Spectra of light reflected by aggregate structures of submicron particles

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Abstract

We analyze the results of numerically exact computer modeling of spectral dependences of the intensity of light scattered by different aggregates of submicron spherical particles in the visible range and compare them to those of individual homogeneous spheres. The computations are performed using the Lorenz–Mie theory and the superposition T-matrix methods for aggregates, respectively. We show that, in the spectra of aggregates, along with the interference extrema peculiar of individual constituents, an additional maximum appears at longer wavelengths. The latter is considered as manifestation of collective effects in aggregate structures and can be explained by the interference of waves singly scattered by aggregate constituents that form groups along the incident radiation direction. In the spectra of randomly oriented fractal-like aggregates, this maximum becomes narrower with increasing the number of constituents and, starting from some number, its position becomes almost resistant to the further growth of aggregates and sensitive only to the refractive index and sizes of constituents. The number of constituents providing a stable collective maximum is higher for fluffier structures. In comparison with rather densely packed clusters, sparser ones exhibit the less expressed collective maximum with a slope weakly declining to the long-wavelength range. In the spectra of clusters containing particles with slightly varied sizes, the collective maximum survives, while the interference features induced by individual constituents naturally become smoother.

Keywords: light scattering, spectrum, aggregate, MSTM modeling

Highlights:

- Spectra of light reflected by fractal clusters of submicron particles are analyzed
- Interference of waves singly scattered by constituents induces new collective details
- Collective maximum is resistant to increasing the constituent number after some value
- Number of constituents providing a stable collective maximum depends of the structure
- Collective maximum is rather weakly affected by variations in the constituent sizes

1. Introduction

Characteristics of the electromagnetic radiation scattered by aggregates of submicron particles are in the focus of many studies in different application, from astrophysics to biology and from materials science to medicine [1-4]. It is often impossible to directly measure the properties of these aggregates (or clusters) of particles, and one have to conclude about their sizes, morphology, and material from the measurements of intensity and polarization of the scattered light. Results of spectral measurements also serve as a source of information on the properties of examined media. The spectral dependences are important, for example, for interpretation of observations of cometary dust or different smokes, the particles of which are aggregates of different morphology. At the same time, when interpreting the spectra, contrary to, for example, polarimetry, the structure of particles is often ignored.

However, recent studies, both theoretical and experimental, show that morphology of particles substantially influences their transmittance/absorption spectra [5–8]. In these papers, the main attention is payed to the IR spectral range and integral characteristics, such as the extinction and scattering cross-sections. The reflectance spectra of different systems of scatterers are considered in many papers (e.g., [9-11]), but they mostly deal with infinite media or layers of particles, while the scattering ensembles comparable to the wavelength in size remain beyond the reflectance spectrum analysis.

It is known that, if the sizes of particles composing aggregates are of the order of the wavelength or somewhat smaller, the aggregate morphology produces a noticeable effect on the angular dependence (the phase function) of intensity and polarization of the scattered light in the backscattering domain (see, e.g., [12–16]). This is caused by strengthening the collective effects in the scattering by ensembles of particles of these sizes. Thus, the spectrum of light scattered by a medium composed of aggregates should differ from that of homogeneous particles and depend on morphology of these aggregates, which makes the heterogeneity of scatterers to be necessarily accounted for in the interpretation of their spectra.

In this paper, we consider manifestations of collective effects in limited aggregate structures of submicron particles observed in the visible spectrum of intensity of the scattered radiation. The intensity depends on both the value of the phase function of particles at a specified scattering angle and their scattering cross-section. The spectral dependence of the latter is caused by the dependence of the scattering efficiency of particles Q_{sca} on their refractive index and the sizes relative to the wavelength, i.e., the size (wave) parameter $x = 2\pi a/\lambda$ (where λ is the wavelength, and a is the particle radius). The analysis of this dependence for homogeneous

particles are well-represented in the literature (see, e.g., [12]). The sizes of particles considered here are smaller than or comparable to the visible wavelengths, which corresponds to the segment of the Q_{sca} curve gradually growing with increasing x and exhibiting a gently sloping hump. Thus, the details in the spectrum of the intensity of radiation scattered by particles of the considered sizes should be caused mainly by the spectral behavior of the phase function of particles. Consequently, it is the changes in the spectra of the phase function under the influence of the structure of aggregates, the sizes of constituents, and the refractive index that is a subject of the present analysis.

We focus here on the spectral behavior of the phase function mainly in the backscattering direction $F(\pi)$ (the scattering angle is $\theta = \pi$), because this behavior provides insight into the spectral dependence of the intensity of radiation scattered to the back hemisphere in a rather wide range of angles. This is explained by the fact that the phase functions of clusters (as those of many irregular particles) are rather smooth in the backscattering domain and exhibit no pronounced features like rainbows and glories characteristic of scattering by individual spherical and other regular particles. The near-field effects, which appear in the light scattering on aggregates of particles, induce no sharp changes in the shape of the phase curve, while the coherent backscattering on clusters of the considered sizes induces no sharp enhancement in the intensity near opposition ($\theta = \pi$) (see, e.g., [14, 17]). (This statement will be illustrated below in Fig. 5.)

The structure of the paper is as follows. First, we describe the parameters that are used in the model calculations. Then, to find manifestations of collective effects in the spectra of aggregates, the spectral dependences of the phase functions of single homogeneous spherical particles and their aggregates are compared. The development of the collective feature identified in the spectra of aggregates is analyzed with the use of definitely orientated structures containing two and three spherical particles, and the changes in the spectra of fractal-like aggregates induced by the increase in the number of constituent particles (CPs) are considered. The results of the analysis are summarized at the end.

2. Input parameters for the simulations

The choice of values for the refractive index and sizes of CPs in the aggregates primarily stems from the purposes of astrophysical applications. The main attention will be payed to the spectral dependences of $F(\pi)$ for ice particles (the refractive index is $m = 1.33 + 0.0i$). To illustrate the influence of changes in the refractive index and absorption, we also consider the so-called dirty ice (the imaginary part of the refractive index was assumed at $m_i = 0.05$) and the artificial material with the real part of the refractive index $m_r = 1.80$ (which approximately

corresponds to the mean value of m_r of Mg-Fe silicates and is close to that of tholins) and m_i = 0.10. We intentionally do not include the spectral dependence of the refractive index into our analysis, because we mean to reveal a pure effect of the refractive index on the spectral pattern of aggregates. At the same time, the model results for the considered values of m may be useful for predicting at least a qualitative behavior of the spectrum for intermediate values of the refractive index.

The values for the CP sizes were chosen in such a way that the size parameters of were roughly between 1.0 and 2.0 for the visible spectral range. This interval of x corresponds to the estimates of x for CPs in the clusters composing cometary dust and regoliths, which were obtained from modeling the negative branch of polarization permanently observed in comets and atmosphereless bodies (see, e.g., [18-20]). Though we actually calculated the light-scattering models for several cases of the CP sizes, we present here the results only for the CP radius a = 0.115 μm, which corresponds to x = 1.2 at λ = 0.60 μm. Obviously, the change in the CP size does not induce essential changes in the spectral behavior of $F(\pi)$ and only shifts the whole pattern along the spectrum (see below).

The shape of particles composing aggregates is assumed to be spherical, since this allows us to distinguish easier the manifestations of properties of individual particles in the scattering characteristics of complex aggregates. This assumption is supported by the results of calculations [20-22] and laboratory measurements [23] of the scattering characteristics of aggregates composed of particles irregular in shape. They show that these characteristics are close to those of aggregates of spherical particles when the CP size parameters are rather small, $x < 2$. This fact is explained by blurring out the peculiarities in the scattering by individual spherical particles of these sizes by near-field effects if they are close to each other in the aggregate.

To understand the effect of the structure of clusters on their spectral pattern, we consider relatively densely packed fractal-like clusters with the following fractal parameters: the pre-factor k_f = 8 and the fractal dimensions D_f = 3, 2.5, and 2.1 (in the following, structures A, B, and C) and a much fluffier fractal-like cluster with k_f = 5.8 and D_f = 1.9 (structure D). We define these parameters according to the description given in the paper [24]. The porosity of clusters is determined from the so-called characteristic radius $R_c = (5/3)^{1/2} \times R_g$ [25], where R_g is the gyration radius of a cluster. Then, the porosity is $p = 1 - N \times (a/R_c)^3$, where N is the number of CPs (monomers). For clusters A–D composed of 42 monomers with radii a = 0.115 μm (which corresponds to the volume equivalent sphere with a radius R = 0.4 μm), p = 0.54, 0.67, 0.78, and 0.93, respectively. All these structures are shown below in the inset of Fig. 1.

Though the structure of the clusters with assumed fractal parameters weakly but depends

on realization peculiarities and, consequently, the characteristics of the scattered light are also realization-dependent [14], we consider here the spectra only for one of the structure realizations. The reason is that we are interested here only in the qualitative behavior of the spectrum in dependence on the properties of the clusters, and we do not average the spectrum over realizations for deriving the quantitative dependence.

3. Spectra of the phase functions of single particles and aggregates: the similarities and differences

At the beginning, let us compare the spectral dependences of $F(\pi)$ for randomly oriented clusters of spherical CPs and individual spherical particles. The phase functions are normalized in the following way: $F = 4a_{11}/Q_{sca}/x_v^2$, where a_{11} is the element of the scattering matrix and $x_v = x$ for a single particle or is defined for an aggregate as the size parameter of the equivalent-volume sphere. The scattering matrix of single homogeneous spherical particles is calculated with the Lorenz–Mie theory (e.g., [12]). To calculate the scattering matrix of aggregates, we use the superposition T-matrix method, which is one of the most versatile and efficient direct computer solvers of the macroscopic Maxwell equations and is widely applied to the computation of electromagnetic scattering by an arbitrary multi-sphere configuration in random or fixed orientation [12]. Consequently, we compute the elements of the single-scattering Stokes matrix for the generated clusters with the corresponding publicly available FORTRAN code (MSTM) [26].

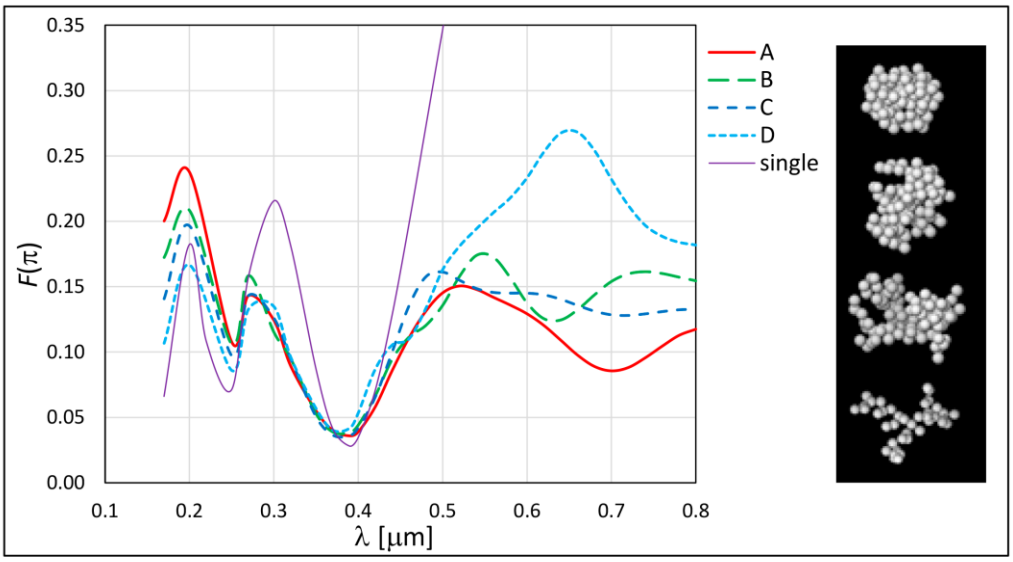

Fig. 1.

Fig. 1. The spectral dependences of $F(\pi)$ for single spherical ice particles with a radius of 0.115 μm and for aggregate structures A–D (shown top-down in the inset) composed of such particles.

Figure 1 shows the spectral dependences of $F(\pi)$ for randomly oriented clusters A–D composed of 42 ice CPs with radii a = 0.115 μm and for single spherical ice particles with the same radius as that of CPs. In the short-wavelength range, all structures exhibit similar patterns corresponding to the features in the spectrum of individual monomers. This means that some features in the spectrum of radiation reflected by individual particles survive in the spectrum of their aggregates. At longer wavelengths, where the scattering by single spheres becomes more isotropic and the $F(\pi)$ curve constantly grows, we observe the formation of additional details in the spectra of aggregates. Obviously, they develop due collective effects induced by scattering on CPs in the aggregates.

To explain the pattern of the spectra of aggregate particles, we will start from the simplest example of scattering by homogeneous spherical particles. The amplitude matrix **S** of the radiation scattered by a homogeneous sphere in the backscattering direction is determined by

$$S_1(\pi) = -S_2(\pi) = \frac{1}{2}\sum_n (2n+1)(-1)^n (a_n - b_n),$$

where a_n and b_n are the Mie coefficients [12]. Note that, though these relationship was obtained for the strictly backscattering direction, it is approximately valid for some angular range around this direction. As is seen from the above formula, for some wavelengths and refractive indices and sizes of particles, the intensity of the reflected radiation may be minimal or even vanish. The latter case results from the equal values of the coefficients a_n and b_n. Consequently, the spectral curves for spherical particles of the same size clearly demonstrate the interference behavior with a characteristic series of minima and maxima, which are well seen in Fig. 2. With increasing the real part of the refractive index m_r, the minima and maxima move to the long-wavelength range of the spectrum. We may say that this shift is caused exactly by changing the real part of the refractive index, since the absorption in the particles produces almost no effect on the position of features in the spectrum of the reflected light and only reduces their amplitude.

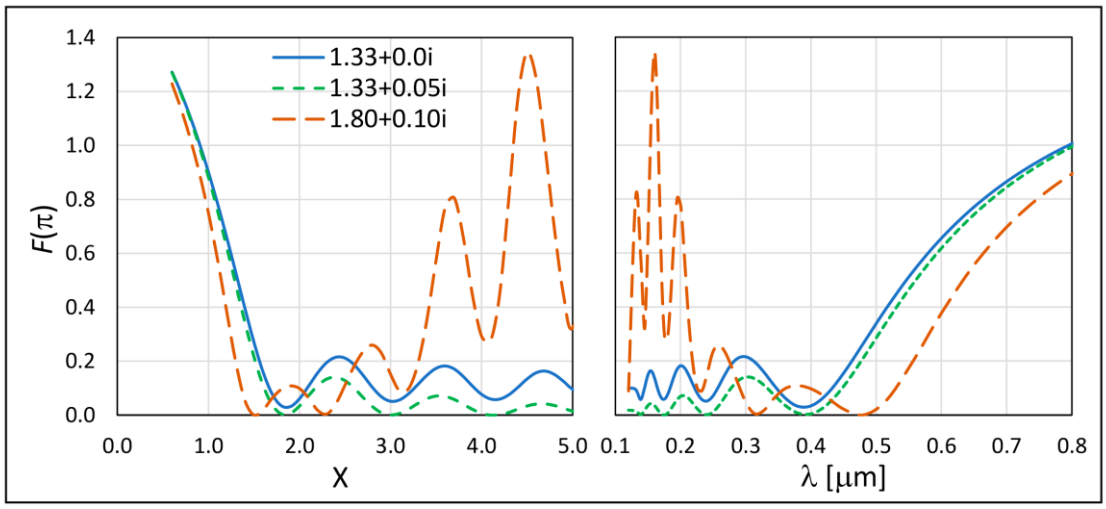

Fig. 2

Fig. 2. The value of $F(\pi)$ for spherical homogeneous particles of the specified materials in dependence on their size parameter (left) and the wavelength (right); the radius of particles is a = 0.115 μm.

Though the geometric optics statements may hardly be applied to the considered size parameters, the estimates of the extrema positions on the spectral curves obtained in the frames of such an approach yield reasonable results. Thus, if we assume that maxima and minima on the curves result from the interference of waves, one of which is reflected from the outer surface of the particle while the second one passes through the particle twice and returns backwards, the maximum and minimum positions x_0 on the curve plotted in dependence on the size parameter will be determined as $x_0 = n\pi/4/m_r$. If n takes odd values, this relationship yields the positions of the minima, while the positions of the maxima correspond to even values of n. If $n \geq 3$, this relationship fits the extrema positions for m_r = 1.33 quite well, while the agreement becomes worse for m_r = 1.80. It is worth noting that this approach may be useful for estimating the sizes or refractive indices of particles from spectral measurements of sparse monodisperse media composed of homogeneous spherical particles (or not strongly differing from spheres) or aggregates of particles of the same sizes.

Though we consider here only the clusters of particles with a = 0.115 μm, the above formula allows one to determine, at least approximately, the positions of minima and maxima in the spectrum for other values of the CP size and refractive index.

Thus, the features observed at short wavelengths in the spectra of aggregates are obviously initiated by the scattering of light on single CPs. To investigate the development of the pattern observed for aggregates at $\lambda > 0.5$ μm, where the scattering on single particles generates no

features, simply organized specific systems of particles turned out to be useful.

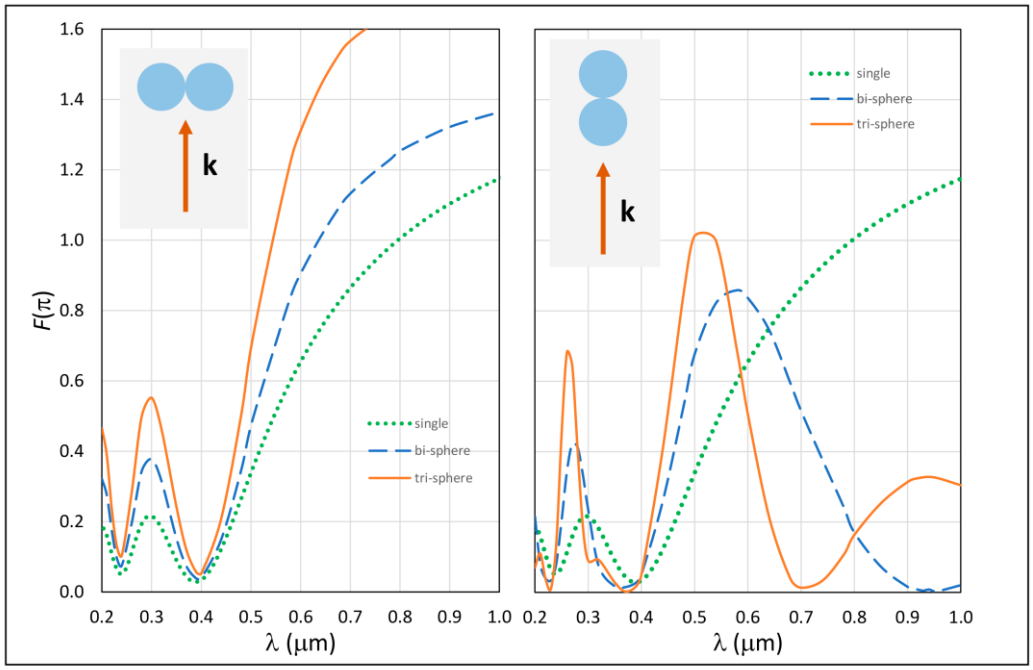

Fig. 3.

Fig. 3. The spectra of $F(\pi)$ for bi- and tri-spheres in contact, the axes of which are oriented perpendicular to (left panel) and along (right panel) the incident radiation direction **k**. The parameters of CPs are a = 0.115 μm and m = 1.33 + 0.0i. The spectral curve for single spherical particles of this properties is shown for comparison.

Figure 3 shows the spectral dependence of $F(\pi)$ for clusters of two and three spherical particles in contact (bi- and tri-spheres), the parameters of constituents of which are a = 0.115 μm and m = 1.33 + 0.0i. Two orientations of the cluster axis relative to the incident radiation direction **k** are considered---along and perpendicular to it. The spectra of the clusters in perpendicular orientation to the **k** direction are averaged by rotation about **k**. As is seen from the figure, when the axes of the bi- and tri-spheres are perpendicular to **k**, no additional features in the $F(\pi)$ spectrum appear, and the amplitudes of the initial features, which are connected with the scattering on single spheres, grow due to the increase in the number of scatterers. At the same time, when the axes of the bi- and tri-spheres are oriented along **k**, additional interference features appear at λ > 0.4 μm. It can be shown that the minima near 0.70 and 0.95 μm for the tri- and bi-spheres, respectively, are caused by the interference of waves singly scattered by different CPs in the system. If the influence of particles above the frontal one relative to the incident wave were ignored in calculations of the interference, these minima would be formed at 0.69 and 0.92 μm, respectively. However, since the wave front immediately behind the frontal particle turns out to be retarded relative to the incident wave front and the near field becomes inhomogeneous

(see, e.g., the structure of the near field in the vicinity of a scatterer in Fig. 1b of a paper [4]), these minima are somewhat shifted to the long-wavelength range and their shape becomes slightly smoothed.

Due to this interference, the additional maximum is also formed; and it becomes more pronounced and moves to short wavelengths with increasing the CP number. At $\lambda < 0.38$ μm, this interference also manifests itself: it shifts the "individual" maximum to shorter wavelengths and makes it narrower with increasing the number of particles. The position of the minimum at $\lambda \approx 0.38$ μm is determined only by the properties of the CPs and almost independent of the number of CPs.

Let us apply the results of the analysis of spectral features in the spectra of individual spherical particles and bi- and tri-spheres to the explanation of the reflectance spectra of aggregates.

4. Changes in the spectra of aggregates caused by increasing the CP number

Since we may expect that the growth of an aggregate may induce changes in its spectrum, let us consider the influence of the number of CPs on the spectral dependences of $F(\pi)$. Figure 4 shows how the spectra of $F(\pi)$ for clusters B and C change with growing the number of CPs. The analogous dependences for cluster A is in Fig. 5 (left panel). We present examples for the clusters composed of ice monomers with $a = 0.115$ μm (the quantity R specified in the legends is the radius of the sphere with a volume equal to the sum of volumes of all CPs in the cluster expressed in micrometers).

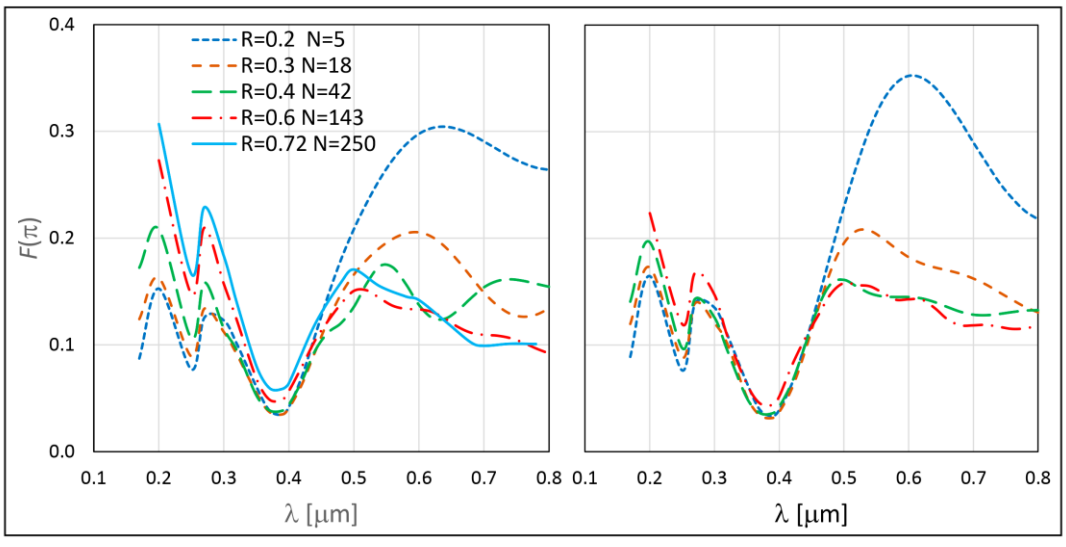

Fig. 4

Fig. 4. Changes in the spectra of $F(\pi)$ for clusters of structures B (left) and C (right) with growing the number of monomers N; their radius is $a = 0.115$ μm, and the material is H_2O ice.

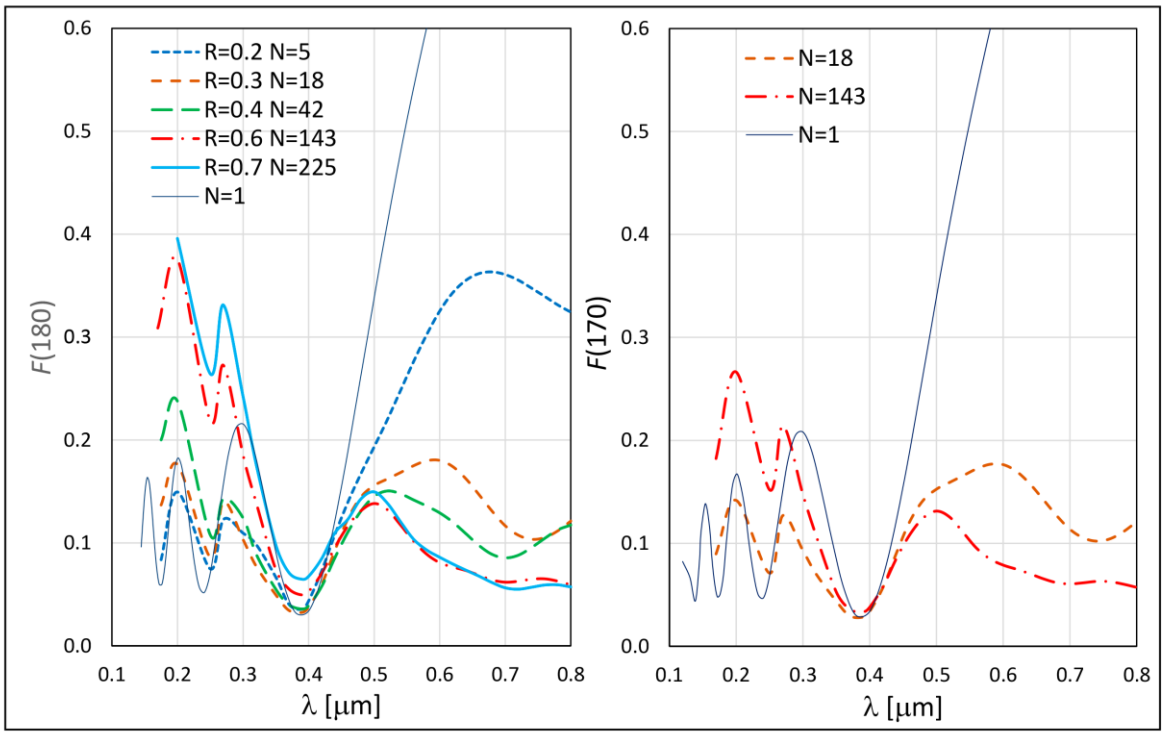

Fig. 5

Fig. 5. Changes in the spectra of $F(\theta=180°)$ (left) and $F(\theta=170°)$ (right) for clusters of structure A with growing the CP number N; their radius is a = 0.115 μm, and the material is H_2O ice. The spectral curve for a single monomer is shown for comparison.

As has been already determined, the spectral features in the short-wavelength range are mainly formed by the scattering on individual CPs. At the same time, the intensity of the reflected light grows here with growing the number of CPs. This increase is mainly induced by the weak-localization (opposition) effect. In particular, this follows from the results of our model calculations of the single scattering matrix for aggregates. They show that the linear polarization changes its sign in the back scattering domain as compared to the polarization sign for individual CPs (we do not show here all the results to avoid overloading the figures). For example, at λ = 0.25 and 0.27 μm, the degree of linear polarization of light scattered by monomers with a = 0.115 μm near opposition is positive, while it is negative for clusters of this monomers. This inversion of the polarization sign is characteristic of the weak-localization effect (see, e.g., [13–15, 17, 18] and references therein). Due to the same effect, the intensity of light scattered at θ = 170° (Fig. 5, right) is lower than that at θ = 180° (Fig. 5, left).

It is worth noting that, in this spectral range, the interference of singly scattered waves also manifests itself: it makes narrower the interference maxima and shifts them to shorter wavelengths (see also Fig. 3).

In Figs. 4 and 5, the minimum at 0.38 μm, which is formed by the scattering on individual CPs of the clusters, draws attention. Regardless the structure properties (see also Fig. 1), it barely moves and changes the shape with increasing the number of CPs. Consequently, this minimum may play an important role in estimating the parameters of particles in the clusters from the light scattering characteristics. In particular, from this minimum, one may determine the size of CPs in the clusters (if the refractive index is known) or the refractive index (if the CP size is known). Naturally, this inference is correct only to the media, where composing aggregates are in the far zones of each other.

At longer wavelengths, at $\lambda > 0.50$ μm, the second noteworthy feature develops with increasing the number of CPs: it is the maximum analogous to that observed for the bi- and tri-spheres in Fig. 3. In this spectral range, the shape of the curve strongly depends on the CP number; and, for a rather large number of CPs, the curve becomes insensitive to its further increase. For aggregates B and C the maxima at $\lambda \approx 0.50$ μm are less expressed than that for structure A, and the spectral curve at wavelengths $\lambda > 0.50$ μm becomes weakly declining or almost flat with growing N, which is explained by averaging the spectrum over several rather densely packed fragments of the aggregates.

We consider the maximum appeared at $\lambda \approx 0.50$ μm in the spectra of the examined aggregates as manifestation of collective effects caused by the interference of waves singly scattered by CPs in the aggregates (see also Fig. 3). There are more groups of particles as bi- and tri-spheres in more densely packed clusters than in fluffier ones with the same number of CPs. Consequently, cluster A exhibits the best developed "collective" maximum near 0.50 μm. This maximum becomes almost resistant to the further growth of N in cluster A already at $N = 42$, while the fluffy cluster D with $N = 42$ exhibits the collective maximum still at long wavelengths (see Fig. 1), where the maximum for the much smaller cluster A is observed.

The influence of different sizes of CPs in clusters was examined by an example of structures constructed in the following way. Since such clusters cannot be built with fractal relationships, we filled a specified volume with spherical particles of different sizes. Their radii were randomly varied around the assumed mean value with a difference ranging within $d = \pm 10\%$. We used the volume of a slightly "distorted" sphere, since the considered fractal-like clusters are more or less equidimensional, but we tried to avoid the wavy spectral features induced by scattering on a large spherical particle. We chose the distorting function that affects the radial position r_i of the ith CP, which is put into the volume in dependence on the angles of the spherical coordinate system θ_i and φ_i. Namely, $r_i = \mathrm{rand}(R - a_i)*(1 + f(\theta_i,\varphi_i) \times \cos^2\theta_i \times$

$\cos^2\varphi_i$), where $f(\theta_i,\varphi_i)$ was chosen in a different way in different realizations.

The number of CPs and the packing density were specified according to the values for a rather densely packed cluster of structure A. The size of the "quasi-sphere", being filled up, was varied until the required packing density of the polydisperse cluster was obtained. Then, the same volume was filled up with spherical particles with sizes differing within a range of d = ±20%; the obtained packing density of CPs in this cluster was smaller than in the previous case. To reach the same packing density, we decreased the filled volume. Thus, on the basis of the packing densities of two fractal-like clusters of type A composed of 42 and 143 monomers, we constructed one polydisperse cluster with d = ±10% and two polydisperse clusters, differing in size and packing density, with d = ±20% for each of them.

As one may expect, variations in the sizes of CPs in the above described aggregates substantially smooth the short-wavelength features that are caused by the interference of light scattered by individual CPs. The minimum near 0.38 μm may simultaneously be shifted to the short-wavelength range, and the wider the range of CP sizes, the shallower this minimum. At the same time, the collective maximum at longer wavelengths survives (see Fig. 6).

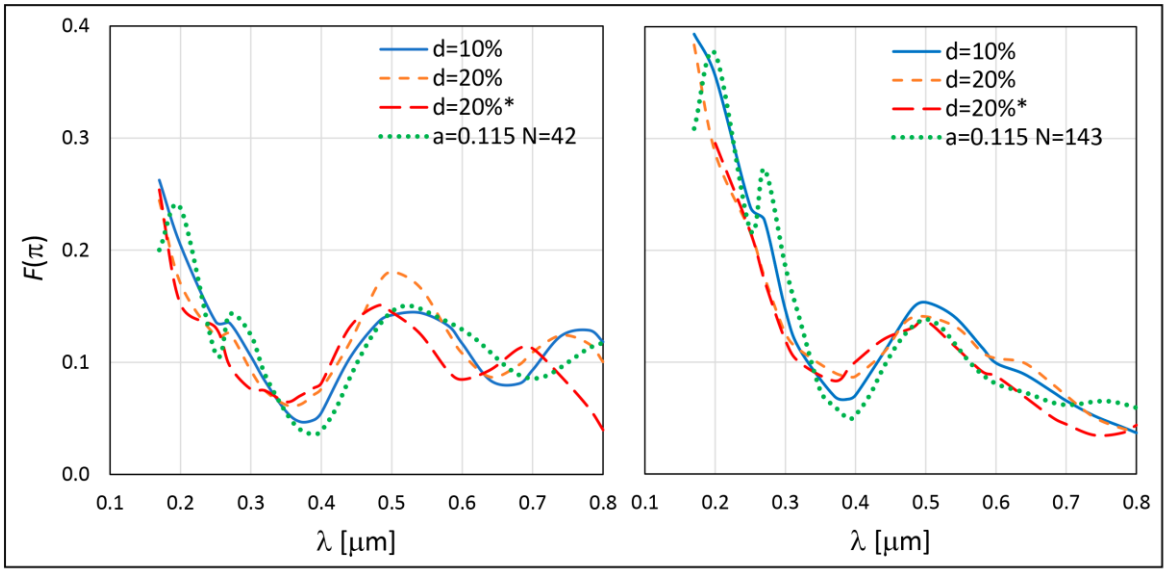

Fig. 6

Fig. 6. The spectral dependence of $F(\pi)$ for clusters composed of polydisperse spherical ice particles; the number of CPs is N = 42 (left) and 143 (right). The deviations in the CP sizes from the mean value (a = 0.115 μm) are within d = ±10 and 20% (the star marks the smaller cluster (see text)). The spectral curves for the clusters of structure A, containing the same number of identical CPs, are shown for comparison.

4. Concluding remarks

In this paper we examined the manifestations of collective effects in the visible spectrum of the intensity of light scattered by aggregate structures of submicron spherical particles. For this, we compared the model spectra of light scattered by different ensembles of particles and their individual constituents.

The results of the superposition T-matrix calculations showed that the spectra of randomly oriented fractal-like aggregates contain the interference extrema characteristic of individual constituents, and their positions in the spectra expectedly depends on the refractive index and the sizes of CPs. At longer wavelengths, where the intensity of light scattered by individual particles constantly grows, an additional maximum appears in the spectra of aggregates. We consider this maximum as manifestation of collective effects in aggregate structures caused by the interference of waves singly scattered by different CPs in the ensembles. In the spectra of randomly oriented aggregates, the position of this maximum, starting from some number of CPs, becomes almost resistant to the further aggregate growth and dependent only on the refractive index and sizes of CPs. The collective maximum survives even in the spectra of clusters with CPs of slightly varied sizes, while the interference features induced by individual constituents naturally become smoother.

It is worth noting that the most expressed minimum caused by the scattering at individual CPs in clusters, which is closest to the collective maximum, is not much sensitive to variations in the sizes of CPs within the clusters and strongly resistant to changes in the aggregate structure and CP number. Consequently, even in the case of somewhat differing CP sizes, it may serve for estimating the parameters of particles in the aggregates from the spectra measured for the media, where aggregates are in the far zones of each other.

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Figure captions

Fig. 1. The spectral dependences of $F(\pi)$ for single spherical ice particles with a radius of 0.115 μm and for aggregate structures A–D (shown top-down in the inset) composed of such particles.

Fig. 2. The value of $F(\pi)$ for spherical homogeneous particles of the specified materials in dependence on their size parameter (left) and the wavelength (right); the radius of particles is a = 0.115 μm.

Fig. 3. The spectra of $F(\pi)$ for bi- and tri-spheres in contact, the axes of which are oriented

perpendicular to (left panel) and along (right panel) the incident radiation direction **k**. The parameters of CPs are a = 0.115 μm and m = 1.33 + 0.0i. The spectral curve for single spherical particles of this properties is shown for comparison.

Fig. 4. Changes in the spectra of $F(\pi)$ for clusters of structures B (left) and C (right) with growing the number of monomers N; their radius is a = 0.115 μm, and the material is H_2O ice.

Fig. 5. Changes in the spectra of $F(\theta=180°)$ (left) and $F(\theta=170°)$ (right) for clusters of structure A with growing the CP number N; their radius is a = 0.115 μm, and the material is H_2O ice. The spectral curve for a single monomer is shown for comparison.

Fig. 6. The spectral dependence of $F(\pi)$ for clusters composed of polydisperse spherical ice particles; the number of CPs is N = 42 (left) and 143 (right). The deviations in the CP sizes from the mean value (a = 0.115 μm) are within d = ±10 and 20% (the star marks the smaller cluster (see text)). The spectral curves for the clusters of structure A, containing the same number of identical CPs, are shown for comparison.