An algorithm of scattering matrix calculation by core-shell spherical particles

The theory of light scattering by spherical particles with a shell was established in a long-standing work by Aden and Kerker [1]. Subsequently, efforts of researchers were directed to the development of efficient methods for calculating expansion coefficients of the scattered field in the vector spherical harmonics (see, for example, the papers [2-6] and references there). However, until now the light scattering by core-shell spherical particles is an open computational problem, especially in the case of large strongly absorbing particles. The algorithm of calculation of the expansion coefficients of the scattered field described below shows high accuracy of calculation of these coefficients in a wide range of particle parameters.

The expansion coefficients a_n and b_n of the scattered field in the vector spherical harmonics can be represented as (see, for example, [6,7]):

$$a_n = \frac{\psi_n(x_s)\Psi_n'(A_n) - m_s\psi_n'(x_s)\Psi_n(A_n)}{\xi_n(x_s)\Psi_n'(A_n) - m_s\xi_n'(x_s)\Psi_n(A_n)}, \tag{1}$$

$$b_n = \frac{m_s\psi_n(x_s)\Psi_n'(B_n) - \psi_n'(x_s)\Psi_n(B_n)}{m_s\xi_n(x_s)\Psi_n'(B_n) - \xi_n'(yx_s)\Psi_n(B_n)}, \tag{2}$$

where $x_s = ka_s$, $k = 2\pi/\lambda$, λ is the wavelength of the incident light, a_s is the radius of entire particle, $\psi_n(x_s)$, $\xi_n(x_s)$ are the Riccati-Bessel functions of the first and third kind respectively and their derivatives are $\psi_n'(x_s)$, $\xi_n'(x_s)$, $\xi_n(x_s) = \psi_n(x_s) + \mathrm{i}\chi_n(x_s)$, $\chi_n(x_s) = x_s y_n(x_s)$, and $y_n(x_s)$ is the spherical Bessel functions of the second kind [7], $\mathrm{i} = \sqrt{-1}$.

$$\Psi_n'(X_n) = \psi_n'(m_s x_s) - X_n\xi_n'(m_s x_s), \tag{3}$$

$$\Psi_n(X_n) = \psi_n(m_s x_s) - X_n\xi_n(m_s x_s), \tag{4}$$

$$A_n = \frac{m_s\psi_n(m_s x_c)\psi_n'(m_c x_c) - m_c\psi_n'(m_s x_c)\psi_n(m_c x_c)}{m_s\xi_n(m_s x_c)\psi_n'(m_c x_c) - m_c\xi_n'(m_s x_c)\psi_n(m_c x_c)}, \tag{5}$$

$$B_n = \frac{m_c\psi_n(m_s x_c)\psi_n'(m_c x_c) - m_s\psi_n'(m_s x_c)\psi_n(m_c x_c)}{m_c\xi_n(m_s x_c)\psi_n'(m_c x_c) - m_s\xi_n'(m_s x_c)\psi_n(m_c x_c)}. \tag{6}$$

Here $x_c = ka_c$, a_c is the radius of the core, m_s and m_c are the complex refractive index of the shell and core respectively, $m_{s,c} = \mathrm{Re}(m_{s,c}) + \mathrm{i}\cdot\mathrm{Im}(m_{s,c})$. Coefficients (5) and (6) are the Lorenz–Mie coefficients of a core particle immersed in an medium with refractive index m_s.

To perform numerical calculations, it is convenient to reduce Eqs. (1)-(2) to the form:

$$a_n = \frac{\left(\frac{\widehat{D}_n}{m_s} + \frac{n}{x_s}\right)\psi_n(x_s) - \psi_{n-1}(x_s)}{\left(\frac{\widehat{D}_n}{m_s} + \frac{n}{x_s}\right)\xi_n(x_s) - \xi_{n-1}(x_s)}, \tag{7}$$

$$b_n = \frac{\left(m_s \hat{G}_n + \frac{n}{x_s}\right)\psi_n(x_s) - \psi_{n-1}(x_s)}{\left(m_s \hat{G}_n + \frac{n}{x_s}\right)\xi_n(x_s) - \xi_{n-1}(x_s)}, \tag{8}$$

where

$$\hat{D}_n = \frac{D_n(m_s x_s) - \hat{A}_n H_n(m_s x_s)}{1 - \hat{A}_n}, \tag{9}$$

$$\hat{G}_n = \frac{D_n(m_s x_s) - \hat{B}_n H_n(m_s x_s)}{1 - B_n}, \tag{10}$$

$$\hat{A}_n = W_n(m_s, x_s, x_c) \cdot \frac{\hat{m} D_n(m_c x_c) - D_n(m_s x_c)}{\hat{m} D_n(m_c x_c) - H_n(m_s x_c)}, \tag{11}$$

$$\hat{B}_n = W_n(m_s, x_s, x_c) \cdot \frac{D_n(m_c x_c) - \hat{m} D_n(m_s x_c)}{D_n(m_c x_c) - \hat{m} H_n(m_s x_c)}. \tag{12}$$

Here $\hat{m} = m_s/m_c$, $D_n(z)$ and $H_n(z)$ are the logarithmic derivatives of Riccati–Bessel functions:

$$D_n(z) = \frac{\psi_n'(z)}{\psi_n(z)}, \tag{13}$$

$$H_n(z) = \frac{\xi_n'(z)}{\xi_n(z)}, \tag{14}$$

and function $W_n(m_s, x_s, x_c)$ is

$$W_n(m_s, x_s, x_c) = \frac{\xi_n(m_s x_s)}{\xi_n(m_s x_c)} \cdot \frac{\psi_n(m_s x_c)}{\psi_n(m_s x_s)}. \tag{15}$$

Functions (7)-(15) are limited at all values of the parameters x_s, x_c, m_s, m_c [2,3,6]. The basic algorithms and methods for their calculation are considered in many works (see, for example, [2-6] and references there). It has been shown [2,6] that for functions of real arguments $\psi_n(x_s)$ and $\xi_n(x_s)$ in Eqs. (7), (8) the upward recurrence relation is stable

$$\eta_{n+1}(x) = \frac{2n+1}{x}\eta_n(x) - \eta_{n-1}(x). \tag{16}$$

Here $\eta_n(x)$ is one of the functions $\psi_n(x_s)$ or $\xi_n(x_s)$. We use this relation to calculate the Riccati-Bessel functions of the first kind $\psi_n(x)$ and the function $\chi_n(x)$. The function $\xi_n(x)$ is then defined as $\xi_n(x) = \psi_n(x) + \mathrm{i}\chi_n(x)$. The starting values of these functions are: $\psi_0(x) = \sin(x)$, $\psi_1(x) = \psi_0(x)/x - \cos(x)$ and $\chi_0(x) = -\cos(x)$, $\chi_1(x) = \chi_0(x)/x - \sin(x)$. Functions $H_n(z)$ of real and complex argument can also be obtained by upwards recurrence using the relations [2,3,5-7]

$$H_n(z) = -\frac{n}{z} + \frac{1}{\frac{n}{z} - H_{n-1}(z)} \tag{17}$$

starting from $H_0(z) =$ i.

When computing the functions $\psi_n(z_1)/\psi_n(z_2)$ and $\xi_n(z_2)/\xi_n(z_1)$, the upward recurrence relations [2,3,5,6] are also stable. It is convenient to represent these relations in the form

$$\frac{\psi_n(z_1)}{\psi_n(z_2)} = \frac{\psi_0(z_1)}{\psi_0(z_2)} R_n(z_1, z_2), \tag{18}$$

$$\frac{\xi_n(z_2)}{\xi_n(z_1)} = \frac{\xi_0(z_2)}{\xi_0(z_1)} F_n(z_2, z_1). \tag{19}$$

where $z_1 = m_s x_c$ and $z_2 = m_s x_s$ Both functions $R_n(z_1, z_2)$ and $F_n(z_2, z_1)$ are calculated using the upward recurrence relations

$$R_n(z_1, z_2) = R_{n-1}(z_1, z_2) \frac{\left[\frac{n}{z_2} + D_n(z_2)\right]}{\left[\frac{n}{z_1} + D_n(z_1)\right]}, \tag{20}$$

$$F_n(z_2, z_1) = F_{n-1}(z_2, z_1) \frac{\left[\frac{n}{z_1} + H_n(z_1)\right]}{\left[\frac{n}{z_2} + H_n(z_2)\right]} \tag{21}$$

beginning with $R_0(z_1, z_2) = F_0(z_2, z_1) = 1 + \mathrm{i}0$. With this representation of these functions, the analysis of the coefficients $\hat{A}_n$ and $\hat{B}_n$ in relations (11), (12) becomes comparatively simple. The common factor for these coefficients has the form

$$W_n(m_s, x_s, x_c) = W_0(m_s, x_s, x_c) R_n(z_1, z_2) F_n(z_2, z_1), \tag{22}$$

where

$$W_0(m_s, x_s, x_c) = \exp[(\mathrm{i}(z_2 - z_1)] \frac{\sin(z_1)}{\sin(z_2)} = \frac{1 - \exp(-\mathrm{i}2z_1)}{1 - \exp(-\mathrm{i}2z_2)} \tag{23}$$

It follows from relations (15), (22) at $x_s \mathrm{Im}(m_s) \gg 1$ that

$$W_0(m_s, x_s, x_c) \cong \exp[-2\mathrm{Im}(m_s)(x_s - x_c)] \tag{24}$$

and when $\mathrm{Im}(m_s)(x_s - x_c) \gg 1$ then $W_0(m_s, x_s, x_c) \cong 0$. In this case, $\hat{A}_n = \hat{B}_n = 0$ and coefficients (9) and (10) take the form

$$\widehat{D}_n = \hat{G}_n = D_n(m_s x_s). \tag{25}$$

Equalities (25) are also satisfied in the case $\widehat{m} = 1$, which corresponds to a homogeneous spherical particle with parameters x_s and m_s.

In contrast to the functions mentioned above, the downward recurrence relation is stable for the function $D_n(z)$ [2,6]

$$D_{n-1}(z) = \frac{n}{z} - \frac{1}{\frac{n}{z} + D_n(z)} \tag{26}$$

beginning with $D_N(z) = 0$ [3,6] for some sufficiently large value of N.

The maximum values of the indices n_{max} when calculating the coefficients (7), (8), as well as the Riccati-Bessel functions of the first kind, are recommended to be selected in the form

$$n_{max} = \text{int}\left[|z| + 4.05|z|^{1/3} + p\right] \tag{27}$$

with a minimum value of $p = 2$ [8]. The expression for N has the same form, but p must be significantly larger (see below).

To calculate the scattering characteristics of particles, a FORTRAN code has been completed, in which the above algorithm to determine the values of the coefficients (7), (8) is used. The coefficient values were then used to calculate the scattering matrix, scattering efficiency factors q_{sca} and extinction efficiency factors q_{ext} [7, 9]. The code was tested in detail for $1 \le x_s \le 1000$ and selectively for $1000 \le x_s \le 2500$ in the range of refractive index $1.1 \le \text{Re}(m_s), \text{Re}(m_c) \le 2$ and $0 \le \text{Im}(m_s), \text{Im}(m_c) \le 2$ for different values of x_c. The following criteria were chosen for the correct operation of the code. For non-absorbing particles the single scattering albedo $\omega = q_{sca}/q_{ext}$ should satisfy the condition $|\omega - 1| < 10^{-6}$. For absorbing particles in depending on the parameters of the shell and core, starting from some sufficiently large value of x_s, the condition $0.5 < \omega < 1$ must be satisfied. Note that even for particles with the parameter $x_s \sim 100$, there may be cases when $q_{ext} < 2$ and $\omega < 0.5$, which is not unusual. All of the above conditions were guaranteed to be satisfied with the value $p = 6$ for indices n_{max} (27) and $p = 25$ for index N. In addition, for particles with $x_s \le 60$, the calculations results of the scattering matrix and efficiency factors were controlled using the code described in [10,11]. All these control calculations were very well consistent with each other.

The particle parameters required for the calculations are specified in the file "*File_Input.txt*". They are all described in detail here. The calculation results will be in the file "*Matrix.txt*", which contains the extinction and scattering parameters, the single scattering albedo, and the scattering matrix. The scattering matrix **S** is normalized, so that

$$\frac{1}{2}\int_0^{\pi} S_{11} \sin\vartheta \mathrm{d}\vartheta = 1. \tag{28}$$

1. A. L. Aden and M. Kerker, Scattering of electromagnetic waves from two concentric spheres. J. Appl. Phys. Vol. 22, 1242–1246 (1951). https://doi.org/10.1063/1.1699834.
2. O. B. Toon, T. P. Ackerman. Algorithms for the calculation of scattering by stratified spheres. Appl. Opt. Vol. 20,No 20. 3657-3660 (1981). https://doi.org/10.1364/AO.20.003657..

3. D. W. Mackowski, R. A. Altenkirch, and M. P. Menguc. Internal absorption cross sections in a stratified sphere. Appl. Opt. Vol. 29, No 10. 1551-1559. (1990). https://doi.org/10.1364/AO.29.001551.
4. K. A. Fuller. Scattering and absorption cross sections of compounded spheres. III. Spheres containing arbitrarily located spherical inhomogeneities. J. Opt. Soc. Am. A, Vol. 12, No. 5. 893-904 (1995). https://doi.org/10.1364/JOSAA.12.000893.
5. I. Gurvich, N. Shiloah, M. Kleiman. Calculations of the Mie scattering coefficients for multilayered particle with large size parameters. J. Quant. Spectrosc. Radiat. Transfer.Vol. 70, 433-440 (2001). https://doi.org/10.1016/S0022-4073(01)00021-8.
6. X. Jia. Calculation of auxiliary functions related to Riccati–Bessel functions in Mie scattering. Journal of Modern Optics. (2016). https://doi.org/10.1080/09500340.2016.1203462.
7. C.F. Bohren, D.R. Huffman. Absorption and scattering of light by small particles. New York: Wiley; (1983).
8. W. J. Wiscombe, Improved Mie Scattering Algorithms. Appl. Opt. Vol. 19, 1505-1509 (1980). https://doi.org/10.1364/AO.19.001505.
9. M.I. Mishchenko, L.D. Travis, A.A. Lacis. Scattering, absorption, and emission of light by small particles. Cambridge, UK: Cambridge University Press; (2002).
10. D.W. Mackowski, M.I. Mishchenko. A multiple sphere T-matrix Fortran code for use on parallel computer clusters. J Quant Spectrosc Radiat Transfer 112:2182–92. (2011). https://doi.org/10.1016/j.jqsrt.2011.02.019.
11. D.W. Mackowski. The extension of the Multiple Sphere T Matrix code to include multiple plane boundaries and 2-D periodic systems. J Quant Spectrosc Radiat Transfer 290: 108292. (2022). https://doi.org/10.1016/j.jqsrt.2022.108292