spherical particles embedded in absorbing host medium (matrix). T_c and R_c are coherent transmission and reflection coefficients of the system consisting of the monolayer of particles with diameter D and matrix layer with thickness D. I_{inc} is the intensity of the incoherent light.*

Averaging the Eqs. (2) and (3) over all possible ensemble configurations results in the following equations [4]:

$$\begin{aligned}\langle \mathbf{E}(\mathbf{r})\rangle &= \mathbf{E}_0(\mathbf{r}) + \sum_{j=1}^{N}\langle \mathbf{F}_j(\mathbf{r},\mathbf{R})\rangle = \mathbf{E}_0(\mathbf{r}) + \\ &+ \frac{({k_p}^2-k_h^2)}{4\pi}\rho_0\int d\mathbf{R}\int_{V_p} d\mathbf{r}'\vec{\mathrm{G}}(\mathbf{r},\mathbf{R}+\mathbf{r}')\cdot\langle \mathbf{E}(\mathbf{r}')\rangle_1\end{aligned}, \quad (5)$$

$$\begin{aligned}\langle \mathbf{E}(\mathbf{r}')\rangle_1 &= \mathbf{E}_0(\mathbf{r}') + \frac{({k_p}^2-k_h^2)}{4\pi}\int_{V_p} d\mathbf{r}''\vec{\mathrm{G}}(\mathbf{r}',\mathbf{r}'')\cdot\langle \mathbf{E}(\mathbf{r}'')\rangle_1 + \\ &+ \frac{({k_p}^2-k_h^2)}{4\pi}\rho_0\int d\mathbf{R}g(R)\int_{V_p} d\mathbf{r}''\vec{\mathrm{G}}(\mathbf{r}',\mathbf{R}+\mathbf{r}'')\cdot\langle \mathbf{E}(\mathbf{r}'')\rangle_1\end{aligned}. \quad (6)$$

Here ρ_0 is averaged number density (concentration) of particles, g(R) is the radial distribution function (RDF) [4,5] characterizing the probability to find the particle centred at distance R relative the particle centred at the coordinate origin, $R=|\mathbf{R}|$, $\langle\mathbf{E}(\mathbf{r}')\rangle_1$ is the field in the particle centred at the coordinate origin, averaged over the positions of all other particles (averaged internal field). It determined by Eq. (6) with applying the quasicrystalline approximation (QCA) [6].

The field in the far zone in the direction $\hat{\mathbf{r}}$ for the single monolayer particle is determined via the amplitude scattering function:

$$\mathbf{f}(\hat{\mathbf{r}}) = \frac{({k_p}^2-k_h^2)k_h}{4\pi}\int_{V_p} d\mathbf{r}'\left(\vec{\mathrm{I}}-\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}\right)e^{-ik_h\hat{\mathbf{r}}\cdot\mathbf{r}'}\cdot\langle \mathbf{E}(\mathbf{r}')\rangle_1 \cdot \quad (7)$$

It takes into account the multiple scattering of waves. To determine $\mathbf{f}(\hat{\mathbf{r}})$ we expand the functions entered in (6), (7) in terms of vector spherical wave functions [4]. As a result, we obtain:

$$\mathbf{f}(\hat{\mathbf{r}}) = E_0[\mathrm{f}_\theta(\hat{\mathbf{r}})\hat{\boldsymbol{\theta}} + \mathrm{f}_\varphi(\hat{\mathbf{r}})\hat{\boldsymbol{\varphi}}], \quad (8)$$

where

$$\mathrm{f}_\theta(\hat{\mathbf{r}}) = i\sum_{l=1}^{\infty}\frac{(2l+1)}{l(l+1)}\left[\begin{aligned}&\pi_l^{(1)}(\mu)\cdot\left(b_{lM}^{(o)}\cos\varphi + ib_{lM}^{(e)}\sin\varphi\right) + \\ &+\tau_l^{(1)}(\mu)\cdot\left(b_{lE}^{(e)}\cos\varphi + ib_{lE}^{(o)}\sin\varphi\right)\end{aligned}\right], \quad (9)$$

$$\mathrm{f}_\varphi(\hat{\mathbf{r}}) = -\sum_{l=1}^{\infty}\frac{(2l+1)}{l(l+1)}\left[\begin{aligned}&\tau_l^{(1)}(\mu)\cdot\left(b_{lM}^{(e)}\cos\varphi + ib_{lM}^{(o)}\sin\varphi\right) + \\ &+\pi_l^{(1)}(\mu)\cdot\left(b_{lE}^{(o)}\cos\varphi + ib_{lE}^{(e)}\sin\varphi\right)\end{aligned}\right]. \quad (10)$$

Angular functions $\pi_l^{(1)}(\mu)=\mathrm{P}_l^{(1)}(\cos\theta)/\sin\theta$, $\tau_l^{(1)}(\mu)=d\mathrm{P}_l^{(1)}(\cos\theta)/d\theta$, where $\mu=\cos\theta$, $\mathrm{P}_l^{(1)}(\cos\theta)$ are associated Legendre function, $\hat{\boldsymbol{\theta}}$ and $\hat{\boldsymbol{\varphi}}$ are unit vectors in the directions determined by polar θ and azimuthal φ scattering angles.

The expansion coefficients $b_{lM}^{(o,e)}$ и $b_{lE}^{(o,e)}$ take into account multiple scattering of waves and determine the contribution of multipoles of different orders into the total scattering field. They are calculated by analogy with the technique described in [4]. Using these coefficients, we can write the equations for coherent transmission T_c and coherent reflection R_c coefficients:

$$T_c = e^{-4x\beta_h}\left[\begin{aligned}&\left|\varepsilon_x - \frac{\eta}{x^2(1+i\beta_h)^2}\sum_{l=1}^{\infty}(2l+1)\left(b_{lM}^{(o)}+b_{lE}^{(e)}\right)\right|^2 + \\ &+\left|\varepsilon_y - i\frac{\eta}{x^2(1+i\beta_h)^2}\sum_{l=1}^{\infty}(2l+1)\left(b_{lM}^{(e)}+b_{lE}^{(o)}\right)\right|^2\end{aligned}\right], \quad (11)$$

$$R_c = e^{-4x\beta_h}\left(\frac{\eta}{x^2(1+i\beta_h)^2}\right)^2\left[\begin{aligned}&\left|\sum_{l=1}^{\infty}(-1)^l(2l+1)\left(b_{lM}^{(o)}-b_{lE}^{(e)}\right)\right|^2 + \\ &+\left|\sum_{l=1}^{\infty}(-1)^l(2l+1)\left(b_{lM}^{(e)}-b_{lE}^{(o)}\right)\right|^2\end{aligned}\right], \quad (12)$$

where $x=\pi Dn_h/\lambda$ is the particle size parameter in the host medium, $\beta_h=\kappa_h/n_h$, η is the monolayer filling factor describing the ratio of the area of all particle projections into the monolayer plane to the area where the particles are distributed.

The incoherent scattering coefficient F_{inc} of a monolayer is determined as follows:

$$F_{inc} = \int_0^{2\pi}d\varphi\int_0^{\pi}I_{inc}^{rd}(\theta,\varphi)e^{-2x\beta_h[1+1/|\cos\theta|]}\sin\theta\, d\theta \cdot \quad (13)$$

Here I_{inc}^{rd} is the reduced intensity [4] of incoherently scattered light:

$$I_{inc}^{rd}(\theta,\varphi) = \frac{\eta \mathrm{S}_2(\sin\theta)}{\pi x^2(1+\beta_h^2)}\times$$
$$\times\left\{\begin{aligned}&\left|\sum_{l=1}^{\infty}\frac{(2l+1)}{l(l+1)}\left[\begin{aligned}&\pi_l^{(1)}(\mu)\cdot\left(b_{lM}^{(o)}\cos\varphi+ib_{lM}^{(e)}\sin\varphi\right)+\\&+\tau_l^{(1)}(\mu)\cdot\left(b_{lE}^{(e)}\cos\varphi+ib_{lE}^{(o)}\sin\varphi\right)\end{aligned}\right]\right|^2 + \\ &+\left|\sum_{l=1}^{\infty}\frac{(2l+1)}{l(l+1)}\left[\begin{aligned}&\tau_l^{(1)}(\mu)\cdot\left(b_{lM}^{(e)}\cos\varphi+ib_{lM}^{(o)}\sin\varphi\right)+\\&+\pi_l^{(1)}(\mu)\cdot\left(b_{lE}^{(o)}\cos\varphi+ib_{lE}^{(e)}\sin\varphi\right)\end{aligned}\right]\right|^2\end{aligned}\right\}, \quad (14)$$

were $\mathrm{S}_2(\sin\theta)$ is the structure factor of a monolayer [4,7]:

$$\mathrm{S}_2(\sin\theta) = 1 + 8\eta\int_0^{\infty}[\mathrm{g}(u)-1]\mathrm{J}_0(2xu\sin\theta)u\,du, \quad (16)$$

$u=R/D$ is the dimensionless distance in the monolayer plane expressed in the particle diameters D.

The absorption coefficient A of a monolayer is defined using the (11)-(13) as follows:

$$A=1-T_c-R_c-F_{inc}. \quad (15)$$

These coefficients characterize only the system consisting of the layer of host medium of thickness D (bounded by dashed lines in Figure 1) containing the monolayer of spheres with diameter D. They do not take into account absorption of host medium outside the monolayer.

3 Results of Calculations

We applied the developed method to consider the optical properties of the system consisting of the partially ordered submicron spherical voids in a homogeneous silver plate.

Figure 2 shows the spectral dependences of T_c, R_c, F_{inc} and A coefficients of such system at different diameters D of voids. One can see that in the spectral region of plasmonic resonance (λ=0.3 - 0.4 μm) the coherent transmittance, as a whole, grows with decreasing the void diameter (Figure 2a). For small voids, D≤0.05 μm, the "extraordinary transmittance" is observed. Less pronounced peaks occur outside this region for larger voids.

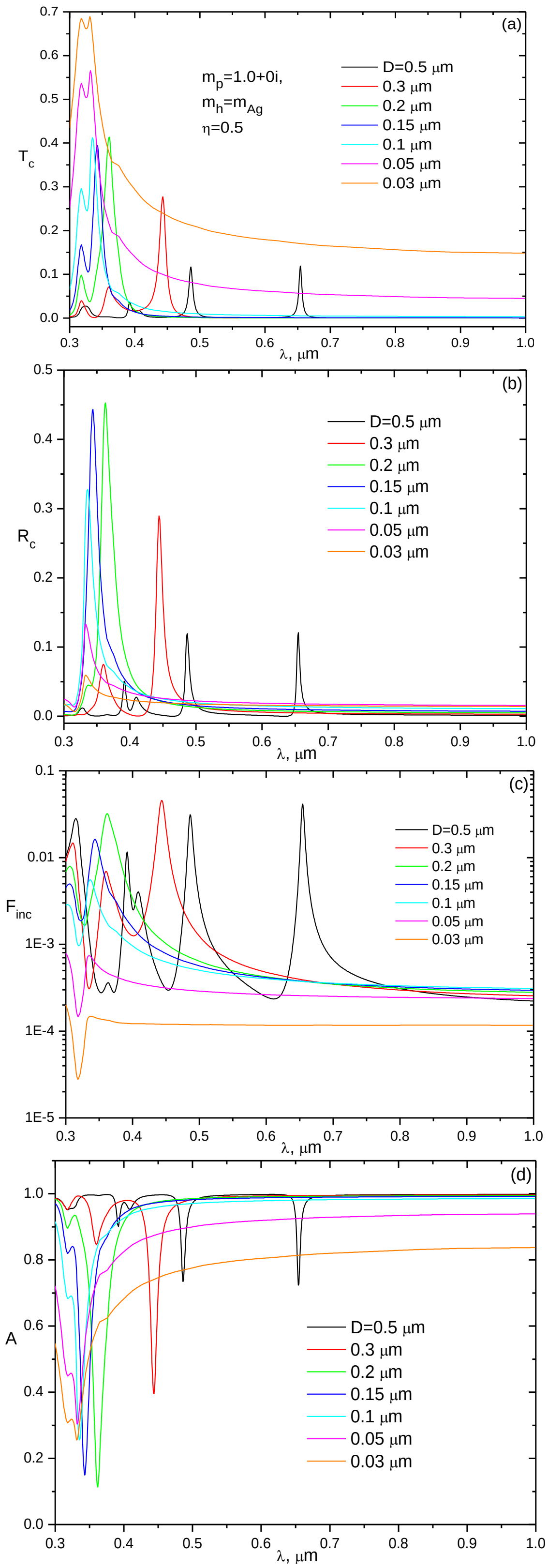

Figure 2 *Spectral dependences of T_c, R_c, F_{inc} and A coefficients of the partially ordered monolayer of spherical voids in silver (Ag) plate at different diameters D of voids. η=0.5*

From Figure 2 it follows that for larger voids, the strong correlation takes place between T_c, R_c, F_{inc} and A spectra, especially in the wavelength range outside the plasmonic resonance region. In contrast to the monolayer of silver particles in air [8] where dips in the transmission and peaks in the absorption coefficients spectra take place, here the peaks in $T_c(\lambda)$ and dips in $A(\lambda)$ are observed. Also, the low amount of incoherently scattered (Figure 2c) and large amount of absorbed (Figure 2d) light take place.

4 Conclusions

The method to describe optical properties of a normally illuminated monolayer of homogeneous identical spherical particles in an absorbing host medium is developed. It is based on multipole expansion of electromagnetic (EM) fields and dyadic Green's function in terms of vector spherical waves functions and takes into account multiple scattering of waves in the quasicrystalline approximation.

The method is applied to describe scattering and absorbing properties of the system consisting of the monolayer of voids of various diameters D in the silver matrix. The effect of "extraordinary transmittance" is observed for the monolayer of nanosized (D≤0.05 μm) voids.

The method and obtained results can be used to investigate and optimize various optical, optoelectronic, and photonic devices, such as solar cells, light emitting diodes, optical filters, etc.

5 References

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