

The electromagnetic radiation scattering in the assembly of the two scales spatially ordered cylinders

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Based on the interference approximation of the statistical theory of multiple wave scattering for planar systems characterized by two ordering scales, a model is proposed for calculating the coefficients of coherent transmission of T_{coh} , reflection R_{coh} and the angular distribution of the intensity of scattered radiation $I(\theta)$. The components of such systems are multimers, integral and angular characteristics, which can be calculated by the method of formalism of the volume integral equation. The radial distribution function of $g_m(r)$ multimers depends on their surface concentration η_m .

It was found that dense packing of multimers results in near-orderedness, which amplification electrodynamic coupling between the multimers and manifests itself in a nonmonotonic concentration dependence of the transmission coefficient.

Keywords: finite cylinders, multimers, electrodynamic coupling, two scales spatially ordered environment, surface concentration.

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Introduction

One of the most important tasks of modern photonics is the creation of active nanoelements for multifunctional devices in optics, laser physics and microelectronics. In recent years, the problem associated with monitoring and controlling the optical properties of nanomaterials by changing their internal structure has been intensively studied [1–3]. Research is actively developing in the field of creating metamaterials [4–6], which currently include both plasmonic metal-containing and optically resonance dielectric nanosystems, the unique electromagnetic properties of which are due to an artificially created spatially-ordered structure. Various types of so-called photonic crystals (PC) have been proposed, in which light can propagate only in given directions or can generally be localized in certain spatial areas [7–9].

To date, the patterns of propagation, scattering and localization of electromagnetic waves of the optical range in one-dimensional, two-dimensional, three-dimensional photonic crystals, which are strictly periodic structures, have been studied in sufficient detail [10,11]. At the same time, in order to identify new opportunities for controlling the spectral and angular characteristics of electromagnetic radiation, an urgent task remains the study of its interaction with partially ordered systems of scatters, in which the characteristic scales of spatial structuring are commensurable with the wavelength of electromagnetic radiation. The presence of this kind of ordering leads to the manifestation

of effects of collective electrodynamic interaction in the system, depending both on the type of spatial ordering of particles in the dispersed structure and on the characteristics of single scattering on these particles [12–14].

For the present, the effects of electrodynamic interaction in structured partially ordered dielectric systems, as well as their influence on the spectral and angular characteristics of such systems, have been theoretically studied mainly for ensembles consisting of spherical particles or infinite cylinders [15–17]. At the same time, structures based on oriented dielectric cylinders of finite length, the characteristic dimensions of which are commensurate with the wavelength of impinging electromagnetic radiation, are of particular interest. Such structures with specified morphological parameters can be formed, for example, by electrochemical anodic process of thin aluminum films [18]. The physicochemical, structural and optical properties of thin porous anodic aluminum oxide films (AAOP) make them a promising material for various applications. AAOP membranes are used as substrates for the formation of composite materials and xerogels, templates for the synthesis of nanostructures. The practical significance of AAOP membranes doped with nanoparticles and dyes is determined by the opportunity of implementing photoluminescent screens and selective filter elements with tunable spectral and angular characteristics.

Additional opportunities for controlling collective effects arise in structures with two characteristic scales of heterogeneities (nanometer and micron sizes). The so-called two-scale spatially-ordered media (TSSOM) are porous and/or

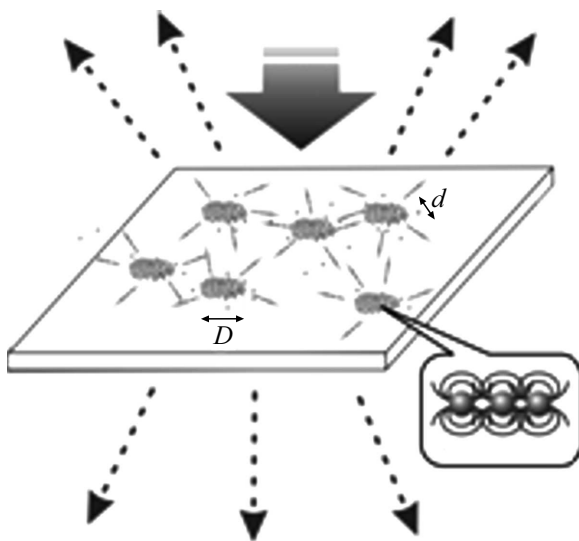


Figure 1. Schematic representation of a two-scale planar structure.

composite materials containing stable structural elements (SSE) in the form of small-particle ensembles formed by a group of smaller particles [19–21]. In such structures, the first ordering scale describes the arrangement of small particles inside the SSE, and the second ordering scale describes the location of the SSE relative to each other (Fig. 1).

This paper proposes a theoretical model for studying the features of the interaction of electromagnetic waves in the optical and microwave range with two-scale dispersed systems. Using the example of spatially-ordered planar structures of hexagonal multimers formed by finite cylinders, the influence of optical-geometric parameters on the transmittance and scattering characteristics of these systems is assessed.

The paper is aimed at identifying the opportunities of controlling the spectral and angular characteristics of scattered radiation through the use of collective electrodynamic interactions in ordered systems of cylinders of finite length. These studies are important from a practical point of view for establishing additional capabilities for controlling the scattering and localization of electromagnetic radiation in the optical and microwave ranges, which is required in the development of new materials and functional elements for laser physics, optoelectronics, microwave technology and radiophysics.

Calculation method

Let us divide the procedure for simulation of the optical properties of planar TSSOM into two intervals: (1) simulation of the optical characteristics of the SSE and (2) simulation of the optical characteristics of the TSSOM as a whole.

Simulation of SSE optical characteristics

At the first interval of simulation, the simplest approach is to consider stable structural elements consisting of „primary“ particles as homogeneous disks with a size of D and an effective complex refractive index depending on the concentration of nanoscale inclusions in this disk. The effective complex refractive index of $n_{D(\text{app})}$ disks can be determined, for example, from Maxwell–Garnett relations or their modifications applicable over a wider range of nanoinclusion concentrations. Parameter D is taken equal to the characteristic size of the SSE. Then, using admissible approximate relations [17,22] or the volume integral equation formalism (VIEF) [22–25] it is possible to calculate the attenuation efficiency factors $Q_{D(\text{ext})}$, absorption $Q_{D(\text{abs})}$, scattering $Q_{D(\text{sca})}$ and amplitude scattering functions $S(\theta)_D$ of such model disks .

The entity of the volume integral equation description (VIEF) is as follows. The homogeneous or heterogeneous particle under study is divided into elementary cubic cells of the same size. The refractive index of these cells is compared to the corresponding material. It is assumed that each point in space at which the refractive index is not equal to unity emits as a dipole, with corresponding phase and amplitude characteristics. These characteristics are determined by the local field and the refractive index.

The main integral equation of the VIEF has the form

$$E(r_1) = E_{\text{in}}(r_1) + k^2/(4\pi) \times \iiint [m^2(r_2) - 1]E(r_2)G(r_1, r_2)d^3r_2,$$

where E — electric field strength at the point under consideration, E_{in} — electric field strength of the impinging wave, m — complex refractive index of the particle, k — wave vector, $G(r_1, r_2)$ — tensor Green’s function.

When the scatter is divided into N cubic shape lattice cells, the integral equation is reduced to a matrix equation, which is defined in the $3N$ -dimensional space of complex numbers:

$$\mathbf{A}\mathbf{E}_v = \mathbf{E}_v^{\text{in}},$$

where \mathbf{A} — a square matrix of complex numbers taking into account the interactions of lattice cells, \mathbf{E}_v — a column vector containing the values of the desired field in each cell, \mathbf{E}_v^{in} — a column vector containing the values of the incident field in the cells. Next, the matrix equation is solved using numerical methods by minimizing the function $|\mathbf{A}\mathbf{E}_v - \mathbf{E}_v^{\text{in}}|^2$. Reducing the volume of lattice cells increases the accuracy of calculating the characteristics of the scatter.

The attenuation efficiency factor Q of the scatter under consideration is determined as the ratio of the attenuation cross section C_{ext} to the geometric cross section of the scatter in a plane perpendicular to the direction of incidence of the electromagnetic wave. It can be found from the attenuation formula $C_{\text{ext}} = 4\pi\text{Re}S(0)/k^2$, where $S(0)$ — amplitude scattering function in the forward direction, k — wave number.

The calculation of the scattering indicatrices $x(\theta)$ and from the amplitude scattering functions $S_1(\theta)$, $S_2(\theta)$, corresponding to the orthogonal polarizations of the incident light, is carried out using the following relations:

$$x(\theta) = |S(\theta)|^2 / \pi \rho^2 Q_{\text{sca}},$$

$$i(\theta) = |S(\theta)|^2 = 1/2(|S_1(\theta)|^2 + |S_2(\theta)|^2),$$

where $S(\theta)$, Q_{sca} and diffraction parameter ρ correspond to the object under consideration, i.e. in our case, a separate SSE.

The procedure for transition from integration over the entire particle to summation assumes that the particle is represented as an ensemble of lattice cells of the same cubic shape. The electric field within each unit cell is assumed to be constant, and the refraction index within each lattice cell is set according to the composition and structure of the scatter in question. If the effective medium approximation is used to model the optical characteristics of individual SSE, then the scatter under consideration is homogeneous, its dimensions coincide with the characteristic dimensions of the SSE, and the refraction index inside each lattice cell is equal to the effective complex refraction index.

The applicability of the effective medium approximation does not allow, however, to provide a sufficiently strict account of collective interactions inside the SSE filled with „primary“ particles. The most significant difficulties arise in the case of non-spherical „primary“ particles with the same orientation, for example, oriented cylinders. Therefore, it is better to take into account the strong electrodynamic interactions inside the SSE, which is a multimer of a certain type, by directly applying the VIEF to such a multimer, considered as a single heterogeneous scattering formation.

As an example, in this paper we will review SSE, which are multimers of hexagonal symmetry, which consist of oriented cylinders of finite length. The hexagonal multimers we are reviewing consist of seven coaxial cylinders, one of which is located in the center of a circle with a radius of R , and the centers of the other six are uniformly located on this circle. A schematic representation of such a multimer is shown in Fig. 2. Hexagonal multimers of this type reproduce the structure of the primary cells of anodized aluminum films (honeycomb patterns of cylindrical pores). To calculate the optical characteristics of such hexagonal multimers, we used a special software package VIEF–M [24,25].

Simulation of TSSOM optical characteristics

At the second interval, when modeling the optical characteristics of the TSSOM as a whole, the development of the model is carried out by applying the statistical theory of multiple wave scattering (STMWS) to a partially ordered ensemble of SSE. In STMWS, the resulting field is represented as the sum of the fields of all possible multiply scattered waves, taking into account their phase, and each

heterogeneity is not in the field of the incident wave, but in some effective field. STMWS is one of the most effective approaches to simulation and analyzing the propagation and scattering of coherent radiation in partially ordered media, since this approach allows to take into account electrodynamic interactions between scattering objects [26–29]. The need to take into account electrodynamic interactions that arise as a result of coherent re-irradiation of heterogeneities with each other depends on the degree of correlation of the spatial location of these heterogeneities. Typically, in close-packed discrete systems, the correlation of the location of heterogeneities is described by the binary correlation distribution function $g(\mathbf{r}_1, \mathbf{r}_2)$, the form of which is determined by the size and surface concentration of heterogeneities [30].

To date, the most rigorous STMWS method for taking into account cooperative effects in partially ordered ensembles of particles is the so-called quasicrystalline approximation (QCA). In those cases where coherent re-irradiation between heterogeneities is insignificant, the so-called single coherent scattering approximation (SCSA), or interference approximation (IA), gives fairly good results. In this approximation, it is assumed that each particle is located only in the field of the incident wave [26–29], and the collective interaction consists of the interference of waves scattered once by each particle. The contribution of the effects of coherent re-irradiation by heterogeneities to each other decreases with a decrease in their surface concentration and an increase in the distance between them. Comparison of the results of calculations using IA and using QCA for monolayers of spherical heterogeneities [29] shows that calculations in these two approximations agree quite well at $\eta < 0.3$. In addition, it is important to note that the concentration range of applicability of the IA expands with increasing degree of elongation of single scattering indicatrices in the forward direction.

Applicability of the interference approximation for monolayers formed by oriented cylinders of finite length

In the interference approximation, the following expressions are used to calculate the coefficients of coherent transmittance, coherent reflection and intensity of scattered light:

$$T_{\text{coh}} = 1 - \eta Q_{\text{ext}} + \eta^2 Q_{\text{ext}} \frac{4\pi x(0)\Lambda}{\rho^2},$$

$$R_{\text{coh}} = \eta^2 Q_{\text{ext}} \frac{4\pi x(\pi)\Lambda}{\rho^2},$$

$$I(\theta, \phi) = F_0 \lambda Q_{\text{ext}} \eta x(\theta, \phi) H(\theta, \phi),$$

$$H(\theta, \phi) = 1 + N \int_V \int_V [g(\mathbf{r}_i, \mathbf{r}_j) - 1] \times \exp[ik(\mathbf{r}_i - \mathbf{r}_j)(\mathbf{s} - \mathbf{s}_0)] \frac{d\mathbf{r}_i d\mathbf{r}_j}{V}, \quad (1)$$

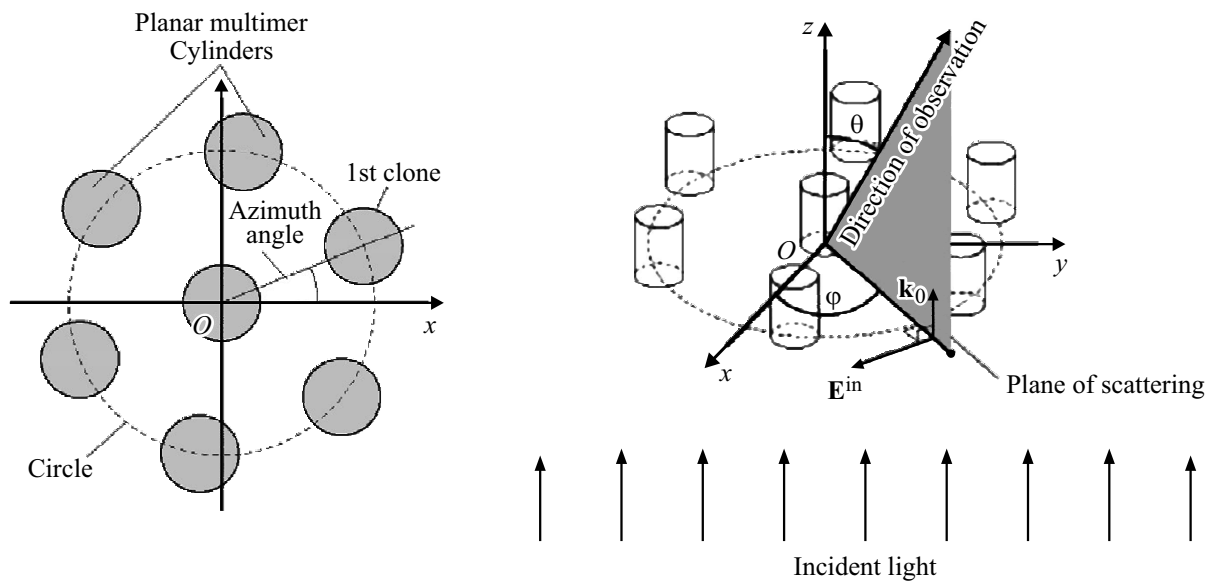


Figure 2. Schematic representation of a hexagonal multimer made of identical cylinders.

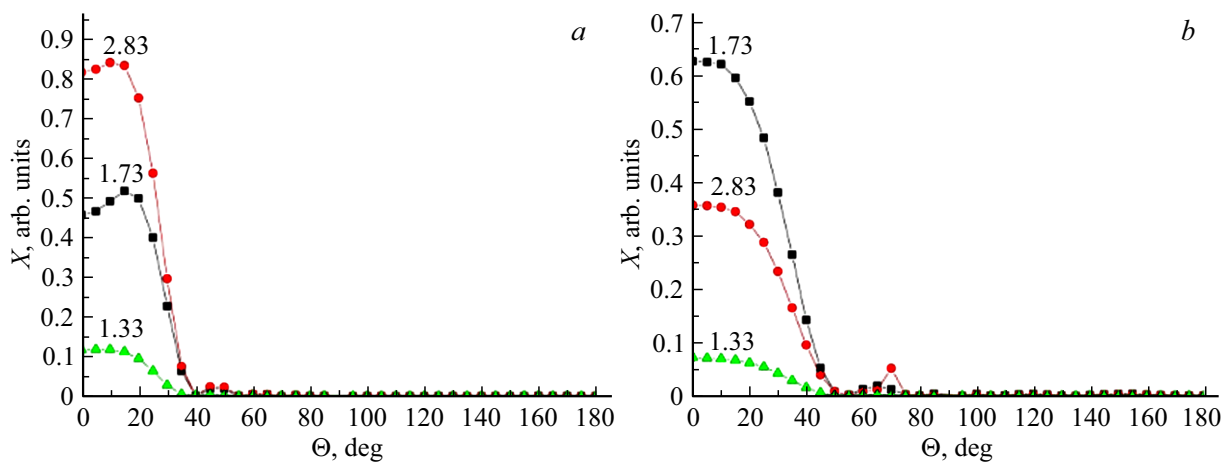


Figure 3. Scattering indicatrices of cylindrical particles with dimensions $d = 100$ nm, $l = 1500$ nm and refraction index 1.33, 1.73, 2.83; wavelength of incident radiation 300 (a) and 600 nm (b); the incident radiation wave propagates along the OZ axis.

where $\Lambda = Q_{sca}/Q_{ext}$ — survival parameter, Q_{sca} and Q_{ext} — scattering and particle attenuation efficiency factors, respectively, η — overlap parameter proportional to the surface concentration of particles, $\rho = \pi d/\lambda$ — diffraction parameter; d — particle diameter, $x(\theta)$ — particle scattering indicatrix, value F_0 — power of incident radiation, $H(\theta, \phi)$ — structure factor depending on the binary correlation function $g(\mathbf{r}_1, \mathbf{r}_2)$, N — number of particles, (θ, ϕ) — angle between incident and scattered beam.

Taking into account the interference of waves scattered once by each particle in the interference approximation leads, in particular, to the appearance of a third term quadratic in the number of particles in the formula for the coherent transmittance coefficient. The contribution of the third term in formula (1) is determined by the intensity of radiation scattering in the forward direction and the degree of elongation of the scattering indicatrix

of an individual particle. These characteristics depend on the optical and geometric parameters of the particle. For cylindrical particles, such studies were carried out in the papers [24,25]. An increase in the intensity of scattering in the forward direction was found with a decrease in the wavelength of the incident radiation and with an increase in the diffraction parameter due to an increase in the particle size or the refractive index of the surrounding medium. In addition, it is shown that the transition from the direct system to the inverse system leads to a significant increase in the degree of elongation of the IR of the final cylinder in the direction of propagation of the incident radiation and an increase in the scattering intensity in the forward direction.

As an example, Fig. 3 shows the effect of the relative refractive index of the particle and the wavelength of the incident radiation.

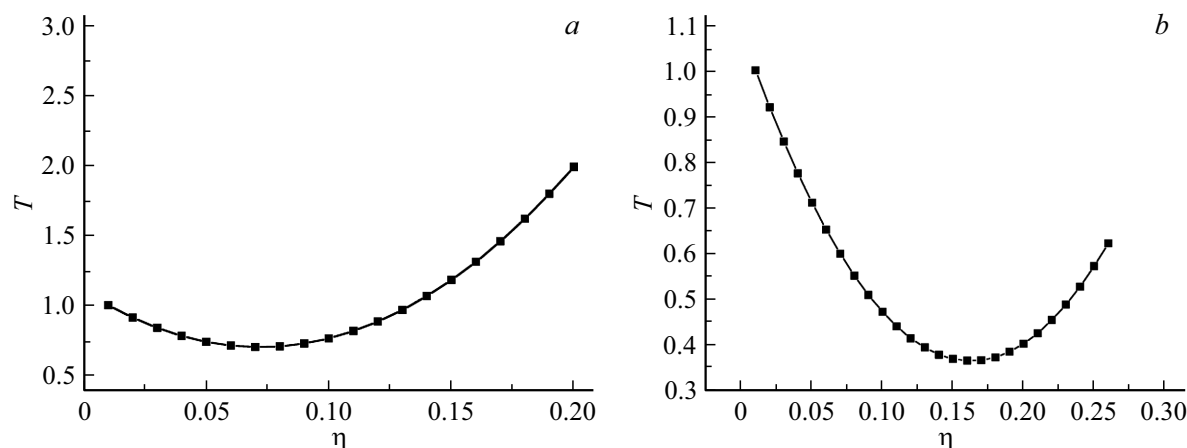


Figure 4. Dependence of the transmittance of a cylindrical particles monolayer on the overlap parameter (a) — cylinder diameter 100 nm, relative refractive index 1.33; (b) cylinder diameter 200 nm, relative refractive index 1.73.

Figure 4 shows an example of calculation in the IA of the transmittance of a monolayer formed by individual (not combined into multimers) oriented cylinders located perpendicular to the surface of the monolayer. The length of the cylinders is 1500 nm, the wavelength of the incident radiation is 300 nm.

The dependence of the transmittance of a cylindrical particles monolayer on the overlap parameter calculated in this approximation is non-monotonic. This nature of the concentration dependence reflects the fact of competition between two processes that take place with an increase in the number of particles in the monolayer — a decrease in free space and an increase in the intensity of radiation coherently scattered in the forward direction. In the area of low surface concentrations, with an increase in the number of particles in the monolayer, the predominant influence is exerted by a decrease in free space, which leads to a decrease in the transmittance. In the area of high surface concentrations, when the correlation in the arrangement of particles increases, the processes of interference of radiation coherently scattered in the forward direction become significant. In this concentration area, as the number of particles in the monolayer increases, the contribution of interference effects increases and the transmittance increases. The minimum transmittance is achieved at values of h that depend on the scattering and absorption characteristics of individual particles. For example, as can be seen from the comparison of Fig. 3, *a* and 3, *b*, a decrease in the diameter and refractive index of the cylinder leads to a narrowing of the area of surface concentrations, within which the concentration decrease of the transmittance coefficient is realized.

However, it is clear from the figure that for small values of the refractive index ($n = 1.33$) already in the area of overlap parameters $\eta > 0.15$ the transmittance values exceed unity and become nonphysical. For larger refractive index values ($n = 1.73$) the $T < 1$ area expands to $\eta = 0.3$. This fact is due to the fact that as the relative refractive index

of a particle decreases, the scattering indicatrix becomes more diffuse, which leads to more significant errors when the coherent over-radiation of particles in a monolayer is not taken into account. The area of applicability of the interference approximation can be expanded by taking coherent over-radiation in the immediate environment of particles into account at a preliminary stage. In the case of two-scale systems, it is first necessary to take into account the electrodynamic interactions inside the SSE.

Modified interference approximation

A feature of spatially-ordered two-scale systems is that the distances between the primary particles that make up the SSE are smaller than between individual SSE. Accordingly, the packing density of the primary particles forming the SSE is higher than the packing density of the SSE. In addition, an increase in the diffraction parameter of the SSE compared to the diffraction parameter of the primary particles leads to an increase in the degree of elongation of the scattering indicatrix of the SSE in the forward direction. This circumstance helps to reduce coherent over-radiation between SSE. Therefore, the most significant role in two-scale structures is played by coherent over-radiation between the particles forming the SSE. As already mentioned, the most effective way to take into account coherent over-radiation inside SSE, which are a multimer of a small number of cylinders of finite length, is to use a VIEF. Coherent re-irradiation between individual SSE is less significant and in a large number of practically important cases it can be neglected.

Thus, to evaluate the optical characteristics of a two-dimensional ensemble of multimers in the region of surface concentrations of $\eta_m < 0.3$ multimers, the following approximate expressions can be proposed:

$$T_m = 1 - \eta_m Q_m + \eta_m^2 Q_m \frac{4\pi x_m(0) \Lambda_m}{\rho_m^2},$$

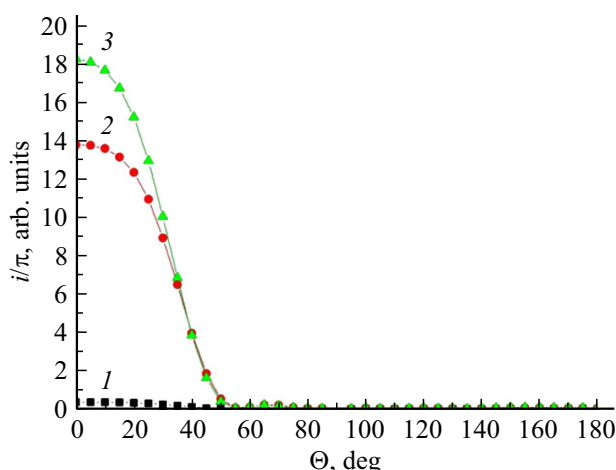


Figure 5. Normalized functions of the angular distribution of the scattering intensity of an individual cylinder (1) and hexagonal multimers with radii 100 (2) and 140 nm (3). The relative refraction index of the cylinders is 2.83, the wavelength of the incident radiation is 600 nm, the length and diameter of each cylinder are 1500 nm and 100 nm, respectively.

$$R_m = \eta_m^2 Q_m \frac{4\pi x_m(\pi) \Lambda_m}{\rho_m^2}, \tag{2}$$

$$I(\theta, \varphi) = F_0 \Lambda_m Q_m \eta_m x_m(\theta, \varphi) H_m(\theta, \varphi),$$

$$H_{m(\theta, \varphi)} = 1 + N_m \int_V \int_V [g_m(\mathbf{r}_i, \mathbf{r}_j) - 1] \times \exp[ik(\mathbf{r}_i - \mathbf{r}_j)(s - s_0)] \frac{d\mathbf{r}_i d\mathbf{r}_j}{V},$$

where η_m — overlap parameter, proportional to the surface concentration of multimers; $Q_m, \Lambda_m, x_m(\theta)$ — attenuation efficiency factor, survival parameter and multimer scattering indicatrix, respectively, $H_m(\theta, \varphi)$ — structure factor describing the arrangement of multimers, $g_m(\mathbf{r}_1, \mathbf{r}_2)$ — binary correlation function of multimer distribution, N_m — number multimers.

The relationship between the overlap parameters of hexagonal multimers η_m and primary particles η_c is given by the expressions:

$$\eta_m = \frac{N_m \frac{\pi D_m^2}{4}}{S} = (N_0)_m \frac{\pi D_m^2}{4},$$

$$\eta_c = (N_0)_c \frac{\pi d^2}{4} = (7N_0)_m \frac{\pi d^2}{4} = \frac{7}{4} \pi d \frac{4\eta_m}{\pi D_m^2} = \frac{7d^2}{D_m^2} \eta_m, \tag{3}$$

$$\eta_c = \frac{7d^2}{D_m^2} \eta_m.$$

Here $D_m = 2R + d$ — characteristic transverse size of a multimer, $(N_0)_c$ and $(N_0)_m$ — surface density of primary particles and multimers, respectively.

For statistically homogeneous ensembles of partially ordered multimers, the two-dimensional correlation function

depends only on the distance between the multimers, which allows to introduce the radial distribution function $g_m(r)$. The radial distribution function $g_m(r)$ can be determined either in the approximation of solid incompressible disks with a size equal to the size of the multimer, or using suitable approximation expressions [30]. The absorbing and scattering properties of multimers are calculated, as already mentioned, using the VIEF method.

As an example, Fig. 5 shows the results of calculating the normalized functions of the angular distribution of the scattering intensity of an individual cylinder and hexagonal multimers.

As can be seen from Fig. 5, with an increase in the transverse diffraction parameter of the heterogeneity due to an increase in its size, i.e. when passing from a single cylinder to an ensemble of cylinders, an elongation of the scattering indicatrix in the forward direction is observed. The values of the angular distribution function of the scattering intensity for multimers increase by an order of magnitude, and the main maximum also shifts to the area of small angles. Similar conclusions can be drawn from the results of calculations for inverse hexagonal multimers, which are air pores in a dielectric matrix.

Figure 6 shows calculations of the transmittance of a monolayer consisting of multimers of cylindrical particles using formula (2). Multimers consist of cylinders with a diameter of 200 nm, a length of 1500 nm and a refraction index of 1.73. The radius of multimers is 400 nm, wavelength is 300 nm.

Figure 6 shows a monotonic decrease in transmittance as the surface concentration of multimers increases. This kind of concentration dependence of the transmittance indicates that with an increase in the number of multimers in a monolayer, in this case, the effect of decreasing free space is predominant compared to the effect of increasing the

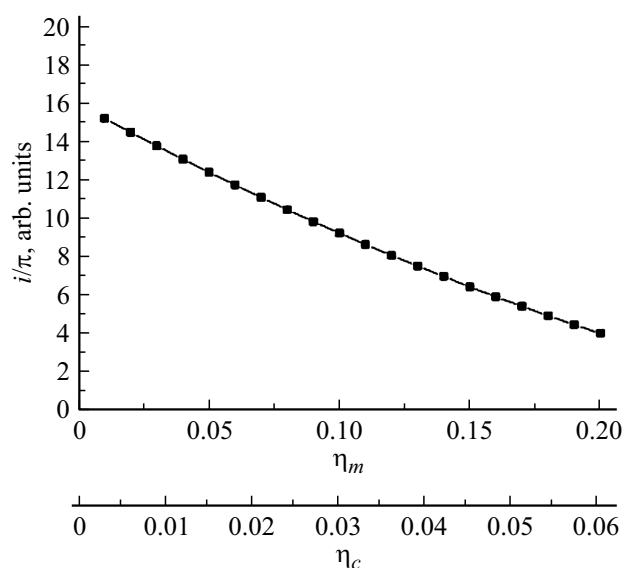


Figure 6. Concentration dependence of the transmittance of a monolayer consisting of multimers of cylindrical particles.

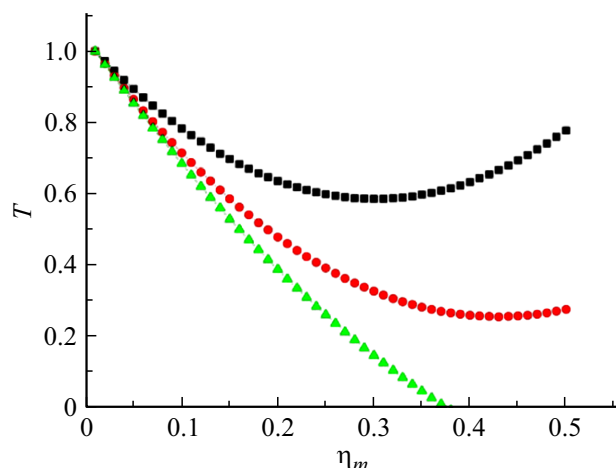


Figure 7. Dependence of the transmittance of a monolayer consisting of multimers on the overlap parameter. Relative refraction index of cylinders $n = 1.73$; $R = 300, 350, 400$ nm.

intensity of radiation coherently scattered by multimers in the forward direction. This relationship between two competing concentration processes is typical for „optically hard“ multimers.

The size effect of multimers on the concentration dependence of transmittance is illustrated in Fig. 7. This figure shows the results of calculations for monolayers of multimers of different sizes formed by cylinders with a diameter of 100 nm and a refraction index of 1.73. The wavelength is 300 nm.

As can be seen from the figure, throughout the entire considered range of surface concentrations of multimers, the transmittance varies from 0 to 1.

As the diameter of the multimer increases, the qualitative nature of the dependence of the transmittance on the overlap parameter changes. The non-monotonic dependence of the transmittance observed for small radii of multimers is replaced by a monotonically decreasing dependence as the multimer size increases.

Conclusion

Thus, based on the joint use of the interference approximation STMWS and the formalism of the volumetric integral equation, a method is proposed for calculating the transmittance and scattering characteristics of monolayers consisting of multimers of cylindrical particles of finite length. In the proposed method, in addition to taking into account the interference of waves scattered by different particles in the same direction, an approximate account is taken of coherent over-radiation by particles included in the immediate environment of the scattering center under consideration. The proposed model is based on a two-stage consideration of electrodynamic interactions in a two-scale discrete monolayer and allows to analyze the optical

characteristics of a monolayer under conditions of dense packing of its constituent heterogeneities.

Conflict of interest

The authors declare that they have no conflict of interest.

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