

A T-matrix method and computer code for randomly oriented, axially symmetric coated scatterers

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Received 26 April 2003; received in revised form 6 August 2004; accepted 6 August 2004

Abstract

A computer code is described for the calculation of light-scattering properties of randomly oriented, axially symmetric coated particles, in the framework of the T-matrix theory. The underlying mathematical background is outlined briefly and convergence procedures are discussed. After outlining the input–output interaction between user and code, benchmark results are presented for two distinct shapes: coated, centered spheroids and offset coated spheres.

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Keywords: T-matrix; Nonspherical scatterers; Coated particles; Fortran code

1. Introduction

The T-matrix method [1], based on the linearity of Maxwell's equations, is a widely used technique for the determination of light-scattering properties by nonspherical particles. While it is applicable to any kind of shape, most practical applications are centered on revolution (axisymmetric) particles, for it is then that the T-matrix divides itself into independent submatrices [2]. Further simplifications, like particles with a plane of symmetry, allow for a faster and more efficient calculation of light-scattering properties [3]. Finally, Mishchenko's averaging scheme [4], or an equivalent angle integration routine, allow for an orientation averaging, a situation typically

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encountered in real-life systems, ranging from atmospheric aerosols to colloidal suspensions to algal ocean particles.

The study of nonhomogeneous particles is the next logical extension. Peterson and Ström [5,6] showed in 1974 how the T-matrix can be generalized for multilayered objects. However, during the last 30 years, the number of applications dealing with coated nonspherical particles have been scarce. This can partly be explained by the fact that many applications do not need orientation averaging, so the T-matrix loses attraction as compared to alternative methods like point matching [7] or separation of variables [8]; in addition, the challenge of computing the T-matrix for coated particles is higher than for the homogeneous case.

There are, however, quite a few systems where the scattering particle is both layered and nonspherical. For example, algal and bacterial particles are known to be both nonspherical and nonhomogeneous, but the lack of adequate models force researches in fields such as oceanography to rely on simpler models, be it exact (Mie, Aden-Kerker) or approximate (Anomalous Diffraction, Geometric Optics) [9,10].

In the present paper, a method is outlined to calculate the T-matrix of a coated, axisymmetric, nonspherical particle. This paper is to be used as a “user manual” for a Fortran code (“LISA”) publicly available on the World Wide Web. Light-scattering results for two particle sets have also been included for benchmark purposes.

2. Theory

In the framework of the T-matrix approach [1], both incident $E_{\text{inc}}(r)$ and scattered $E_{\text{sca}}(r)$ electric fields are expanded in a series of vector spherical harmonics M_{mn} , N_{mn} [11]:

$$\begin{aligned} E_{\text{inc}}(r) &= \sum_{n=1}^{\infty} \sum_{m=-n}^n [a_{mn} Rg M_{mn}(kr) + b_{mn} Rg N_{mn}(kr)], \\ E_{\text{sca}}(r) &= \sum_{n=1}^{\infty} \sum_{m=-n}^n [p_{mn} M_{mn}(kr) + q_{mn} N_{mn}(kr)]. \end{aligned} \quad (1)$$

The linearity of Maxwell’s equations enables us to relate both scattered (p_{mn} , q_{mn}) and incident (a_{mn} , b_{mn}) field coefficients by means of a transition matrix (or T-matrix):

$$\begin{aligned} p_{mn} &= \sum_{n'=1}^{\infty} \sum_{m'=-n'}^{n'} [T_{mm'n'}^{11} a_{m'n'} + T_{mm'n'}^{12} b_{m'n'}], \\ q_{mn} &= \sum_{n'=1}^{\infty} \sum_{m'=-n'}^{n'} [T_{mm'n'}^{21} a_{m'n'} + T_{mm'n'}^{22} b_{m'n'}]. \end{aligned} \quad (2)$$

The T-matrix elements depend on the particle’s size, shape, composition and orientation, but not on the nature of the incident or scattered fields. They can, therefore, be calculated in the so-called natural reference frame (z axis along the revolution axis) and then be averaged for all incidence and scattering directions, which equals averaging on particle orientation. The T-matrix

can be written in compact notation as

$$T = -B * A^{-1} = -\begin{pmatrix} B_{11} & B_{12} \\ B_{21} & B_{22} \end{pmatrix} * \begin{pmatrix} A_{11} & A_{12} \\ A_{21} & A_{22} \end{pmatrix}^{-1} = \begin{pmatrix} T_{11} & T_{12} \\ T_{21} & T_{22} \end{pmatrix}. \tag{3}$$

Expressions for the calculation of the A and B matrix elements are detailed elsewhere [2,3,12] and will not be reproduced here. In the case of a homogeneous scatterer, the matrices depend on the inner and outer indices of refraction m_r , as well as on the particle shape.

For a two-layered particle with refractive indices m_1 (core) and m_2 (shell), the T-matrix can be calculated as [5]

$$T = -B * A^{-1} = -[B_2 + BB_2 * (-B_1 * A_1^{-1})] * [A_2 + AA_2 * (-B_1 * A_1^{-1})]^{-1} \tag{4}$$

with

- $-B_1 * A_1^{-1} = T_1$ is the T-matrix calculated for a particle with refractive index as m_1/m_2 for inner, as 1 for outer and incident radiation wave number as $k_0 m_2$. In other words, it is the T-matrix for the core alone, assuming that the surrounding medium has the same refractive index as the coating.
- A_2 and B_2 are calculated by setting refractive index as m_2 for inner and as 1 for outer, and an incident wavelength as k_0 . If calculated, the product $-B_2 * A_2^{-1}$ would be the T-matrix for a particle with no core.
- Matrices AA_2 and BB_2 are calculated in the same way as A_2, B_2 , except that the Bessel functions of the first kind with argument kr are replaced by Hankel functions with the same argument.

A homogeneous T-matrix code could be adapted to calculate first the (A_1, B_1) matrices for the inner layer, and then the (A_2, B_2, AA_2, BB_2) matrices for the outer layer. Once computed, the T-matrix elements can be used to calculate any light scattering of interest, either cross sections or scattering (Müller) matrix elements. For example, expressions for extinction and scattering cross sections are

$$C_{\text{ext}} = \frac{2\pi}{k^2} \text{Re} \sum_{n=1}^{\infty} \sum_{m=-n}^n (T_{mn}^{11} + T_{mn}^{22}),$$

$$C_{\text{sca}} = \frac{2\pi}{k^2} \sum_{i,j=1}^2 \sum_{n=1}^{\infty} \sum_{n'=1}^{\infty} \sum_{m=0}^{\min(n,n')} (2 - \delta_{m0}) |T_{mn}^{ij}|^2. \tag{5}$$

3. Convergence procedure

Theoretically, the number of expansion coefficients (Eq. (1)) is infinite, and so is the size of the T-matrix. In practical computations, the series expansions are truncated after a finite number of terms n_{max} , the m T-matrices have dimension $(2n_{\text{max}}) * (2n_{\text{max}})$. Enough terms must be included so that the light-scattering functions are calculated to within the desired accuracy; on the other hand, a value of n_{max} too high will result in numerical instabilities and accuracy problems in the T-matrix determinations, as well as in unnecessarily high CPU time and memory usage.

The procedure to estimate the value of n_{\max} in the case of a homogeneous scatterer is to calculate C_{ext} and C_{sca} for the azimuth value $m = 0$ to within the desired accuracy, i.e., with $|C_{\text{ext}}(n_{\max})/C_{\text{ext}}(n_{\max} - 1) - 1| < 0.1\Delta$ ($m = 0$), and likewise for C_{sca} [13]. In the case of a layered scatterer, the value of n_{\max} has to be set for the two sets of matrices corresponding to core (A_1, B_1) and coating (A_2, B_2, AA_2, BB_2). Failure to do so might yield matrices too large or too small in some cases, for instance particles with small cores.

It is also known that the number of terms needed for an accurate computation of the matrices is usually larger than those needed for calculation of light-scattering parameters. Therefore, it is advisable to use a second parameter $n_{2\max}$, which is usually obtained by setting the condition that $|C_{\text{ext}}(n_{2\max})/C_{\text{ext}}(n_{\max}) - 1| < 0.1\Delta$ ($m = 0$), and likewise for C_{sca} . A convergence subroutine obtains the proper values of n_{\max} for both core and coating, as well as $n_{2\max}$ for the coating (full particle); for a better calculation of the full T-matrix, $n_{2\max} = n_{\max}$ is chosen for the inner surface.

In both cases, the starting value for n_{\max} is set as $n = x + 4x^{1/3} + 2 - a$, where x is the dimensionless size parameter kr , and r is a characteristic size such as the radius of the equivalent-volume sphere. If $a = 0$ we can recognize the Wiscombe criterion for spherical particles; it is quite a tight condition—it calls for the sum in the calculation of the extinction efficiency to be accurate to within 14 decimal digits [14]—so a nonzero value of a is preferable.

The calculation of the matrix elements requires a numerical evaluation of surface integrals, which are calculated by using a Gauss quadrature with N_g points. For the calculation of the n_{\max} and $n_{2\max}$ values for both core and coating, the number of N_g is set as $G * n_{\max}$. The final value of N_g is obtained by computing the full T-matrix (m submatrices of dimension $2n_{\max} * 2n_{\max}$) for increasing values of N_g till convergence of the extinction and scattering cross sections have been achieved to within the desired accuracy: $|C_{\text{ext}}(N_g)/C_{\text{ext}}(N_g - G) - 1| < 0.1\Delta$ (all values of m). G is also a parameter to be carefully chosen. The values $a = 5, 10$ and $G = 2, 4$ have been successfully used by the author; the reader is encouraged to experiment with other values to obtain the desired accuracy for his/her particle system.

4. Particle shape

The current T-matrix code can be applied to axisymmetric particles. If they also have a symmetry plane perpendicular to the symmetry axis, additional simplifications are possible: half of the T-matrix elements equals zero, and the rest are calculated along half of the particle surface (in other words, N_g is halved), thereby decreasing natural T-matrix calculations fourfold [3].

Typically, particle size and shape is characterized by two parameters: the equivalent-volume radius r_{eq} , and a nonsphericity parameter. For a typical example of plane symmetry, the spheroidal shape is described by

$$r(\theta) = r_{\text{eq}} \varepsilon^{1/3} [\varepsilon^2 \cos^2 \theta + \sin^2 \theta]^{-1/2}, \quad (6)$$

where a and b are the spheroidal axes (a is the revolution axis), $\varepsilon = b/a$ is the eccentricity and θ is the polar angle.

Different shapes can be chosen for core and coating, the only practical limitation, from a logical point of view, being that the core should not protrude out of the coating. An offset coated sphere in which the centers of both core and shell are separated by a distance l has also been used. The

core is assumed to be centered on the origin of coordinates, and the outer surface can be described as by

$$r(\theta) = a[p \cos^2 \theta + (1 - p^2 \sin^2 \theta)^{1/2}], \quad (7)$$

where a is the outer radius, and $p = l/a$. By defining the core/particle size ratio $q = b/a$, it can be seen that the inner core remains inside the shell as long as the condition $p + q < 1$ is fulfilled. This composite particle has no planar symmetry. Offsetting both core and shell yields a coated sphere, so the results can be checked against the Aden-Kerker theory for coated spherical scatterers [15].

5. The code

A Fortran computer code (“LISA”) has been written and tested for coated, nonspherical particles. It is structured as a main routine and several subroutines for convergence evaluation, natural T-matrix calculation, and computation of the main functions and parameters needed (Gauss quadrature points, Hankel, Bessel and Wigner functions, LU multiplication and inversion, Clebsch–Gordan coefficients). It includes a small set of instructions, short explanations, and recommendations.

In addition to the above mentioned, the following parameters have to be set by the user prior to code compilation:

- $mr1$, $mr2$: Absolute refractive indices for core and coating,
- $ntope$, $ang(ntope)$: Angles to be considered in the calculation of scattering matrix elements,
- a , G : Sets the starting value of n_{max} ($x + 4x^{1/3} + 2 - a$) and the number of Gauss quadrature points ($N_g = G * n_{max}$),
- kr , $deri$: Functions describing the particle surface, $r(\theta)$, $dr/d\theta$,
- SIM : Computing simplifications derived from plane-symmetric particles can be achieved by setting $SIM = 1$. Otherwise, $SIM = 0$.

The following parameters are requested to be entered as the code is run:

- Δ (desired accuracy), or alternatively, fixed values for n_{max} , n_{2max} , N_g ,
- $Choice$: Sets the kind of input data for sizes. If equals one, input parameters are taken as the outer and the inner equivalent-volume-sphere size parameters; if equals one, they are taken as outer size and inner/outer size ratio $q = R_{int}/R_{ext}$,
- Inner and outer size parameters: (R_{int}, R_{ext}) or (R_{int}, q) , depending on the chosen value for $Choice$.

The following data are obtained as output:

- $q.dat$: File containing Q_{ext} , Q_{sca} , Q_{abs} efficiencies,
- $failures.dat$: File containing data about the correctness of several inequalities among the scattering matrix elements [16]. If all inequalities are satisfied to within a factor $\Delta/100$, $failures.dat$ is empty, otherwise it gives information on what inequality is violated, and for what angle(s),

- mm1.dat: File containing the scattering matrix elements $F_{11}(\theta)$, $F_{22}(\theta)$ [17],
- mm2.dat: File containing the scattering matrix elements $F_{33}(\theta)$, $F_{44}(\theta)$,
- mm3.dat: File containing the scattering matrix elements $F_{12}(\theta)$, $F_{34}(\theta)$.

All Müller matrix elements are normalized according to

$$\frac{1}{2} \int_0^\pi F_{11}(\theta) \sin \theta d\theta = 1. \quad (8)$$

LISA has undergone several checks in order to ensure its accuracy. Light scattering data have been calculated for homogeneous particles, such as spheres or spheroids, and results have been

Table 1

Computed values of extinction, scattering and absorption efficiencies Q_{ext} , Q_{sca} , Q_{abs} , and computational parameters for core and shell n_{max} , $n_{2\text{max}}$

| Set | Efficiencies | | | Core parameters | | Shell parameters | | |
|-----|------------------|------------------|------------------|------------------|-------|------------------|-------------------|-------|
| | Q_{ext} | Q_{sca} | Q_{abs} | n_{max} | N_g | n_{max} | $n_{2\text{max}}$ | N_g |
| 1 | 0.98776 | 0.94549 | 0.04227 | 16 | 68 | 22 | 20 | 92 |
| 2 | 3.11020 | 2.97956 | 0.13064 | 10 | 44 | 19 | 17 | 80 |

Table 2

Elements of the scattering matrix for Set 1 (coated spheroids)

| θ | $F_{11}(\theta)$ | $F_{22}(\theta)$ | $F_{33}(\theta)$ | $F_{44}(\theta)$ | $F_{12}(\theta)$ | $F_{34}(\theta)$ |
|----------|------------------|------------------|------------------|------------------|------------------|------------------|
| 0° | 73.0195 | 73.0140 | 73.0140 | 73.0086 | 0.0 | 0.0 |
| 30° | 1.3663 | 1.3642 | 1.3618 | 1.3628 | -0.0465 | 0.0630 |
| 60° | 0.0356 | 0.0352 | 0.0283 | 0.0286 | -0.0164 | -0.0036 |
| 90° | 0.0173 | 0.0165 | 0.0069 | 0.0075 | -0.0133 | 0.0011 |
| 120° | 0.0097 | 0.0091 | -0.0046 | -0.0040 | -0.0076 | -0.0016 |
| 150° | 0.0043 | 0.0042 | -0.0039 | -0.0038 | -0.0011 | -0.0003 |
| 180° | 0.0031 | 0.0028 | -0.0028 | -0.0025 | 0.0 | 0.0 |

Table 3

Elements of the scattering matrix for Set 1 (off-centered spheres)

| θ | $F_{11}(\theta)$ | $F_{22}(\theta)$ | $F_{33}(\theta)$ | $F_{44}(\theta)$ | $F_{12}(\theta)$ | $F_{34}(\theta)$ |
|----------|------------------|------------------|------------------|------------------|------------------|------------------|
| 0° | 82.9206 | 82.9205 | 82.9205 | 82.9205 | 0.0 | 0.0 |
| 30° | 4.3488 | 4.3483 | 4.3409 | 4.3404 | -0.0945 | 0.2041 |
| 60° | 0.1415 | 0.1415 | 0.1237 | 0.1237 | 0.0285 | -0.0486 |
| 90° | 0.0597 | 0.0597 | 0.0535 | 0.0535 | -0.0178 | -0.0063 |
| 120° | 0.0322 | 0.0321 | 0.0275 | 0.0275 | -0.0064 | -0.0053 |
| 150° | 0.0140 | 0.0136 | 0.0058 | 0.0060 | 0.0059 | -0.0070 |
| 180° | 0.0262 | 0.0253 | -0.0253 | -0.0244 | 0.0 | 0.0 |

compared to those from homogeneous T-matrix and Aden-Kerker models. For checking purposes, we also calculated LS data for a coated sphere where both core and shell are offset by the same amount and that gives a nonspherical particle from the code point of view, but it still represents a coated sphere whose LS properties can be calculated within the Mie or Aden-Kerker frameworks.

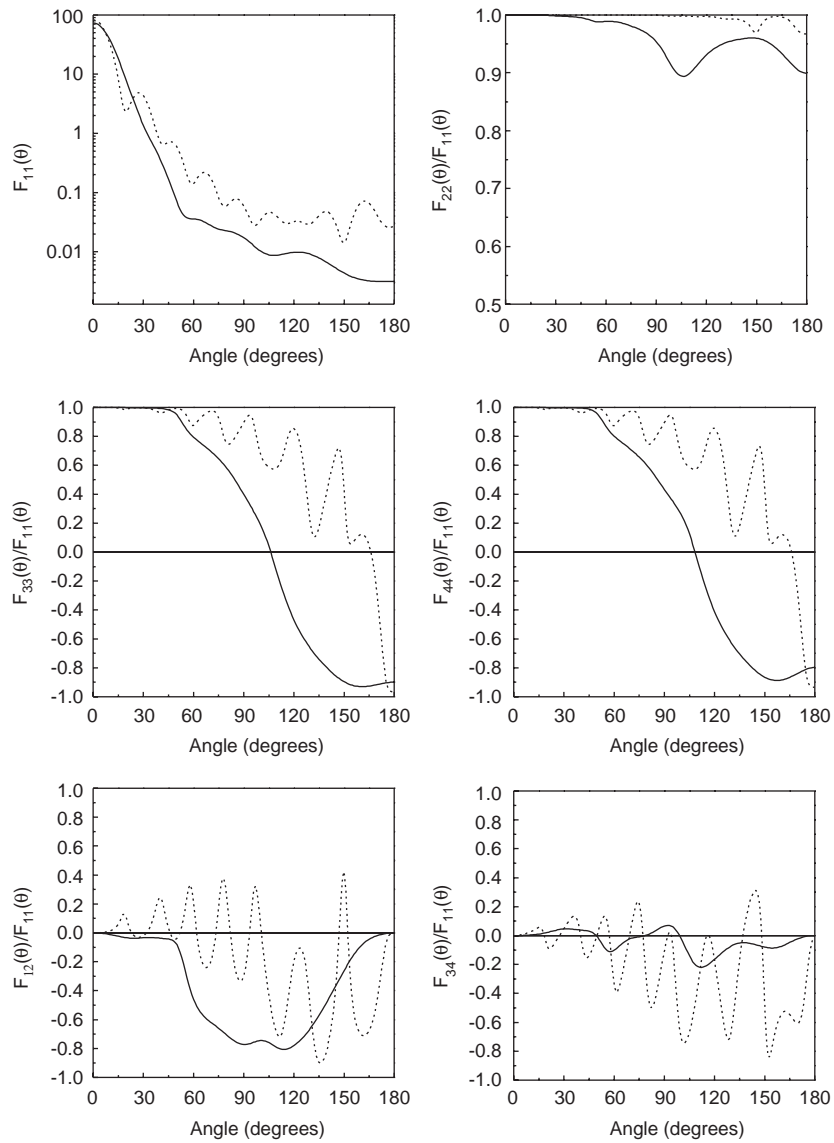


Fig. 1. Elements of the scattering matrix for Set 1 (coated spheroids, full line) and Set 2 (off-centered spheres, dashed line).

6. Benchmark examples

Results for two sets of nonspherical, randomly oriented coated particles are considered, and light-scattering computations have been made for an accuracy parameter $\Delta = 10^{-6}$:

- Set 1: Randomly oriented coated prolate spheroids with inner and outer refractive indices $m_1 = 1.2 + i0.01$, $m_2 = 1.05$, outer equivalent-volume-sphere size parameter $kr_{\text{eq}} = 10$, eccentricity $\varepsilon = 0.5$, and core/particle ratio $q = 0.5$,
- Set 2: Randomly oriented coated spheres with a centered core and an off-centered coating, for inner and outer refractive indices $m_1 = 1.5 + i0.05$, $m_2 = 1.2$, outer size parameter $kr_{\text{eq}} = 10$, an offset ratio $l/kr_{\text{eq}} = 0.3$, and core/particle ratio $q = 0.4$.

Table 1 shows the computed values of extinction, scattering, and absorption efficiencies, as well as computational parameters n_{max} , $n_{2\text{max}}$ for both inner and outer surface. Note that $n_{\text{max}} = n_{2\text{max}}$ for the inner surface. Tables 2 and 3 show the scattering matrix elements for several angles. A graphic representation is given in Fig. 1. The entire computation time for both sets was 10 and 7 s, respectively, on a Pentium IV computer.

Past experience shows that, whenever possible, it is preferable to make one's own code than try to adapt an existing one. However, having a proven code available is of great help to developers for checking and benchmarking purposes, as well as for the researcher with little or no programming expertise. It is hoped that LISA will help those who wish to use light-scattering techniques on nonspherical, nonhomogeneous bodies. LISA is made freely available on a nonprofit basis, on the condition that credit is properly acknowledged. It can be obtained by e-mail request at aquiran@ugr.es, or by downloading from <http://www.ugr.es/local/aquiran/codigos.htm>

Acknowledgements

Financial support from projects MAT2001-3803 (Ministry of Science and Technology, Spain; FEDER funds, EU) and INTAS (EU) 99-00510 is gratefully acknowledged.

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