On checking the calculations of the optical properties of non-spherical particles

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Abstract. All mathematical and computational problems of the light scattering theory for spherical particles (Mie theory) are well studied and resolved. However, they still remain to tackle in the case of non-spherical particles. This requires systematical testing of corresponding numerical algorithms and codes.

We consider different aspects of such checking: *(i)* reliability of the internal convergence test in view of our recent analytical and numerical investigations of some light scattering methods; *(ii)* efficiency of checking based on the reciprocity relation; *(iii)* new possibilities of comparison with other methods that are being opened by the developing WWW database of optical properties (DOP) of non-spherical particles. In particular, a general class of scatterers (called "weakly-Rayleigh scatterers") to which the internal convergence test should be applied with great care is determined. The reciprocity relation is shown to be a useful test (under some conditions), specifically for absorbing scatterers. The DOP will contain original codes and well checked (benchmark) results as well as information on the light scattering theory that can be helpful in researches, applications, and education.

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1. Introduction

There are many methods of solving the problem of light scattering by non-spherical particles [1]. Although both existence and uniqueness of solution to the problem have been proved (see, e.g., [2]), a careful mathematical analysis of even the most popular approaches – the separation of variables (SVM) and T-matrix (TMM) methods – is absent (even for convex axisymmetric particles). In such a situation systematic checking rather than a few particular individual tests is needed to assess the validity of a theoretical method/numerical code. It is well known that a solution may yield an acceptable result in certain cases but fail under other circumstances.

To test light scattering codes, one uses: consideration of internal convergence, relationships drawn from basic physical principles, and comparison with results obtained by other methods/codes.

1.1. Consideration of internal convergence

This approach utilizes "natural" expectation that results should converge with an increase of the number of terms in expansions of the fields or potentials (in the SVM and TMM), the number of knots in integral evaluations (TMM), the number of considered dipoles (in the discrete dipole approximation; DDA), etc.

This criterion can be used for absorbing particles and applied to intensity of radiation scattered in different directions (e.g., like in [3]), which is why it looks preferable to the most popular test based on the energy conservation principle (see section 1.2). However, it should be emphasized that such convergence does not guarantee that the results obtained are correct.

Another disadvantage of the criterion will be seen in section 2 where we demonstrate that the convergence can already be limited by the theoretical approach applied. Then an increase of corresponding parameters will generally not lead to a raise of accuracy of results.

1.2. Usage of physical principles

The energy conservation principle, together with the optical theorem, presents the most often used test. It states that for non-absorbing particles the cross-sections of extinction and scattering (usually calculated in a different way in codes/methods) must be equal [4]. If N digits coincide in the cross-sections, one can expect that N to $N-2$ digits in the results are true. Other tests of this kind (e.g., the relations connecting the scattering matrix elements) can be found in [1].

In this paper we attract attention to the reciprocity relation that is rather seldom considered in the literature but presents a very useful tool of checking (see section 3). The relation is applicable to absorbing particles (and hence well supplements the energy conservation test) and is efficient due to the usage of the differential characteristics of scattered radiation instead of the integral ones such as the scattering cross-section.

1.3. Comparison with other methods

Various theoretical approaches and numerical codes developed to calculate the light scattering by non-spherical particles are now available and can be used for checking. Comprehensive reviews of the methods and various computer codes are presented in [1] and on the website of Th. Wriedt [5], respectively.

Approximate approaches can give accurate results only in some restricted regions of parameter values, and the exact methods are certainly more useful. However, (freely available) codes realizing them are commonly very complicated and have often either insufficient or unclear documentation. This makes applicability of the criterion to some cases rather difficult (and frequently you should guess at accuracy of the test code as well).

Here, available "benchmark" results – the optical characteristics carefully calculated with different codes for a selected set of parameter values (e.g., [6, 7, 8]) – can help. Such benchmarks as well as a number of codes based on approximate and exact methods and developed by several groups will be (some already are) presented in the database of optical properties of non-spherical particles that we announce in section 4.

Thus, in this paper we will consider different aspects of light scattering codes' checking and start with a discussion of the results of our investigations of the basic methods.

2. Analytical and numerical investigations

Among the methods of solving the problem of light scattering by non-spherical particles, only a few ones – mainly the SVM for spheroids and the TMM – appear to be able to provide very accurate results (with 5–10 and more correct digits) for particles larger than the wavelength of incident radiation [1].

It is known (see, e.g., [8]) that if the shape of a scatterer deviates essentially from the spherical one, the classical versions of SVM [9] and TMM [3] work well only for particles small in comparison with the wavelength. Recently we have completed a very general analytical and numerical investigation of the TMM approach (and earlier that of the SVM). The technique used and the results obtained will be described in detail elsewhere [10]. Here we consider these results in the new context of the internal convergence criterion.

2.1. TMM-like methods

Our analytical investigation of infinite algebraic systems arising in the TMM shows that *any* TMM-like method, i.e. a method using expansions in terms of the spherical wave functions, has a limited range of applicability, and this range differs for the near and distant zones.

In the near zone (being considered when one studies phenomena close to a scatterer), the TMM is theoretically proved if the Rayleigh hypothesis is valid. In other words,

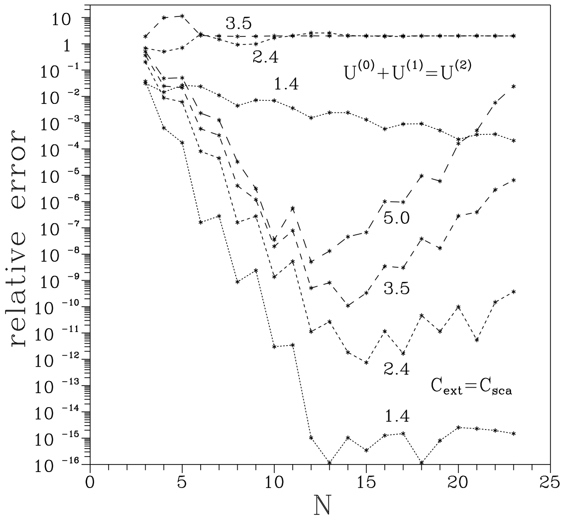

Figure 1. Relative errors of the energy conservation test $C_{\rm ext} = C_{\rm sca}$ and of the boundary condition for a scalar potential U at the particle surface point $(\theta, \varphi) = (45°, 0°)$ against the number of terms in expansions N. Oblate spheroids, the refractive index $m = 2$, the diffraction parameter $x_{\rm V} = 1$, radiation propagates at an angle of $\alpha = 45°$ to the particle symmetry axis. The numbers near the curves give the values of a/b.

the peculiarities of the analytical continuation of the scattered field must be located within a sphere being entirely inside the scatterer [10]. For instance, for spheroids, it occurs when the aspect ratio $a/b < \sqrt{2}$. Hereinafter, we call such particles "Rayleigh scatterers".

In the distant zone (calculations of cross-sections, phase function, etc.), the TMM is proved if the analytical continuations of the scattered and internal fields have a common ring-like region free of peculiarities and lying entirely inside the particles. We suggest to call such scatterers "weakly-Rayleigh" ones. For spheroids, they satisfy the condition $a/b < \sqrt{2} + 1$. For Chebyshev particles, which are non-convex particles in contrast to spheroids, the conditions are given and discussed in [11].

Figure 1 illustrates the conclusions of the analysis in the case of oblate spheroids. One can see that for weakly-Rayleigh (but not Rayleigh) particles ($a/b = 2.4$) there is no (internal) convergence of the potentials U of the fields near the particle surface (and hence the method can lead to incorrect results in the near zone). However, the cross-sections in this case are still calculated with high accuracy. For Rayleigh particles (a/b <1.4), the accuracy of calculations is only restricted by computational errors and the test of internal convergence can be applied without problems.

It should be noted here that in computer realizations of theoretical methods one solves truncated systems of algebraic equations. The properties of such finite systems differ, to some extent, from those of infinite systems we studied. Our numerical investigation [10] shows that for the distant zone relatively accurate results can be obtained for non-weakly-Rayleigh particles as well, if a rather small number of terms

N is kept in expansions of the fields (see curves for $a/b > 2.4$ in figure 1). If one increases N, the TMM algorithm becomes unstable and accuracy drops. The larger is $a/b > \sqrt{2} + 1$, the smaller is the particle size when the TMM gives acceptable results.

Thus, the test of internal convergence should be applied to non-Rayleigh scatterers and in particular to non-weakly-Rayleigh ones with great care.

2.2. SVM and other methods

Apparently, only for one version of the SVM for spheroids [12], a relatively full analytical investigation of the solution was made in our earlier paper [13]. It was found that this SVM version was more or less well mathematically grounded for spheroids of any degree of asphericity (i.e. of any aspect ratio a/b). Our qualitative consideration suggests that the same conclusion should be true for all other versions of the SVM. Although the classical schema of the SVM [9] is known to have efficiency very quickly decreasing with an increase of a/b, which makes it practically inapplicable to very elongated or flattened particles [12], something similar to a proper choice of potentials can make the efficiency nearly independent of a/b.

Thus, the test of internal convergence would be well applicable to the SVM, provided problems did not arise in another place – in calculation of the spheroidal wave functions (in particular, in obtaining eigenvalues for oblate particles or evaluating the radial spheroidal functions with a complex value of c and so on). This difficulty often limits applicability of the approach and requires careful checking of its numerical realizations.

As far as we know, other light scattering approaches besides the TMM and SVM are not well mathematically investigated either and can encounter unexpected problems in usage. For instance, a very popular and powerful numerical technique, DDA, while satisfying its validity criterion for interdipole spacing, significantly deviates from the rigorous solution and the experimental results in some cases of sphere aggregates [14]. Similar problems may occur for other volume integral equation approaches.

All of the preceding emphasizes once again the importance of careful checking of light scattering codes for non-spherical particles. For non-absorbing scatterers, one has a simple and powerful test based on the energy conservation. For absorbing ones, this test does not work and the reciprocity relation can in principle substitute it.

3. Reciprocity relation

Reciprocity is a manifestation of the symmetry of the light scattering process with respect to the inversion of time and directly follows from Maxwell's equations. For the amplitude matrix, the reciprocity relation is (see [1] for more details)

$$\begin{pmatrix} S_{11}(-\boldsymbol{n}', -\boldsymbol{n}) & S_{12}(-\boldsymbol{n}', -\boldsymbol{n}) \\ S_{21}(-\boldsymbol{n}', -\boldsymbol{n}) & S_{22}(-\boldsymbol{n}', -\boldsymbol{n}) \end{pmatrix} = \begin{pmatrix} S_{11}(\boldsymbol{n}, \boldsymbol{n}') & -S_{21}(\boldsymbol{n}, \boldsymbol{n}') \\ -S_{12}(\boldsymbol{n}, \boldsymbol{n}') & S_{22}(\boldsymbol{n}, \boldsymbol{n}') \end{pmatrix}, \quad (1)$$

where $\boldsymbol{n}, \boldsymbol{n}'$ are the unit vectors in the directions of the incident and scattered waves, respectively.

The relation (1) is applicable to absorbing particles and is efficient due to the usage of the differential characteristics of scattered radiation instead of the integral ones.

The relation is comprehensive when the directions of the incident and scattered waves and the axis of the axisymmetric particle do not lie in the same plane. In this case the amplitude matrix (1) is not diagonal, and the check involves the waves of both polarizations (TE and TM modes).

As an illustration, we consider the light scattering by axisymmetric particles. The problem is solved by a recently suggested method [15] that combines the TMM and a version of SVM for spheroids [12]. To simulate an approach of limited or unknown accuracy (hereinafter, "artificial approach"), we take an a priori small number of terms ($N = 8$) in the field expansions.

Two cases are compared: (1) radiation incident at an angle α_0 and scattered at a θ_0 angle to the particle axis, and (2) radiation incident at an angle of $\alpha = \pi - \theta_0$ and scattered at an angle of $\theta = \pi - \alpha_0$. According to the reciprocity relation, the amplitudes of scattered radiation should be the same in both cases, and if the azimuthal angle $\varphi = 0$, they are proportional to S_{11} and S_{22} for the TM and TE modes, respectively. In figures 2–4 we give the normalized differences of the amplitudes S_{ii} (i =1,2)

$$r.e. = \frac{|S_{ii}^{(1)} - S_{ii}^{(2)}|}{|S_{ii}^{(1)} + S_{ii}^{(2)}|} \tag{2}$$

where the superscripts (1) and (2) indicate the considered cases. Some other criteria mentioned in section 1 are also presented in the figures.

For non-absorbing spheroids, the normalized differences of S_{11} and S_{22} as functions of the angle $\pi - \theta_0$ are plotted in figure 2. The systematic one-digit difference of the TM (S_{11}) and TE (S_{22}) modes appears to be an artefact.

In our method [15] practically the same formulae are used to calculate the elements S_{ii} ($i = 1, 2$) and the cross-sections $C_{\rm sca}$. Therefore, comparing results with the SVM [12] code whose accuracy is known to be much better than 10^{-10}, we determined the errors of the $C_{\rm sca}$ (and hence S_{ii}) values obtained by the artificial method. The errors are presented in figure 2 vs. the radiation incidence angle α. One can see that the relative differences of the amplitudes (2) given by the reciprocity relation are of the same order of magnitude as the real relative errors of the calculations.

We also applied the energy conservation test to dielectric spheroids ($m = 1.33$) considered in figure 2 and found that the energy conservation and reciprocity relation criteria looked similar, without regard to details.

For highly absorbing spheroids, the behaviour of the reciprocity relation test is illustrated in figures 3–4, which are quite typical. Generally, one can conclude that the difference of $N_{\rm C}$, the number of correct digits in $C_{\rm sca}$ (obtained by comparison with SVM results), and $N_{\rm S}$, the number of coinciding digits in the elements $S_{ii} (i = 1, 2)$, is in the range $-1 \lesssim (N_{\rm S} - N_{\rm C}) \lesssim 2$, except for the nearly backscattering, that is close to

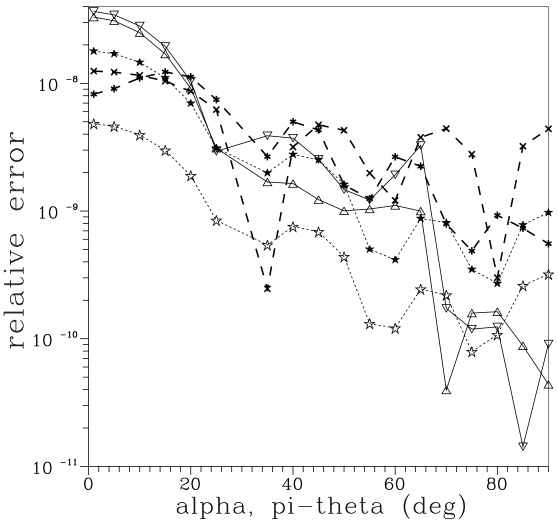

Figure 2. Relative differences of amplitude matrix elements S_{11} and S_{22} (dotted lines), relative errors of the cross-sections $C_{\rm abs} = C_{\rm ext} - C_{\rm sca}$ (dashed lines) and of $C_{\rm sca}$ (solid lines) as functions of the radiation incidence angle. Oblate spheroids, $a/b = 1.5$, $x_{\rm V} = 1$, $m = 1.33$, $\alpha_0 = 30°$ (see definition of the angles α, θ in text). TM modes are shown by triangles, crosses, open stars; TE modes by turndown triangles, asterisks, filled stars.

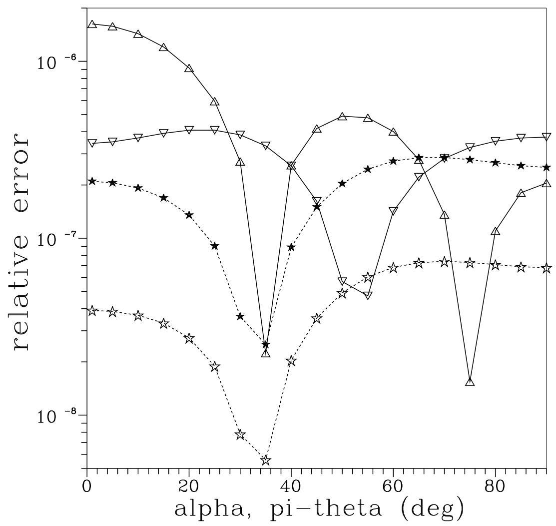

Figure 3. Relative differences of amplitude matrix elements S_{11} and S_{22} (dotted lines) and relative errors of $C_{\rm sca}$ (solid lines) vs. the radiation incidence angle. Oblate spheroids, $a/b = 1.5$, $x_{\rm V} = 1$, $m = 2.5 + 1.5{\rm i}$, $\alpha_0 = 33°$. Notations as in figure 2.

the peculiar case of $\alpha_0 = \pi - \theta_0$ when the test is satisfied automatically.

Thus, one can see that the reciprocity relation generally allows one to predict the number of correct digits in the cross-sections and is a really useful mean for checking results for absorbing particles.

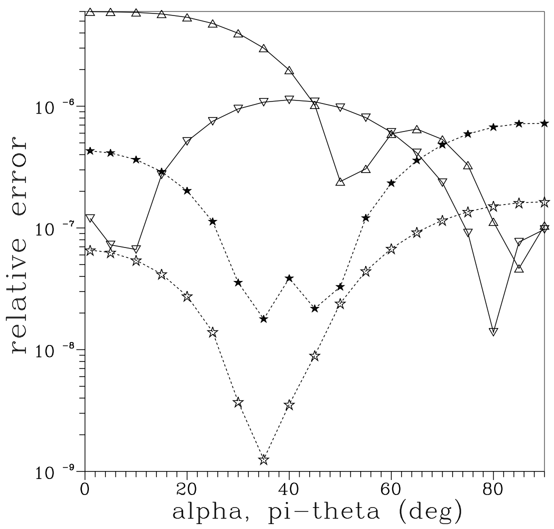

Figure 4. The same as in Fig.3, but for prolate spheroids.

4. The database of optical properties

We start collecting well checked optical characteristics (cross-sections, matrices, etc.) of non-spherical particles of various shapes, sizes, compositions, structures, etc. in the benchmark section of the database of optical properties (DOP) opened for free access at *http://www.astro.spbu.ru/DOP*.

The database is being created by several scientists from different institutions working within a joint INTAS project. Besides the benchmarks, the database will include: *(i)* codes developed in the institutions to treat the light scattering by non-spherical/inhomogeneous particles; *(ii)* a large graphic library of the optical properties obtained by theoretical and experimental methods for particles of different kinds; *(iii)* a collection of over 8000 references to papers on light scattering; *(iv)* links to related Internet resources; *(v)* some sophisticated software under construction (see [16] for more details). Some parts of the DOP are ready, e.g. the Jena-Petersburg database of optical constants (JPDOC) that contains over 700 references, data files and links to the Internet resources related to measurements and calculations of the optical constants in the range from X-rays to radio domain for silicates of different kinds, various ices, oxides, sulfides, carbides, carbonaceous species from amorphous carbon to graphite, etc. [17, 18].

The general aim of the DOP is to help investigators in solution of a wide spectrum of problems arising in scientific and industrial applications of light scattering theory and to serve as an educational tool for high schools.

5. Conclusions

Our analytical and numerical analyses of the T-matrix and separation of variables methods show that these and similar methods can give unreliable results and, in certain cases, the internal convergence test will not be applicable either.

The reciprocity relation is considered as an additional tool for testing results of light scattering calculations for non-spherical particles. The relation is found to be as simple in usage and informative as other tests. For absorbing particles, it substitutes effectively the well known energy conversation test. It is specifically helpful for solutions based on the separation of variables approach where calculations of the radial spheroidal functions for complex values of c are known to meet problems rather frequently.

We also announce the database of optical properties of non-spherical particles that is currently being developed and will be useful not only for testing codes but also for other applications of the light scattering theory.

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