Light scattering by a disperse layer of closely packed two-layered spherical particles

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Light scattering by a layer of closely packed two-layered non-absorbing spherical particles (core-shell particles) is investigated. The interference approximation is used to take into account collective scattering effects. The darkening effect and spectral dependence of the scattering coefficient are considered under this approximation.

INTRODUCTION

Light scattering by natural and artificial media with a high concentration of particles has drawn the attention of researchers recently [1]. For a strict description of scattering in densely packed media it is necessary to use the theory of multiple scattering of waves (MSW) [2]. Because of the complexity and cumbersomeness of the mathematical apparatus of the MSW theory, complete solutions are obtained in rare instances. The effects caused by ordered arrangements of optically soft particles in large concentrations are referred to such instances. In the present work the interference approximation is used to study light scattering by a system of spherical particles of the core-shell type.

ELEMENTARY VOLUME CHARACTERISTICS FOR A MEDIUM WITH SPHERICAL PARTICLES

In accordance with the interference approximation, expressions for differential scattering and extinction coefficients for a medium consisting of identical spherical particles can be written in the form:

$$\sigma_h(\gamma) = w\sigma_{0l}\, p_l(\gamma) S_3(\gamma, w), \tag{1}$$

$$\sigma_h = w\sigma_{0l} u, \tag{2}$$

$$\varepsilon_h = w\left(\varepsilon_{0l} - \sigma_{0l} + \sigma_{0l} u\right), \tag{3}$$

$$u = \int_0^\pi p_l(\gamma) S_3(\gamma, w) \sin\gamma d\gamma, \tag{4}$$

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where $w = Nv/V$ is the volume concentration of the particles; N is the number of particles with volume $v = \frac{4}{3}\pi R^3$, contained in volume V of the medium; R is the particle radius; $\sigma_h(\gamma)$ is the differential scattering coefficient of a medium with a volume concentration w of particles; γ is the scattering angle; $\sigma_{0l} = \Sigma_s/v$; $\varepsilon_{0l} = \alpha_{0l} + \sigma_{0l} = \Sigma_e/v$; $\alpha_0 = \Sigma_a/v$; Σ_a, Σ_s and Σ_e are the absorption, scattering and extinction cross sections of an individual particle; and $p_l(\gamma)$ is the phase function of an individual particle normalised by the condition $\int_0^\pi p_l(\gamma)\sin\gamma d\gamma = 1$.

The structure factor $S_3(\gamma,w)$ takes into account the effect of light interference occurring in a system of correlated particles:

$$S_3(\gamma,w) = 1 + 4\pi n\int_0^\infty [g(r,w) - 1]\frac{\sin zr}{zr}r^2 dr. \tag{5}$$

Here n is the number of particles in the unit volume; $g(r,w)$ is the radial distribution function characterizing the spatial arrangement of particles, $z = 4x\sin\gamma/2$.

For a system with a hard sphere potential, the structure factor can be calculated under the Percus-Yevick approximation [3,4]:

$$S_3(\gamma,w) = \left(1 - 24n\int_0^1 g(r,w)\frac{\sin zr}{zr}r^2 dr\right)^{-1}, \tag{6}$$

where

$$g(r,w) = \begin{cases} -a - b\dfrac{r}{2R} - c\left(\dfrac{r}{2R}\right)^3, r \le 2R \\ 0, r > 2R \end{cases}, \tag{7}$$

$$a = \frac{(1+2w)^2}{(1-w)^4}, \tag{8}$$

$$b = -6w\frac{(1+0.5w)^2}{(1-w)^4}, \tag{9}$$

$$c = 0.5w\frac{(1+2w)^2}{(1-w)^4}. \tag{10}$$

Thus the optical properties of the medium of closely packed particles are defined by the phase function of a separate particle $p_l(\gamma)$ and the structure factor $S_3(\gamma,w)$. To calculate scattering characteristics of particles, the algorithms described in [5] were used.

CHARACTERIZATION OF ENSEMBLE OF SPHERICAL HOMOGENEOUS PARTICLES AND ENSEMBLE OF TWO-LAYERED SPHERICAL PARTICLES

The deviation of parameter *u* from unity characterises the degree of particles interaction. For spherical homogeneous particles, increasing the concentration decreases the value *u* monotonically. The spectral dependence of parameter *u* is not monotonic. The dependence of parameter *u* on relative wavelength $\lambda_r = \lambda/R$ and concentration of homogeneous spherical particles with refractive index n_0 = 1.1 is presented in Fig.1. As seen from the figure, the spectral dependence has a maximum in the range λ_r = 3 – 5 which shifts to small wavelengths with increase of particles concentration. The magnitude of maximum increases as well.

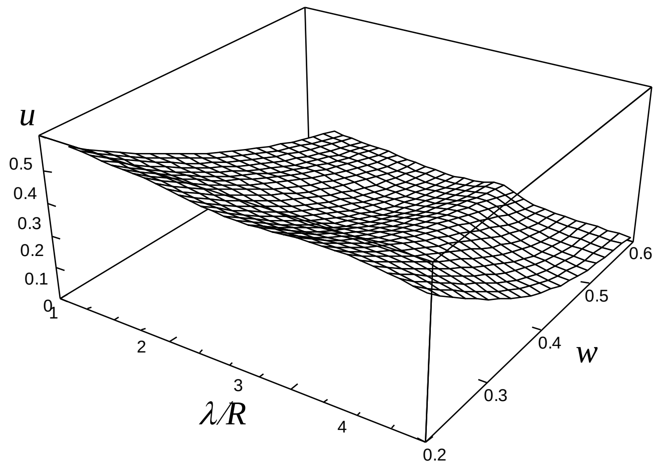

Figure 1. Dependence of $u(\lambda/R, w)$ for homogeneous particles. Radius of particle *R*=0.3 μm, relative refractive index of particle n_0 = 1.1.

The intensity of the forward-scattered light decreases with increasing volume concentration *w*. In the case of large concentrations of particles, the phase function has a characteristic maximum at non-zero scattering angles. The position of such a maximum is shifted to larger angles with increasing volume concentration *w* and to smaller angles with increasing size of particles. At some values of particle concentration the asymmetry parameter $g = \int_{-1}^{1} p(\mu)\mu d\mu$ can be less or equal to zero.

For a medium containing two-layered particles, the dependence of $u(\lambda/R, w)$ differs essentially from that displayed above for ensembles of homogeneous particles. Values of parameter *u* can be more than 1 at certain ratios of refractive indices of core and shell even at small volume concentration. They attain magnitude 2.5 (see Fig. 2) at volume concentration *w* = 0.6. Simulations reveal that the asymmetry parameter attains essentially smaller negative values, than in the case of homogeneous particles.

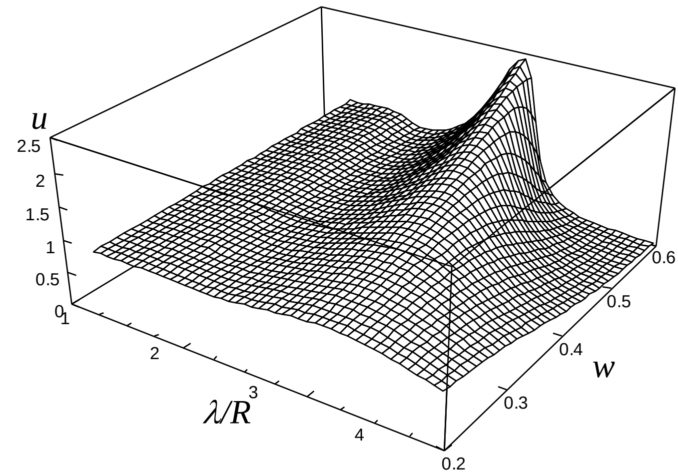

Figure 2. Dependence of $u(\lambda/R, w)$ for ensemble of two-layered particles. Radius of core Rc = 0.18 μm, refractive index of core n_c = 1.10843, radius of shell R = 0.3 μm, refractive index of shell n=0.96364.

CONCLUSION

Light scattering from a layer with closely packed homogeneous and two-layered spherical, non-absorbing particles has been investigated. To model light scattering, the interference approximation and algorithms for light scattering by homogeneous and two-layered particles were used. Simulations disclosed that in a layer of two-layered particles the darkening effect (the optical thickness increasing with the volume concentration) can be implemented. This effect is considered in the details. The developed model can be used to describe the light scattering in liquated glasses, porous glasses and structures, etc.

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