LIGHT SCATTERING BY SMALL NON-SPHERICAL INHOMOGENEOUS (LAYERED) PARTICLES: THE APPLICABILITY OF APPROXIMATIONS

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ABSTRACT

The quasi-static approximation which is a generalization of the Rayleigh and Rayleigh-Gans approximations is considered for (layered) ellipsoids. The approximate expressions for cross-sections and elements of the amplitude matrix are given in the particle and laboratory coordinate systems. Calculations using the quasi-static and other approximations as well as exact methods were performed, and their results are briefly discussed.

1. INTRODUCTION

Among possible models of finite-size non-spherical scatterers, the *ellipsoidal* model occupies an important place. Besides the diffraction parameter characterizing the size of a scatterer relative to the wavelength of incident radiation, the model has two more independent parameters (the semiaxis ratios a/b and b/c) describing deviations of particle's shape from the spherical one in all three perpendicular directions.

Determination of the scattering characteristics of ellipsoids using available exact methods of the light scattering theory is still a very computational time-consuming task. Therefore one often utilizes different approximations: Rayleigh, Rayleigh-Gans, van de Hulst and other ones (see, e.g., Mishchenko et al., 2000 and references therein).

We consider the quasi-static approximation. Its main idea is as follows: the internal field is represented by the incident plane wave (as in the Rayleigh-Gans approximation) taking into account the polarizability of the particle (as in the Rayleigh approximation). Such an approach basically resembles the iteration method of Shifrin (1951) and has been discussed in the literature several times (see, e.g., Volkovitsky et al., 1984 for a review).

In this paper we present formulae of the quasi-static approximation in the case of (layered) ellipsoids and compare results of numerical calculations made using this and other approximations and exact methods.

2. BASIC EQUATIONS

Let us introduce two Cartesian coordinate systems with the origins at the center of an ellipsoidal particle. The axes of the first system (X, Y, Z) are coupled with the (semi)axes of the particle (a, b, c). In the second system (x, y, z), the axis z is parallel to the direction of incident radiation.

In the quasi-static approximation the field inside a particle $\vec{E}^{(2)}$ can be written as follows:

$$\vec{E}^{(2)} = K_1(\vec{E}^{(0)}, \vec{i}_{\rm X})\vec{i}_{\rm X} + K_2(\vec{E}^{(0)}, \vec{i}_{\rm Y})\vec{i}_{\rm Y} + K_3(\vec{E}^{(0)}, \vec{i}_{\rm Z})\vec{i}_{\rm Z}, \tag{1}$$

where $(\vec{i}_{\rm X}, \vec{i}_{\rm Y}, \vec{i}_{\rm Z})$ are the unit vectors of the system (X, Y, Z), $\vec{E}^{(0)}$ is the incident radiation. The coefficients K_j connect the polarizability of the particle $\tilde{\alpha}_j$ with the geometrical factors L_j (see Eq. (4) below).

Bohren and Huffman (1983) give the expressions for elements of the amplitude matrix for ellipsoids in the Rayleigh approximation. It is obvious that in the quasi-static approximation one has only to add the form factor typical of the Rayleigh-Gans approximation to these expressions

$$\begin{aligned} S_1 &= \frac{-ik_0^3}{4\pi}\left(\hat{\alpha}_{11}\sin^2\varphi - 2\hat{\alpha}_{12}\sin\varphi\cos\varphi + \hat{\alpha}_{22} \right. \\ &\quad \left. \times\cos^2\varphi\right) G(u), \\ S_2 &= \frac{-ik_0^3}{4\pi}\left[\cos\theta\left(\hat{\alpha}_{11}\cos^2\varphi + 2\hat{\alpha}_{12}\sin\varphi\cos\varphi \right.\right. \\ &\quad \left.\left. +\hat{\alpha}_{22}\sin^2\varphi\right) - \sin\theta\left(\hat{\alpha}_{13}\cos\varphi + \hat{\alpha}_{23}\sin\varphi\right)\right] \\ &\quad \times G(u), \end{aligned} \tag{2}$$

$$
\begin{aligned}
S_3 &= \frac{-ik_0^3}{4\pi}\left[\cos\theta\left(\hat{\alpha}_{11}\sin\varphi\cos\varphi+\hat{\alpha}_{12}\left(\sin^2\varphi\right.\right.\right. \\
&\quad \left.\left.-\cos^2\varphi\right)-\hat{\alpha}_{22}\sin\varphi\cos\varphi\right)-\sin\theta \\
&\quad \left.-\left(\hat{\alpha}_{13}\sin\varphi-\hat{\alpha}_{23}\cos\varphi\right)\right]G(u), \\
S_4 &= \frac{-ik_0^3}{4\pi}\left[\hat{\alpha}_{11}\sin\varphi\cos\varphi+\hat{\alpha}_{12}\left(\sin^2\varphi-\cos^2\varphi\right)\right. \\
&\quad \left.-\hat{\alpha}_{22}\sin\varphi\cos\varphi\right]G(u),
\end{aligned}
$$

where (θ, φ) are the angles of the spherical coordinate system connected with the system (x, y, z), $k_0 = 2\pi/\lambda$ is the wavenumber in vacuum, and λ the wavelength of incident radiation.

The elements of the complex polarizability tensor in the laboratory system (x, y, z) are

$$
\hat{\alpha}_{ik} = \hat{\alpha}_{ki} = \sum_{j=1}^{3} \tilde{\alpha}_j a_{ji} a_{jk}, \tag{3}
$$

where $\tilde{\alpha}_j$ is the polarizability in the particle system (X, Y, Z), a_{ik} are the elements of the matrix of transition between the coordinate systems (x, y, z) and (X, Y, Z).

For homogeneous ellipsoids, the polarizabilities along each axis of the system (X, Y, Z) can be presented as

$$
\tilde{\alpha}_j = V(\varepsilon - 1)K_j, \quad K_j = \frac{1}{1 + L_j(\varepsilon - 1)}, \tag{4}
$$

where $j = 1$, 2, 3, and V is the volume of the particle. The case of inhomogeneous (multi-layered) particles is considered in the Appendix.

The form factor for ellipsoids is (Volkovitsky et al., 1984)

$$
G(u) = \frac{3}{u^3}(\sin u - u\cos u), \tag{5}
$$

where

$$
u = 2k_0 \sin\frac{\theta}{2}\sqrt{a^2(\vec{i}_{\rm X}, \vec{i}_{\rm bi})^2 + b^2(\vec{i}_{\rm Y}, \vec{i}_{\rm bi})^2 + c^2(\vec{i}_{\rm Z}, \vec{i}_{\rm bi})^2}, \tag{6}
$$

and $(\vec{i}_{\rm X}, \vec{i}_{\rm bi})$, $(\vec{i}_{\rm Y}, \vec{i}_{\rm bi})$, $(\vec{i}_{\rm Z}, \vec{i}_{\rm bi})$ are cosines of the angles between the particle (semi)axes and the bisector of the angle additional to scattering one.

The scattering cross-sections are determined by integrals of square amplitudes of the scattered radiation over all solid angles

$$
\begin{aligned}
C_{\rm sca,x} &= \frac{k_0^4}{16\pi^2}\int_0^{2\pi}\int_0^{\pi}\left[\left|\hat{\alpha}_{11}\sin\varphi-\hat{\alpha}_{12}\cos\varphi\right|^2\right. \\
&\quad +\left|\left(\hat{\alpha}_{11}\cos\varphi+\hat{\alpha}_{12}\sin\varphi\right)\cos\theta\right. \\
&\quad \left.\left.-\hat{\alpha}_{13}\sin\theta\right|^2\right]G^2(u)\sin\theta\, d\theta\, d\varphi, && (7) \\
C_{\rm sca,y} &= \frac{k_0^4}{16\pi^2}\int_0^{2\pi}\int_0^{\pi}\left[\left|\hat{\alpha}_{22}\cos\varphi-\hat{\alpha}_{12}\sin\varphi\right|^2\right. \\
&\quad +\left|\left(\hat{\alpha}_{22}\sin\varphi+\hat{\alpha}_{12}\cos\varphi\right)\cos\theta\right. \\
&\quad \left.\left.-\hat{\alpha}_{23}\sin\theta\right|^2\right]G^2(u)\sin\theta\, d\theta\, d\varphi. && (8)
\end{aligned}
$$

The absorption cross-sections are defined by volume integrals of square amplitudes of the internal field and therefore the expressions for the cross-sections are obviously equivalent to those of the Rayleigh approximation

$$
\begin{aligned}
C_{\rm abs,x} &= k_0\,{\rm Im}\left(\tilde{\alpha}_1 a_{11}^2 + \tilde{\alpha}_2 a_{21}^2 + \tilde{\alpha}_3 a_{31}^2\right), \\
C_{\rm abs,y} &= k_0\,{\rm Im}\left(\tilde{\alpha}_1 a_{12}^2 + \tilde{\alpha}_2 a_{22}^2 + \tilde{\alpha}_3 a_{32}^2\right), && (9)
\end{aligned}
$$

where $\tilde{\alpha}$ is the polarizability in the particle system (X,Y,Z).

3. NUMERICAL RESULTS

We have applied the quasi-static approximation to several kinds of particles. Their size is characterized by $x_{\rm V} = 2\pi r_{\rm V}/\lambda$, where $r_{\rm V}$ is the radius of the equal volume sphere.

3.1 Homogeneous ellipsoids

Cross-sections of homogeneous ellipsoids of various sizes, shapes, and compositions were calculated using the quasi-static and Rayleigh approximations and the DDScat code of Draine and Flatau (1999).

Our results confirm the main conclusion drawn by Voshchinnikov and Farafonov (2000) from a consideration of homogeneous spheroids that for the particles with the ratio of the maximum to minimum dimension (in our case a/c and b/c) exceeding $\sim$3–6, the quasi-static approximation is usually preferable to the Rayleigh approximation and the difference of the approximations sharply grows with an increasing asphericity of scatterer (see Fig. 1 as an illustration).

In general, the range of applicability of the quasi-static approximation outlined in the paper of Voshchinnikov and Farafonov (2000) can be used for rough estimates of that for homogeneous ellipsoids. However, we have revealed some differences (e.g., a more complex dependence on refractive index, orientation, etc), and a systematic study of ellipsoids in a really wide region of their parameter values is required to make the range of the applicability better defined.

3.2 Multi-layered ellipsoids

We modified the code of Draine and Flatau (1999) to treat ellipsoids with three and more layers. After that computations were made for multi-layered confocal ellipsoids of different shapes and structure. Some results are presented in Fig. 1 and Table 1.

Note that for confocal ellipsoids the aspect ratios of the boundaries between layers are larger than the ratio for the particle surface. Therefore, it becomes clear why in Fig. 1 the three-layered ellipsoid with the aspect ratio 4:2:1 shows a larger difference between the quasi-static

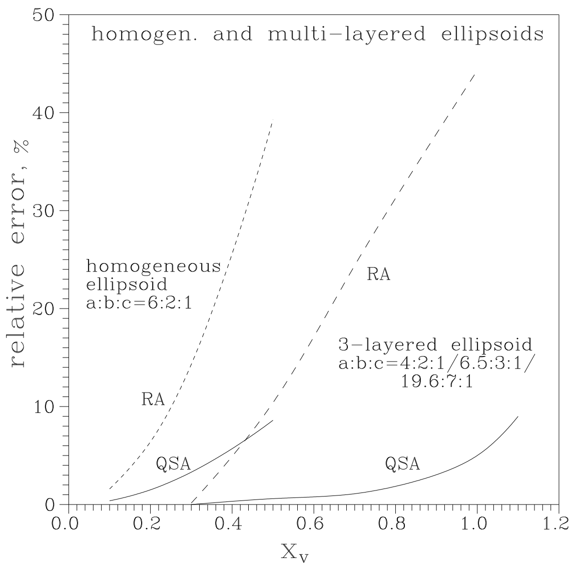

Figure 1: Relative errors of the quasi-static (QSA) and Rayleigh (RA) approximations for a homogeneous ellipsoid with a:b:c = 6:2:1 and m = 1.5 and a three-layered confocal ellipsoid with $(a_i$:b_i:$c_i, m_i)$ = (4:2:1, 1.33 / 6.5:3:1, 1.7+0.03i / 19.6:7:1, 1). The results obtained with the (modified) DDScat code were assumed to be exact.

and Rayleigh approximations than the homogeneous ellipsoid with the ratio 6:2:1.

In Table 1 we present results for another three-layered ellipsoid that seems to be relatively close to that used in Fig. 1. The difference of the quasi-static and Rayleigh approximations in this case is not so remarkable. It is obvious that one needs to consider a larger set of layered particles (in particular with different composition of their layers) in order to reach a full understanding of the range of applicability of the quasi-static approximation.

Table 1: Relative errors of the quasi-static (QSA) and Rayleigh (RA) approximations for three-layered confocal ellipsoids of different size parameter x_V. The parameters of the layers (i =1,2,3) are $(a_i$:b_i:$c_i, m_i, V_i/V)$ = (5:2:1, 1.33+0.01i, 0.56 / 9.85:3.6:1, 1.7+0.03i, 0.23 / 19.6:7:1, 1, 0.21).

x_V	0.25	0.3	0.35	0.4	0.45	0.5
QSA	0.36	2.1	4.2	6.3	8.6	10.9
RA	3.7	7.0	11.1	15.5	20.7	26.3

3.3 Core-mantle spheroids

We considered core-mantle spheroids separately as in this case we can base our approximate methods' comparison on a Separation of variable method (SVM) code of Farafonov et al. (1996) that is much more exact than DDScat. That allowed us to consider the region of (very) small values of the size parameter.

In Fig. 2 we present relative errors for a two-layered confocal spheroid. A higher accuracy of the quasi-static approximation relative to the Rayleigh one is clearly seen through the whole range of x_V values considered.

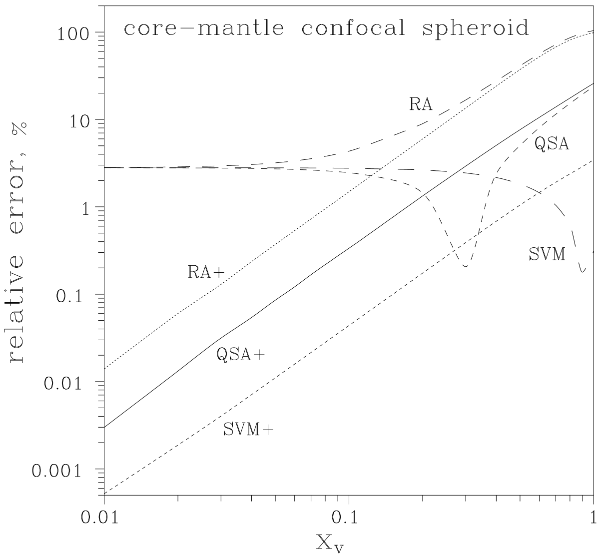

Figure 2: Relative errors of the quasi-static (QSA+) and Rayleigh (RA+) approximations for a two-layered confocal spheroid (mantle a:b = 4:1, m = 1.33 + 0.01i; core a:b = 6.7:1, m = 1.7 + 0.03i; the volume fraction of the core is 33%). The results obtained with the SVM code for core-mantle spheroids (Farafonov et al., 1996) were considered to be exact. SVM+ denotes the data derived for a homogeneous spheroid with the effective permittivity according to Eq. (10); RA, QSA and SVM show results for homogeneous spheroids with the effective permittivity according to the standard Bruggeman mixing rule of the Effective Medium Theory (see Bohren and Huffman, 1983).

On the other hand, it should be noted that Eqs. (10)–(12) (see the section A.1 below) can be considered as a specific rule of the Effective Medium Theory (EMT) for layered particles. Figure 2 demonstrates that this rule can give an effective refractive index that allows one to get very high accuracy results (for x_V <0.1) for multi-layered particles using more simple codes for homogeneous ones (see the curve SVM+ in Fig. 2) in particular in contrast to the commonly used Bruggeman rule of the EMT (see the curves marked by SVM, QSA, RA).

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APPENDIX

A.1 Confocal ellipsoids

Let us consider a n-layered confocal ellipsoid (i.e. for each two layers, the semiaxes obey the rule $a_i^2 - a_{i-1}^2 = b_i^2 - b_{i-1}^2 = c_i^2 - c_{i-1}^2, i = 2, 3, ..., n$).

We shall call by the quasi-static approximation for such particles all the formulae including Eq. (4) where the permittivity ε is replaced by the permittivity $\bar{\varepsilon}$ obtained by Farafonov (2000) in the Rayleigh approximation

$$\bar{\varepsilon} = A_2/A_1, \tag{10}$$

where A_1 and A_2 are determined from the matrix equation

$$\begin{pmatrix} A_1 \\ A_2 \end{pmatrix} = \begin{pmatrix} 1 & L_j^n \\ \epsilon_n & \epsilon_n(L_j^n - 1) \end{pmatrix} \cdot \Psi \cdot \begin{pmatrix} (\epsilon_1 - 1)L_j^1 + 1 \\ -(\epsilon_1 - 1)\delta_1 \end{pmatrix} \tag{11}$$

and the matrix Ψ is

$$\Psi = \prod_{i=2}^{n-1} \begin{pmatrix} (\epsilon_i - 1)L_j^i + 1 & \frac{(\epsilon_i - 1)}{\epsilon_i} L_j^i (L_j^i - 1) \\ -(\epsilon_i - 1)\delta_i & -(\epsilon_i - 1)(L_j^i - 1) + 1 \end{pmatrix}. \tag{12}$$

Here $\epsilon_i = \varepsilon_i/\varepsilon_{i+1}$ is the relative permittivity, $\delta_i = (a_i b_i c_i)/(a_n b_n c_n)$ the ratio of the volume of i-th ellipsoidal surface to the volume of the particle V. For more details, we refer a reader to the paper of Farafonov (2000).

It should be emphasized that in this case we actually have a combination of the quasi-static approximation with a specific rule of the Effective Medium Theory.

A.2 Non-confocal ellipsoids

The Rayleigh approximation for multi-layered non-confocal ellipsoids was considered in the paper of Farafonov (2001) recently submitted to *the Optics & Spectroscopy*.

The paper gives more complex expressions for A_1 and A_2 but their general structure remains similar to that shown above in the section A.1. The only restriction of the model used in that paper is that the a_i, b_i and c_i axes of all the ellipsoidal surfaces between the layers should be parallel.

We have not yet carried out any calculations for multi-layered non-confocal ellipsoids, but plan them in the near future.

A.3 Applicability to scattering matrix

The above-mentioned analysis of the applicability of the quasi-static approximation made by Voshchinnikov and Farafonov (2000) was based on a consideration of the scattering cross-sections of homogeneous spheroids for an axial incidence of radiation. We extended it by study of all the elements of the scattering matrix for oblique incidence of radiation on homogeneous spheroids.

It was found that not all results of the authors were applicable to the scattering matrix elements. The main conclusion that the larger is the aspect ratio the better works the quasi-static approximation is in principle correct. But for the elements it may not be true for scattering angles outside the forward (and backward) peaks.

It can be also mentioned that the dependence of accuracy of different scattering matrix elements on the parameters is in general similar, with F_{34} having practically always the lowest accuracy. These and other results of our consideration will be presented in a paper of Prokopjeva et al. (2000) preparing for publication in *the Optics & Spectroscopy*.