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Volume Scattering Function of Light for a Mixture of Polydisperse Small Particles with Various Optical Properties

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Abstract – Scattering of electromagnetic waves by a polydisperse system of small spherical hydrosol particles in the scalar approximation is considered. The equation for the scattering cross-section which generalizes the well-known Rayleigh-Gans-Rocar formula for a scattering medium containing particles characterized by continuous distributions in their dielectric permittivities at any given size is derived. For a natural situation, such as a coastal environment, the difference between volume scattering functions proposed in this paper and conventionally derived scattering functions can be as large as 50% for the particle size distributions with the maximum size of particles less than ten wavelengths of light.

INTRODUCTION

Conventional volume scattering function calculations use a particle size distribution and a fixed dielectric permittivity for each size class. This approach became widely accepted due to two main reasons: first, it is mathematically simpler compared to a more general approach, and, second, it is accurate enough for open ocean situations. In these simple cases we can divide all scattering particles into a few different particle types, each with an assigned dielectric permittivity. However, as emphasis has switched to coastal environments, the number of particle types in each size class has increased. We include this diversity of particle types in the calculation of the volume scattering function.

In order to incorporate this real-life complexity we utilize a two-dimensional distribution of particles with respect to particle size and dielectric permittivity. We consider multiple scattering but, because of the complexity of the problem, restrict ourselves to the case of small particles and use the scalar approximation to Maxwell's equations for electromagnetic theory. Using this approach we calculate the contribution to the volume scattering function due to small particles for various coastal-type hydrosols.

APPROACH

Consider the scattering of scalar electromagnetic waves in a medium represented by a polydisperse system of N particles. Let ε_0 denote the dielectric permittivity of the surrounding medium, while ε_n is the dielectric permittivity of a particle with index n. The dielectric permittivity of a mixture is a function of the random distribution of the particles' coordinates $\{\vec{r}_n\}$. For a particular configuration of a polydisperse mixture of hydrosol particles with different electric properties it can be represented in the following form [1, 2]

$$\tilde{\varepsilon}(\vec{r},\{\vec{r}_n\}) = \varepsilon_0 + \sum_{n=1}^{N} (\varepsilon_n - \varepsilon_0)\,\theta(a_n - |\vec{r} - \vec{r}_n|), \qquad (1)$$

where $\theta(x)$ is the Heavyside (or step) function defined as : $(\theta(\mu) = 1, \mu > 0; \theta(\mu) = 0, \mu \le 0)$, and a_n is the size associated with the n^{th} particle determined from

$$a_n = (3V_n / 4\pi)^{1/3}, \qquad (2)$$

where V_n is the volume of the n^{th} particle. For a spherical particle, a_n coincides with its radius. In the above equation, $\vec{r}$ is the field vector, while $\vec{r}_n$ is the position vector of n^{th} particle. The field scattered by a system of N particles is a random function of both the coordinates and scattering properties of the particles in that system.

In order to calculate average values of physical quantities of interest, we use Foldy's probability distribution method [3] for the ensemble of possible configurations of the dielectric scatterers. Foldy represents "the probability of finding the scatterers in a configuration in which the first scatterer lies in the element of volume $d\vec{r}_1$ about the point $\vec{r}_1$ and has a scattering parameter lying between S_1 and $S_1 + dS_1$, the second scatterer lies in the element of volume $d\vec{r}_2$ about the point $\vec{r}_2$ and has a scattering parameter lying between S_2 and $S_2 + dS_2$, etc." as

$$F(\vec{r}_1, \vec{r}_2, ...\vec{r}_N, S_1, S_2, ...S_N)\, dr_1\, dr_2 ... dr_N\; dS_1\, dS_2 ... dS_N. \qquad (3)$$

In this article we consider only scattering properties such as the dielectric permittivity ε and the particle size x. Then: $dS_n = d\varepsilon_n dx_n$. To find the configurational average of a physical quantity *f*, we multiply its value for a particular configuration by (3) and integrate over all values of the $\vec{r}_n$ and S_n accessible to the scatterers:

$$\langle f \rangle = \iint F(\{S\},\{\vec{r}\})\, f\, d\{S\}\, d\{\vec{r}\}. \qquad (4)$$

In the spirit of Foldy's article [3], we write

$$F(\{S\},\{\vec{r}\}) = n(S_1,\vec{r}_1)\, n(S_2,\vec{r}_2) ... n(S_N,\vec{r}_N), \qquad (5)$$

which is equivalent to assuming that “the probability that a particular scatterer is located in some volume element and has a value of S located within some range dS is independent of the locations and scattering parameters of the other scatterers and is the same for all scatterers”. We note that

$$n(S,\vec{r}) = n_{Foldy}(S,\vec{r}) / N, \qquad (6)$$

so that $n(S,\vec{r})\, d\vec{r}\, dS$ represents the *percentage* of scatterers in the element of volume $d\vec{r}$ about the point $\vec{r}$ with scattering parameters between S and $S + dS$.

In what follows, we restrict ourselves to spherical particles, neglect thermal and turbulent fluctuations of the medium and assume homogeneity. We can then write:

$$n(S,\vec{r}) = n(S) = V^{-1} f_x(\varepsilon)\, \varphi(x), \qquad (7)$$

where V is the volume of the system, $f_x(\varepsilon)$ is the dielectric permittivity distribution function for a particle with size *x*, and $\varphi(x)$ is the size-distribution function. The distribution functions in (7) are normalized according to

$$\iint n(S,\vec{r})\, dS\, d\vec{r} = 1, \quad \int f_x(\varepsilon)\, d\varepsilon = 1, \quad \int_0^\infty \varphi(x)\, dx = 1. \qquad (8)$$

Using (4) and (5) we can calculate the average dielectric permittivity of the medium from (1):

$$\left\langle \tilde{\varepsilon}(\vec{r},\{\vec{r}\}) \right\rangle = \varepsilon_0 + C_H\, \varepsilon_H, \qquad (9)$$

In the above equation

$$C_H = N V_H / V \qquad (10)$$

is the volume concentration of particles as a fraction of the total volume,

$$V_H = (4\pi/3) \int_0^\infty x^3 \varphi(x)\, dx \qquad (11)$$

is the average particle volume, and

$$\varepsilon_H = \int_0^\infty \varepsilon_x\, \varphi(x)\, x^3 dx \Big/ \int_0^\infty \varphi(x)\, x^3 dx \qquad (12)$$

is the standard deviation of the average dielectric permittivity ε_x due to the particles with respect to the dielectric permittivity ε_0 of the surrounding medium, such that

$$\varepsilon_x = \int (\varepsilon - \varepsilon_0) f_x(\varepsilon)\, d\varepsilon. \qquad (13)$$

In this case the formula (1) for the dielectric permittivity of a mixture can be rewritten as

$$\tilde{\varepsilon} = \varepsilon_w - \varepsilon; \quad \varepsilon_w = \varepsilon_0 + C_H\, \varepsilon_H, \qquad (14)$$

$$\varepsilon = C_H\, \varepsilon_H - \sum_{n=1}^{N} (\varepsilon_n - \varepsilon_0)\, \theta(a_n - |\vec{r} - \vec{r}_n|), \ \langle \varepsilon \rangle = 0. \qquad (15)$$

The equation for a monochromatic scalar wave field in a scattering medium [4] will be

$$\left(\Delta + \tilde{\varepsilon}\, \omega^2 / c^2\right) \tilde{\Psi} = f, \qquad (16)$$

where $\Delta \equiv \nabla^2$ is the Laplace operator, ω is the circular frequency, c is the speed of light in vacuo, $\tilde{\Psi}$ is the wave field amplitude, and f is the source function. Let us introduce the following notation:

$$\kappa^2 = \varepsilon_w\, \omega^2 / c^2; \quad \kappa_0^2 = \omega^2 / c^2, \qquad (17)$$

$$\tilde{\Psi} = \Psi + \varphi; \quad \langle \varphi \rangle = 0, \qquad (18)$$

$$\hat{L} = \Delta + \kappa^2, \qquad (19)$$

so that $\hat{L}$ is the Helmholtz operator.

We derive an equation for Ψ by averaging (16), and an equation for φ by subtracting the averaged equation from (16). Then we have the system of equations

$$\left.\begin{aligned} \hat{L}\ \Psi &= \kappa_0^2\, \langle \varepsilon \varphi \rangle + f \\ \hat{L}\ \varphi &= \kappa_0^2\, \varepsilon\, \Psi + \kappa_0^2 [\varepsilon, \varphi] \end{aligned}\right\}, \qquad (20)$$

where

$$[\varepsilon, \varphi] = \varepsilon \varphi - \langle \varepsilon \varphi \rangle. \qquad (21)$$

The Helmholtz operator (19) has an inverse operator $\hat{G}$. Operator $\hat{G}$ is an integral operator with a Green function G (or a fundamental solution of the Helmholtz equation [5]) as kernel

$$(\hat{L})^{-1} = \hat{G}, \quad G(r) = -\exp(i\kappa r)/(4\pi r), \qquad (22)$$

$$(\hat{G} f)\,(\vec{r}) = \int G(\vec{r} - \vec{r}\,')\, f(\vec{r}\,')\, d\vec{r}\,', \qquad (23)$$

with

$$\hat{L}\,\hat{G} = \hat{G}\,\hat{L} = \hat{1}, \quad \hat{L}\, G(\vec{r}) = \delta(\vec{r}), \qquad (24)$$

$$(\hat{1}\ f)(\vec{r}) \equiv \int \delta(\vec{r} - \vec{r}\,')\, f(\vec{r}\,')\, d\vec{r}\,' = f(\vec{r}). \qquad (25)$$

Let us introduce a random operator $\hat{K}$, which acts on an arbitrary (random or deterministic) function F according to the rule

$$\hat{K}\, F = \hat{G} [\varepsilon, F] \equiv \hat{G} \left(\varepsilon\, F - \langle \varepsilon\, F \rangle \right). \qquad (26)$$

When acting on a deterministic function F_0 it gives

$$\hat{K}\, F_0 = \hat{G} [\varepsilon, F_0] = \hat{G}\, \varepsilon\, F_0. \qquad (27)$$

Let us also introduce the operator

$$\left(1 - \kappa_0^2 \hat{K}\right)^{-1} = \sum_{n=0}^{\infty} \kappa_0^{2n}\, \hat{K}^n\ , \qquad (28)$$

and act with the operator $\hat{G}$ on the system of equations (20). Then, instead of (20) we get

$$\left(\hat{L} - \kappa_0^2 \left\langle \varepsilon\, \hat{Q} \right\rangle \right) \Psi = f, \quad \varphi = \hat{Q}\, \Psi, \qquad (29)$$

where

$$\hat{Q} = \sum_{n=1}^{\infty} \hat{Q}_n \equiv \sum_{n=1}^{\infty} \kappa_0^{2n}\, \hat{K}^n\ . \qquad (30)$$

Let us restrict ourselves presently to a first order approximation for which

$$\hat{L}^0 \Psi = \left(\hat{L} - \kappa_0^4 \left\langle \varepsilon\, \hat{G}\, \varepsilon \right\rangle \right) \varphi^0 = f, \quad \varphi^0 = \kappa_0^2\ \hat{G}\ \varepsilon\ \Psi^0 \qquad (31)$$

Let us examine the structure of the operator

$$\hat{L}^0(1,2) = \hat{L}(1,2) - \kappa_0^4\, G(1,2)\, B_\varepsilon(1,2), \qquad (32)$$

where the spatial correlation function

$$B_\varepsilon(1,2) = \langle \varepsilon(1)\, \varepsilon(2) \rangle = B_\varepsilon(\vec{r}_1, \vec{r}_2) = B_\varepsilon(\vec{r}_1 - \vec{r}_2), \qquad (33)$$

and the square of field fluctuations is

$$\left\langle \varphi^0(1)\varphi^{*0}(1)\right\rangle = \kappa_0^4\, G(1,2)\,G^*(1,3)\,B_\varepsilon(2,3)\,\Psi^0(2)\,\Psi^{*0}(3)\ ,(34)$$

where asterisk denotes complex conjugates. In order to simplify the notation in what follows, we introduce numerical indices *1, 2,…* instead of $\vec r_1, \vec r_2, \ldots$, with implied integration over repeating indices.

When $N >> 1$, and with the use of (4) and (15), it is not difficult to derive the following expression for the correlation function $B_\varepsilon(R)$ (see APPENDIX):

$$B_\varepsilon(R) = \frac{N}{V}\int_{R/2}^{\infty}\frac{4\pi}{3}x^3\,\varepsilon_x^{(2)}\,\varphi(x)\left(\frac{R}{2x}-1\right)^2\left(\frac{R}{4x}+1\right)dx,\ (35)$$

where $$\varepsilon_x^{(2)} = \int(\varepsilon-\varepsilon_0)^2\, f_x(\varepsilon)\,d\varepsilon. \qquad (36)$$

represents the variance of the dielectric permittivity fluctuations.

The Fourier transform of this correlation function will be needed later and is given by:

$$\Phi_\varepsilon(k) = (2\pi)^{-3}\int B_\varepsilon(R)\exp(-i\,\vec k\,\vec r)\,d^3R$$

$$= \left[1/(2\pi^2 k)\right]\int_0^\infty B_\varepsilon(R)\sin(kR)\,R\,dR. \qquad (37)$$

Using (35) it is also not difficult to show that

$$\Phi_\varepsilon(k) = \frac{N}{V}\frac{1}{(2\pi)^3}\int_0^\infty\left[\frac{4\pi}{3}x^3\,f(kx)\right]^2\varepsilon_x^{(2)}\,\varphi(x)\,dx, \qquad (38)$$

where $$f(kx) = 3\left[\sin(kx) - kx\cos(kx)\right]/(kx)^3. \qquad (39)$$

For a monodisperse hydrosol where $\varphi(x)=\delta(x-a)$, and $f_x(\varepsilon)=\delta(\varepsilon-\varepsilon_m)$, we find that

$$\Phi_\varepsilon^{mono}(k) = \frac{N}{V}\frac{(\varepsilon_m-\varepsilon_0)^2}{(2\pi)^3}\left[\frac{4\pi}{3}a^3 f(ka)\right]^2 = C_H\,\frac{(\varepsilon_m-\varepsilon_0)^2}{6\pi^2}\,a^3 f^2(ka). \qquad (40)$$

SCATTERING CROSS-SECTION

Consider a plane-parallel wave $A_0\exp(i\kappa_0'\,\vec n\,\vec r)$ falling on a certain part V_S of the volume V. (here $\vec n$ is a unit vector pointing in the direction of propagation; $\kappa_0' = \varepsilon_0\,\kappa_0$). The fluctuating field outside of the scattering volume V_S, caused by scattering due to the particles inside this volume, is given by (31):

$$\varphi^0 = \kappa_0^2\,\hat G\,\varepsilon\,\Psi^0; \qquad (41)$$

where the average field Ψ^0 is represented by a plane-parallel wave [4]

$$\Psi^0(\vec r) = A_0\exp(iq\,\vec n\,\vec r). \qquad (42)$$

Here q is a wave vector. It corresponds to the Fourier transform of a bi-local Green function $G^0 = (\hat L^0)^{-1}$ and can be expressed by the equation

$$q^2 = \kappa^2 + \left(\kappa_0^4/q\right)\int_0^\infty B_\varepsilon(R)\exp(i\kappa R)\sin(qR)\,dR. \qquad (43)$$

When the following condition is valid,

$$C_H\int\left(\varepsilon/\varepsilon_0 - 1\right)^2 f_x(\varepsilon)\,d\varepsilon\ <<\ 1, \qquad (44)$$

the integral in (43) is small compared to κ^2 so that $q\cong\kappa$ Neglecting higher order terms we obtain

$$q = q' + i\,q'', \qquad (45)$$

$$q' = \kappa\left[1 + (1/8)\left(\kappa_0/\kappa\right)^4\right]\int_0^\infty B_\varepsilon\left(z/2\kappa\right)\sin z\,dz, \qquad (46)$$

$$q'' = \left(\kappa/2\right)\left(\kappa_0/\kappa\right)^4\int_0^\infty B_\varepsilon\left(z/2\kappa\right)\sin^2 z\,dz. \qquad (47)$$

During the derivation of the scattering cross-section we will consider that the field point is situated in the Fraunhofer diffraction zone, *i. e.*

$$|\,\vec r - \vec r'\,| \cong \vec r - \vec n^s\,\vec r, \qquad (\vec n^s \equiv \vec r/r).$$

Then

$$\left\langle\varphi^0\,\varphi^{*0}\right\rangle \cong \left[\kappa_0^4 A_0^2\big/\left(16\pi^2 r^2\right)\right]\times \int_{V_s}\int_{V_s}\exp\{\,i[\kappa\,\vec n_s(\vec r''-\vec r') - q'\,\vec n\,(\vec r''-\vec r')] - q''\,\vec n\,(\vec r'+\vec r'')\}\,B_\varepsilon(\vec r'-\vec r'')\ d^3\vec r'\,d^3\vec r''. \qquad (48)$$

Introducing the new variable $\vec R = \vec r' - \vec r''$ and integrating over $\vec r''$, which under the assumption that $q''V_s^{1/3} << 1$ gives the scattering volume V_s, we get

$$\left\langle\varphi^0\,\varphi^{*0}\right\rangle \cong \frac{\kappa_0^4 A_0^2 V_s}{16\pi^2 r^2}\int_{V_s}\exp[-i(\kappa\,\vec n_s - q'\,\vec n)\,\vec R]\,B_\varepsilon(\vec R)\,d^3\vec R. \ (49)$$

Regarding the linear size of the scattering volume as big in comparison with the correlation length a of the function B_ε (it corresponds to an average radius of an ensemble of scattering particles) $V_s << a^3$, we can expand the integration in (49) to infinity, and obtain

$$\left\langle\varphi^0\,\varphi^{*0}\right\rangle = \frac{\pi\,\kappa_0^4 A_0^2 V_s}{2\,r^2}\Phi_\varepsilon(\kappa\,\vec n_s - q'\,\vec n) \equiv \frac{\pi\,\kappa_0^4 A_0^2 V_s}{2\,r^2}\times \Phi_\varepsilon\left(\sqrt{4\kappa^2[1-(q'-\kappa)/\kappa]\sin^2(\theta/2) + (q'-\kappa)^2}\right) \qquad (50)$$

where $\theta = \cos^{-1}(\vec n\,\vec n')$ is the scattering angle. The energy scattered by the volume V_s in the direction of $\vec n_s$ in a solid angle $d\Omega$ will be

$$dE = \left[c\left\langle\varphi^0\,\varphi^{*0}\right\rangle\big/(8\pi)\right]r^2 d\Omega = S_0\,\sigma_0\,V_s\,d\Omega, \qquad (51)$$

where $S_0 = c\,A_0^2/(8\pi c)$ is the energy flux density of the incident wave. From this equation we derive an effective scattering cross-section from a unit volume to a unit solid angle in the direction of $\vec n_s$

$$\sigma_s = \left(\pi\,\kappa_0^4/2\right)\Phi_\varepsilon\left(2\kappa\sqrt{[1+\Delta(\kappa)]\,\sin^2(\theta/2) + \Delta^2(\kappa)/4}\right), (52)$$

where

$$\Delta(\kappa) = \frac{q'-\kappa}{\kappa} = \frac{\kappa^2}{2\,\varepsilon_w^2}\int_0^\infty B_\varepsilon(R)\sin R\,dR, \quad \text{or} \quad \Delta(\lambda) = \frac{2\pi^2}{\varepsilon_w\lambda^2}\int_0^\infty B_\varepsilon(R)\sin R\,dR, \qquad (53)$$

$$B_\varepsilon(R) = \frac{4\pi N}{3V} \int_{R/2}^{\infty} \varepsilon_x^{(2)} \left(\frac{R}{2x} - 1\right)^2 \left(\frac{R}{4x} + 1\right) x^3 \varphi(x)\, dx, \quad (54)$$

$$\varepsilon_x^{(2)} = \int_0^{\infty} (\varepsilon - \varepsilon_0)^2 f_x(\varepsilon)\, d\varepsilon, \ \varepsilon_w = \varepsilon_0 + C_H\, \varepsilon_H. \quad (55)$$

The transition from the scalar wave to the electromagnetic field is made by introducing the coefficient $\sin^2\chi$ [4, 6], which takes into account the vector nature of the electromagnetic field. Here $\chi = \cos^{-1}(\vec{n}^E \vec{n}^S)$, where $\vec{n}^E$ is the unit vector of polarization of the incident wave. Let β be the azimuthal angle between vector $\vec{n}^E$ and the projection of the scattering vector $\vec{n}^S$ on a plane orthogonal to the wave vector $q\vec{n}$. Then

$$\sin^2\chi = 1 - \cos^2\beta \ \sin^2\theta. \quad (56)$$

The scattering cross-section in this case will then become

$$\sigma_s(\theta,\beta) = \frac{\pi}{2}\kappa_0^4 (1 - \cos^2\beta \ \sin^2\theta) \times \Phi_\varepsilon\left(2\kappa\sqrt{[1+\Delta(\kappa)] \sin^2(\theta/2) + \Delta^2(\kappa)/4}\right). \quad (57)$$

If $q' \to \kappa$, then $\Delta \to 0$, and (57) will coincide with the well-known Rayleigh-Gans-Rocar equation [7, 8], averaged over the particle size distribution function (without considering depolarization effects inside particles which are irrelevant to the shape of phase function).

For unpolarized light (57) should be averaged over the azimuthal angle β, which gives

$$\sigma_s(\theta,\beta) = \frac{\pi}{2}\kappa_0^4 (1 - \cos^2\beta \ \sin^2\theta) \times \Phi_\varepsilon\left(2\kappa\sqrt{[1+\Delta(\kappa)] \sin^2(\theta/2) + \Delta^2(\kappa)/4}\right), \quad (58)$$

with Φ_ε given by (38).

PHASE FUNCTION

The phase function for scattering by a polydisperse inhomogeneous ensemble of dielectric particles is given by the following equation

$$p(\theta) = \left[2(1+\cos^2\theta)/A_p\right] \times \Phi_\varepsilon\left((2\pi/\lambda)\sqrt{\varepsilon_w\{[1+\Delta(\lambda)] \sin^2(\theta/2) + \Delta^2(\lambda)/4\}}\right), \quad (59)$$

where ε_w is given by (14), (15) and (10), and the constant of normalization can be computed according to

$$A_p = \int_0^{\pi} (1+\cos^2\theta)\ \Phi_\varepsilon \sin\theta\, d\theta. \quad (60)$$

The phase function (59) is normalized according to the condition

$$0.5\int_0^{\pi} p(\theta) \sin\theta\, d\theta = 1. \quad (61)$$

Eqn. (59) together with (53)-(55), (12), (14) and (39) can be used to calculate the scattering phase function for a polydisperse system of particles of different types. The input parameters for calculation of $p(\theta)$ are: the size distribution function of particles $\varphi(x)$, the distribution function of values of dielectric permittivity for a given size $f_x(\varepsilon)$, and the volume concentration of particles C_H.

EXAMPLE

In order to demonstrate the difference in phase functions calculated by the polydisperse and monodisperse approaches we considered the following two simple cases:

Case 1.

The size distribution function is given by

$$\varphi(x) = \begin{cases} c_x\, x^{-2.5}, & a_x \le x \le b_x, \\ 0, & elsewhere, \end{cases} \quad (62)$$

here

$$c_x = 1.5\Big/\left(a_x^{-1.5} - b_x^{-1.5}\right). \quad (63)$$

The dielectric permittivity distribution function for a given size x is represented as a superposition of rectangular and Dirac delta-function:

$$f_x(\varepsilon) = \alpha_x \Pi(\varepsilon) + (1-\alpha_x)\,\delta(\varepsilon - \varepsilon_m), \quad (64)$$

here

$$\alpha_x = (x - a_x)/(b_x - a_x) \quad (65)$$

$$\Pi(\varepsilon) = \begin{cases} \pi_\varepsilon, & \varepsilon_0 < \varepsilon < 2\varepsilon_m - \varepsilon_0, \\ 0, & elsewhere, \end{cases} \quad (66)$$

$$\pi_\varepsilon = 1\Big/\left[2(\varepsilon_m - \varepsilon_0)\right]. \quad (67)$$

This case corresponds to a polydisperse hydrosol with the size-dependent dielectric permittivity distributed between values of ε_0 and $2\varepsilon_m - \varepsilon_0$.

Case 2.

The size distribution function is taken identical to the one used in *Case 1* and given by (62)-(63). The dielectric permittivity distribution function for a given size x is represented by Dirac delta function [5]:

$$f_x(\varepsilon) = \delta(\varepsilon - \varepsilon_m). \quad (68)$$

This case corresponds to a monodisperse hydrosol with the unique value for the dielectric permittivity equal to ε_m.

Due to spatial limitations we cannot display the pictures associated with both considered cases. We strongly emphasize that neglecting the dielectric permittivity distribution function will cause differences in the resulting phase function in the range of 50%, which can be regarded as very significant.

CONCLUSION

We have shown here that our formulation gives a more general expression for scattering of light by a polydisperse ensemble of dielectric particles than in the Rayleigh-Gans-Rocar approximation [7, 8]. For the special case of a particle size distribution represented by a single dielectric permittivity (a one-component hydrosol) our results reduce to the Rayleigh-Gans-Rocar approximation. For a natural situation such as a coastal environment, the difference between our volume scattering function and conventionally derived scattering functions can be as large as 50% for the particle size distributions $(x < 10\,\lambda)$ considered here. In view of such potential differences, a similar approach to deal with the larger particles in coastal waters should be considered.

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APPENDIX: DERIVATION OF THE CORRELATION FUNCTION

Using (15) we can write

$$\langle \varepsilon(\vec r_1)\varepsilon(\vec r_2)\rangle \equiv B_\varepsilon(\vec r_1,\vec r_2) = (C_H\varepsilon_H)^2 - C_H\varepsilon_H + \left\langle \sum_{n=1}^{N}(\varepsilon_n-\varepsilon_0)\big(\theta(a_n-|\vec r_1-\vec r_n|)+\theta(a_n-|\vec r_2-\vec r_n|)\big)\right\rangle + \left\langle \sum_{n=1}^{N}\sum_{m=1}^{N}(\varepsilon_n-\varepsilon_0)(\varepsilon_m-\varepsilon_0)\theta(a_n-|\vec r_1-\vec r_n|)\,\theta(a_m-|\vec r_2-\vec r_n|)\right\rangle, \quad (1a)$$

The average in the second and third summations according to (1) and (14) is equal to $C_H\varepsilon_H$. In the double summation term we extract the $m=n$ term:

$$B_\varepsilon(\vec r_1,\vec r_2) = -(C_H\varepsilon_H)^2 + \left\langle \sum_{n=1}^{N}(\varepsilon_n-\varepsilon_0)^2\theta(a_n-|\vec r_1-\vec r_n|)\theta(a_n-|\vec r_2-\vec r_n|)\right\rangle + \left\langle \sum_{n=1}^{N}(\varepsilon_n-\varepsilon_0)\theta(a_n-|\vec r_1-\vec r_n|)\right\rangle \times \left\langle \sum_{m=1,m\neq n}^{N}(\varepsilon_m-\varepsilon_0)\theta(a_m-|\vec r_2-\vec r_m|)\right\rangle. \quad (2a)$$

By neglecting the terms where $\sim 1/N$, (because $N>>1$) and explicitly introducing the averaging operation according to (4), (5) and (7), we have

$$B_\varepsilon(R) = (N/V)\int_V d^3\rho\int_0^\infty dx\,\varphi(x)\int d\varepsilon\, f_x(\varepsilon) \times(\varepsilon-\varepsilon_0)^2\theta(x-|\rho|)\theta(x-|\vec R+\vec\rho|). \quad (3a)$$

By integrating over the polar angle, we obtain

$$B_\varepsilon(R) = (2\pi N/V)\int_0^\infty \varepsilon_x^{(2)}\varphi(x)\,dx\int_0^x \rho^2 d\rho \times\int_{-1}^{1} d\eta\;\theta\left(x-\sqrt{\rho^2+R^2+2\rho R\eta}\right), \quad (4a)$$

where

$$\varepsilon_x^{(2)} = \int d\varepsilon\, f_x(\varepsilon)(\varepsilon-\varepsilon_0)^2. \quad (5a)$$

For convenience let us introduce the function

$$D(x,R,\rho) = (x^2-R^2-\rho^2)/(2R\rho), \quad (6a)$$

with which we can rewrite (4a) as

$$B_\varepsilon(R) = (2\pi N/V)\int_0^\infty \varepsilon_x^{(2)}\, I(x)\,\varphi(x)\,dx, \quad (7a)$$

where

$$I(x) = \int_0^x J(x,\rho)\rho^2 d\rho, \quad (8a)$$

and

$$J(x,\rho) = \int_{-1}^{1}\theta(D-\eta)\,d\eta = 2\theta(D-1)+(1+D)\theta(1-D^2). \quad (9a)$$

Integrating (9a) with (8a)and after analyzing the behavior of D in a $R\rho$-plane, we find

$$I(x) = (2/3)\theta(2x-R)x^3[R/(2x)-1]^2[R/(4x)+1]. \quad (10a)$$

After inserting (10a) in (7a), we obtain formula (35) for the correlation function

$$B_\varepsilon(R) = \frac{N}{V}\int_{R/2}^{\infty}\frac{4\pi}{3}x^3\varepsilon_x^{(2)}\varphi(x)\left(\frac{R}{2x}-1\right)^2\left(\frac{R}{4x}+1\right)dx. \quad (11a)$$

We also present here the value of the correlation function and its derivative at $R=0$

$$B_\varepsilon(0) = \frac{N}{V}\int_0^\infty\frac{4\pi}{3}x^3\varepsilon_x^{(2)}\varphi(x)\,dx = C_H\,\varepsilon_H^{(2)}, \quad (12a)$$

where

$$\varepsilon_H^{(2)} = \int_0^\infty \varepsilon_x^{(2)}x^3\varphi(x)dx \Big/ \int_0^\infty x^3\varphi(x)dx, \quad (13a)$$

$$\left.\frac{\partial B_\varepsilon(R)}{\partial R}\right|_{R=0} = -\frac{N}{V}\int_0^\infty \pi x^2\varepsilon_x^{(2)}\varphi(x)\,dx = -\left[3C_H/(4a^3)\right]\int_0^\infty x^2\varepsilon_x^{(2)}\varphi(x)\,dx. \quad (14a)$$

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