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COMPUTATIONS OF SCATTERING MATRICES OF FOUR TYPES OF NON-SPHERICAL PARTICLES USING DIVERSE METHODS

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Abstract—The scattering matrix as a function of scattering angle has been computed for four different homogeneous particles: a prolate spheroid, an oblate spheroid, a finite cylinder and a bisphere with touching components. The directions of the incident and scattered light beams, as well as the rotation axis of each particle, lie in the same plane. The particles considered have the same refractive index, volume and orientation of the rotation axis with respect to the incident light. The computations were performed with the (superposition) T-matrix method, the separation of variables method for spheroids and the discrete-dipole approximation. The results are presented in the form of tables and graphs. Their usefulness as benchmark results and some other aspects are also discussed. Copyright © 1996 Elsevier Science Ltd

1. INTRODUCTION

It is well known that many small particles occurring in nature differ appreciably from homogeneous spheres. Consequently, the classical Lorenz–Mie theory[1,2] can not be used for accurate computations of light scattering properties of such particles. This has inspired many scientists to develop methods for computing light scattering by non-spherical particles (see, e.g., Refs. 3–19). In some of these methods certain basic features of the Lorenz–Mie theory, such as separation of variables, were retained, but in other approaches completely new paths were followed.

During a workshop on Light Scattering by Non-Spherical Particles, held in May 1995 in Amsterdam, the idea was born to tackle a few well-defined scattering problems with different methods and to compare the results. The main reasons for starting the project were that this would provide more insight into each of the methods and that some very reliable benchmark numbers could be created in such a way. The primary purpose of this paper is to present the results of the project.

The scattering problems considered are described in Sec. 2. The methods used to solve these scattering problems are the (superposition) T-matrix method, the separation of variables method for spheroids and the discrete-dipole approximation. These are briefly treated in Sec. 3. Section 4 contains the computational results in the form of tables and graphs. These results pertain to the elements of the scattering matrix as functions of the scattering angle for directions in the plane containing the direction of the incident light and the rotation axis of the particle. A short discussion is given in Sec. 5 and concluding remarks are presented in Sec. 6.

2. DESCRIPTION OF THE PROBLEMS

We consider four different scattering problems, namely light scattering by a prolate spheroid, an oblate spheroid, a finite circular cylinder and a bisphere with equal touching components. Each particle is homogeneous, made of some optically inactive substance and has a refractive index of 1.5–0.01 i. The size of each particle is such that $2\pi r/\lambda = 5$, where r is the radius of the equal-volume-sphere and λ is the wavelength in the surrounding medium. The rotation axis of each particle is defined in such a way that any rotation about this axis maps all points of the surface of the particle into the surface. The prolate and oblate spheroid are both obtained by rotation of an ellipse whose major axis is twice as large as its minor axis. The height of the cylinder is twice as large as the diameter of its circular cross section.

The scattering geometry of each particle is as follows. The particle is illuminated by a plane harmonic wave propagating along the positive z-axis of a Cartesian coordinate system (x, y, z). The origin of this coordinate system lies inside the particle (see Fig. 1). The rotation axis of the particle makes an angle $\Theta_p = 50°$ with the positive z-axis and lies in the x–z plane. The scattering angle, Θ, is the angle between the direction of the incident light and that of the scattered light. In this paper we only consider scattering directions in the x–z plane having non-negative x-components.

The scattering of light in a particular direction can be described by (Ref. 1)

$$\begin{pmatrix} I^{sc} \\ Q^{sc} \\ U^{sc} \\ V^{sc} \end{pmatrix} = \frac{F}{k^2R^2} \begin{pmatrix} I^{in} \\ Q^{in} \\ U^{in} \\ V^{in} \end{pmatrix}. \tag{1}$$

Here Stokes parameters are used for the incident (superscript "in") and scattered (superscript "sc") light and the scattering plane acts as the plane of reference for defining the Stokes parameters. Furthermore, $\boldsymbol{F}$ is the scattering matrix, $k = 2\pi/\lambda$, and R is the distance between the point of detection and the particle. The elements of $\boldsymbol{F}$ are written as F_{ij} with i,j = 1, 2, 3, 4. It should be noted that they are all dimensionless. For the four scattering problems considered in this paper, the elements F_{13}, F_{14}, F_{23}, F_{24}, F_{31}, F_{32}, F_{41} and F_{42} vanish identically. Furthermore, $F_{11} = F_{22}$, $F_{12} = F_{21}$, $F_{34} = -F_{43}$ and $F_{33} = F_{44}$ for all scattering angles. Therefore, we will only report computational results in this paper for the elements F_{11}, F_{21}, F_{33} and F_{43}.

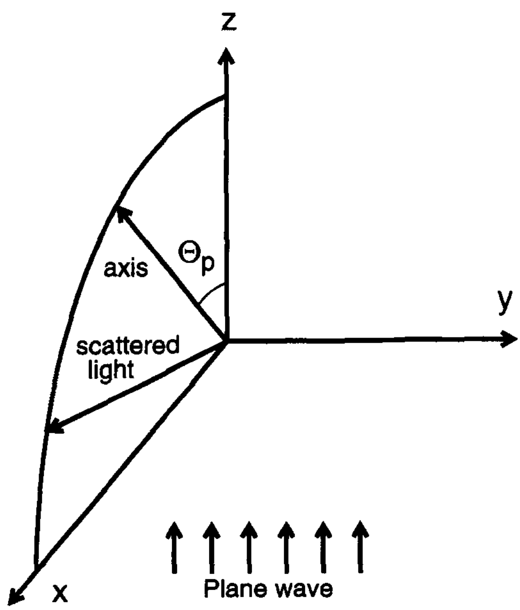

Fig. 1. A plane wave scattered by a non-spherical particle located at the origin of a Cartesian coordinate system. The (rotation) axis of the particle makes an angle Θ_p with the positive z-direction and lies in the x–z-plane, which also contains the direction of the scattered light.

Table 1. The elements F_{11}, F_{21}, F_{33} and F_{43} of the scattering matrix for various values of the scattering angle (in degrees). This table refers to the prolate spheroid described in the text.

Θ (deg)	F_{11}	F_{21}	F_{33}	F_{43}
0	654.032186	29.977488	651.970555	-42.353812
5	570.099227	26.236151	568.210226	-38.235212
10	416.303708	22.379107	414.231995	-34.925736
15	254.517251	19.537602	251.984936	-30.015082
20	129.014741	17.349058	125.951591	-21.909139
25	53.967561	14.659070	50.605928	-11.689715
30	20.630846	10.777187	17.448021	-2.247357
35	11.649349	6.125455	9.160858	3.776879
40	12.016994	1.947290	10.533918	5.445436
45	13.358025	-.467244	12.826912	3.699846
50	13.253511	-.437450	13.232194	.610945
55	12.621991	1.871544	12.376925	-1.619786
60	13.250280	5.597229	11.925697	-1.420807
65	16.286833	9.519488	13.092340	1.797473
70	21.702240	12.406708	16.080288	7.647560
75	28.427030	13.274932	20.183307	14.983535
80	34.846911	11.571469	24.243361	22.196119
85	39.374609	7.353046	27.180508	27.522946
90	40.882224	1.475275	28.343652	29.424772
95	38.907807	-4.373663	27.553418	27.120060
100	33.716280	-8.123074	24.936727	21.188743
105	26.332118	-8.392097	20.813578	13.774908
110	18.455097	-5.642785	15.756630	7.776773
115	11.968646	-2.261953	10.598905	5.078905
120	8.027310	-.921420	6.121562	5.110302
125	6.390260	-2.306316	2.641840	5.342005
130	5.797329	-4.490982	.063335	3.665528
135	5.165163	-4.896891	-1.604322	.354281
140	4.335211	-2.872681	-2.050105	-2.517702
145	3.642342	-.087487	-1.225073	-3.429023
150	3.130457	1.490084	.134539	-2.749784
155	2.557525	1.505215	.972050	-1.824932
160	1.979901	1.056556	.922133	-1.397630
165	1.844747	1.192896	.567927	-1.287459
170	2.388185	1.974793	.631563	-1.185222
175	3.219522	2.768502	1.142141	-1.181623
180	3.665305	2.997046	1.502259	-1.481686

3. METHODS OF SOLUTION

Three computational methods have been used to obtain numerical solutions in the form of tables and graphs (shown in the next section) for the scattering problems described in the preceding section. These methods are (i) the (superposition) T-matrix Method (TMM), (ii) The Separation of Variables Method for spheroids (SVM) and (iii) The Discrete-Dipole Approximation (DDA). In this section a brief description of each of these methods will be presented with references to other papers for further details.

(i) The T-matrix Method

The T-matrix approach for calculating light scattering by single non-spherical particles and the (superposition) T-matrix method for aggregated scatterers are reviewed in Ref. 8. It should be noted that in the case of aggregated spheres the (superposition) T-matrix method is equivalent to the separation of variables method.[20,21] Numerical aspects of computer calculations are discussed in Refs. 17, 22 and 23, while numerical accuracy checks of the TMM codes used in this study are described in detail in Ref. 17 for spheroids, Ref. 24 for bispheres, and Ref. 25 for cylinders. The parameter n_{max} specifying the maximum value of the index n in the expansion of the scattered electric field in vector spherical functions [see Eq. (6) of Ref. 8] was increased until all scattering matrix elements converged to within $\pm\Delta$, where Δ was equal to 10^{-6} for the spheroids and 10^{-4} for the cylinder and the bisphere. Although this internal convergence of T-matrix computations does not necessarily imply that T-matrix results are indeed accurate to within $\pm\Delta$, the comparison of internally converged T-matrix computations for spheroids with analogous SVM computations strongly suggests that for the particles under consideration internal convergence is also an excellent measure of the absolute accuracy of TMM as well as SVM computations.

T-matrix results for spheroids and bispheres were obtained using double-precision arithmetic, while convergent computations for cylinders required the use of extended-precision floating-point variables, as described in Ref. 22. The parameter n_{max} was equal to 23 for the prolate spheroid, 20 for the oblate spheroid, 79 for the cylinder, and 30 for the bisphere.

(ii) Separation of Variables Method for spheroids

This is the classical approach based on the light scattering theory of a plane linearly polarized electromagnetic wave which is scattered by a particle with any size and refractive index. In this method the vector wave equation is separated in a special coordinate system which is chosen in such a way that the surface of the particle coincides with one of the coordinate surfaces. Then the solution of the wave equation is expanded in terms of the corresponding wavefunctions, and the expansion coefficients are determined under the boundary conditions. The single-particle solutions by the SVM were obtained for spheres (Mie theory), infinitely long circular cylinders, homogeneous spheroids and spheroids having a core–mantle structure.

For homogeneous spheroidal particles, the solution was first developed by Asano and Yamamoto.[13] It is based on expansions of the radiation fields in the form of Debye potentials (as for spherical particles). Another approach was used by Farafonov,[14] (see also Ref. 26) who introduced a special basis for expansion of the electromagnetic fields: a combination of Debye and Hertz

Table 2. As Table 1, but for an oblate spheroid.

Θ (deg)	F_{11}	F_{21}	F_{33}	F_{43}
0	758.151698	-2.693769	757.186135	38.156226
5	741.327842	1.822435	740.770617	28.679981
10	643.920727	7.100306	643.784415	11.185498
15	488.665877	11.966287	488.466987	-7.151935
20	315.978622	15.230261	315.058005	-18.681061
25	167.601091	15.883501	165.687728	-19.632041
30	69.184670	13.541881	66.726587	-12.275939
35	22.249716	8.942556	20.148441	-3.020081
40	9.906180	3.908154	8.813401	2.276553
45	10.928921	.436690	10.724085	2.060248
50	11.639137	-.548136	11.586164	-.964288
55	8.784380	.295981	8.220722	-3.081795
60	4.903274	1.458333	3.799228	-2.735182
65	2.533494	1.838393	1.415124	-1.018000
70	1.871537	1.389537	1.241145	.177192
75	1.780490	.740772	1.617439	.072748
80	1.474315	.405614	1.250125	-.668034
85	1.039549	.380773	.169950	-.952256
90	.908869	.335628	-.784941	-.311872
95	1.260279	.014100	-.918094	.863253
100	1.899870	-.563533	-.167918	1.806582
105	2.507129	-1.183531	1.002748	1.969632
110	2.885247	-1.633155	1.996702	1.292530
115	3.025313	-1.824744	2.410565	.109553
120	3.020455	-1.797216	2.157741	-1.112348
125	2.953068	-1.647173	1.414964	-2.001327
130	2.839171	-1.460720	.486243	-2.385530
135	2.641826	-1.282542	-.334409	-2.285279
140	2.321617	-1.120201	-.857545	-1.843820
145	1.884915	-.963051	-1.037294	-1.244771
150	1.403657	-.796017	-.958690	-.646160
155	.997891	-.599450	-.784362	-.145678
160	.788261	-.340276	-.676967	.217447
165	.839114	.028370	-.721336	.427765
170	1.121989	.538256	-.877908	.445440
175	1.526847	1.141671	-.997111	.183355
180	1.926611	1.642682	-.902574	-.445853

Table 3. As Table 1, but for a cylinder.

Θ (deg)	F_{11}	F_{21}	F_{33}	F_{43}
0	748.9332	37.5036	747.4812	-27.6815
5	638.6521	28.9379	637.3573	-28.5455
10	441.3904	21.1998	439.5997	-33.5885
15	246.2887	17.5768	243.2641	-34.2307
20	110.3032	17.2234	105.7549	-26.1927
25	42.6053	16.8952	37.0739	-12.4617
30	21.6631	14.2745	16.2889	.4487
35	20.2613	9.5299	16.1415	7.6913
40	20.6450	4.4885	18.2874	8.4642
45	17.0194	.9818	16.1952	5.1392
50	11.1250	-.1370	11.0767	1.0265
55	6.8836	1.0245	6.6610	-1.4021
60	7.0144	3.8974	5.7256	-1.1083
65	11.9433	7.8116	8.8350	1.8883
70	20.2477	11.9286	14.6851	7.2129
75	29.7223	14.9271	21.2385	14.4751
80	38.3174	14.9718	26.8259	22.9006
85	44.5038	10.4771	30.5410	30.6278
90	47.1100	1.5231	31.8189	34.7072
95	45.1688	-9.1845	29.9344	32.5545
100	38.3524	-17.1733	24.3916	24.1048
105	27.8701	-18.9294	16.1213	12.5906
110	16.7569	-14.5024	7.9549	2.6822
115	8.5410	-7.5306	3.0548	-2.6281
120	4.8827	-2.3830	2.3149	-3.5782
125	4.4227	-.8926	3.5475	-2.4858
130	4.3343	-1.5874	3.7570	-1.4669
135	3.1224	-1.8904	2.1872	-1.1799
140	1.6090	-.9008	.6322	-1.1737
145	1.1999	.1001	.7233	-.9521
150	1.8581	-.2564	1.7294	-.6293
155	2.3169	-1.4107	1.7138	-.6640
160	1.9324	-1.5314	.3655	-1.1204
165	1.5273	-.0034	-.4351	-1.4640
170	2.1937	1.5824	.9006	-1.2237
175	3.7787	1.3022	3.4925	-.6212
180	4.9823	-.7983	4.9095	-.2861

potentials (a superposition of the approaches for spheres and infinitely long cylinders). The resulting solution is rather simple and can be implemented with a relatively small numerical code. From a computational point of view, it is more effective than the approach of Asano and Yamamoto, especially for large values of the ratios of major to minor axes (see Refs. 26 and 27 for details). In this paper, Farafonov's solution is used for calculations. The numerical calculations were performed on a PC-AT/486-50 with double precision.

(iii) The Discrete-Dipole Approximation

A new powerful method to compute, at least in principle, electromagnetic scattering by a particle of any shape and structure was developed by Purcell and Pennypacker.[12] In this discrete-dipole approximation (DDA) method a particle is modelled as a group of interacting dipoles. The dipole moments and thus the electric fields radiated by the dipoles are unknown. The total electric field at the dipole positions is determined by solving the system of linear equations for the individual dipole fields.

One difficult point in all DDA applications is the question of the correct expression for the polarizability of a dipole. This happens because we are dealing with finite sized spherical (hard core) dipoles which do not fill a particle completely. Lumme and Rahola,[19] chose to use for the polarizability the first three terms of the expansion of the Mie coefficient, a_1, in terms of the size parameter. The first term is equal to the classical Clausius–Mossotti relation and the third term gives the radiation reaction. The physical meaning of the second term, proportional to the fifth power of the size parameter, x_0, of a dipole, still lacks an interpretation.

The other approximation needed in the DDA is a relation between the refractive index of a dipole and that of the bulk of the medium. For this purpose an effective medium theory is needed. Lumme and Rahola,[19] showed that a semi-empirical model by Sihvola,[28] produced very good results in the case of spheres when compared to Mie theory. The relative errors of F_{11} never exceed a few per cent and the absolute errors of the other elements are typically less than a few times 0.01.

In our numerical computations we have used a new algorithm, the quasi-minimal residual method,[29] to solve the large systems of linear equations. This, together with using fast Fourier transform, makes our DDA code very efficient (see Rahola,[30] for details).

Among the users of the DDA there seems to be a consensus that fairly accurate results are obtained if the size parameter of a dipole x_0 is smaller than about 0.3. For our calculations we used $x_0 \approx 0.2$. The exact value depends on the particle considered and on the relation

$$2\pi r/\lambda = 5 = x_0\ (6N/\pi)^{1/3} \tag{2}$$

where N is the number of dipoles.[19] Since all four particles under consideration in this paper have an octagonal symmetry, N must be a multiple of 8. The combinations (x_0, N) used for the prolate spheroid, the oblate spheroid, the cylinder and the bisphere were, respectively, (0.20, 8320), (0.20, 8664), (0.22, 6656) and (0.20, 8448).

The other free parameter in our DDA code comes from a semi-empirical model for the effective medium theory which involves a parameter ν.[28] The best choice turned out to be $\nu = -0.2$, which is consistent with our earlier,[19] conclusion $\nu = -0.3 \pm 0.3$. Thus the effective refractive index for a dipole assumes the value $2.25 - 0.037i$.[19]

Table 4. As Table 1, but for a bisphere.

Θ (deg)	F_{11}	F_{21}	F_{33}	F_{43}
0	791.1861	26.6659	789.9327	-35.6460
5	667.5445	27.3329	665.9841	-36.5208
10	445.4292	24.2288	443.3876	-35.0357
15	231.6601	19.5437	229.1450	-27.8748
20	90.1434	14.7204	87.4396	-16.2315
25	26.1503	9.9846	23.7187	-4.6448
30	11.1767	5.3408	9.3928	2.8581
35	14.0979	1.2673	13.0541	5.1706
40	17.0633	-1.5183	16.5720	3.7711
45	15.5742	-2.6177	15.3212	.9826
50	11.8903	-2.1769	11.5960	-1.4739
55	9.2327	-.6845	8.7265	-2.9364
60	9.1894	1.3658	8.3936	-3.4825
65	11.5358	3.6488	10.4180	-3.3504
70	15.0093	5.9049	13.5648	-2.5314
75	18.1173	7.6698	16.3957	-.7688
80	19.7231	8.1367	17.8520	2.0250
85	19.4541	6.4630	17.5889	5.2272
90	17.8394	2.5167	16.0401	7.3910
95	15.8687	-2.4887	14.0227	6.9990
100	14.0172	-6.2745	11.9481	3.7888
105	11.6906	-6.9134	9.4122	-.5346
110	8.1457	-4.4155	5.9915	-3.3101
115	4.1522	-.9028	2.5610	-3.1411
120	1.9229	1.1637	1.0596	-1.1049
125	2.5528	1.1444	2.2099	.5690
130	4.1854	.4416	4.0694	.8734
135	4.0847	.4987	4.0222	.5081
140	2.2526	1.0814	1.9717	.1317
145	1.3044	1.0415	.2238	-.7527
150	2.5793	.3184	.2291	-2.5493
155	4.2317	.1413	.9894	-4.1120
160	4.0060	1.0810	1.0856	-3.7015
165	2.3750	1.8607	.7497	-1.2714
170	1.6649	.7765	.9671	1.1106
175	2.9481	-2.1237	1.5315	1.3549
180	5.0554	-4.8831	1.1584	-.6088

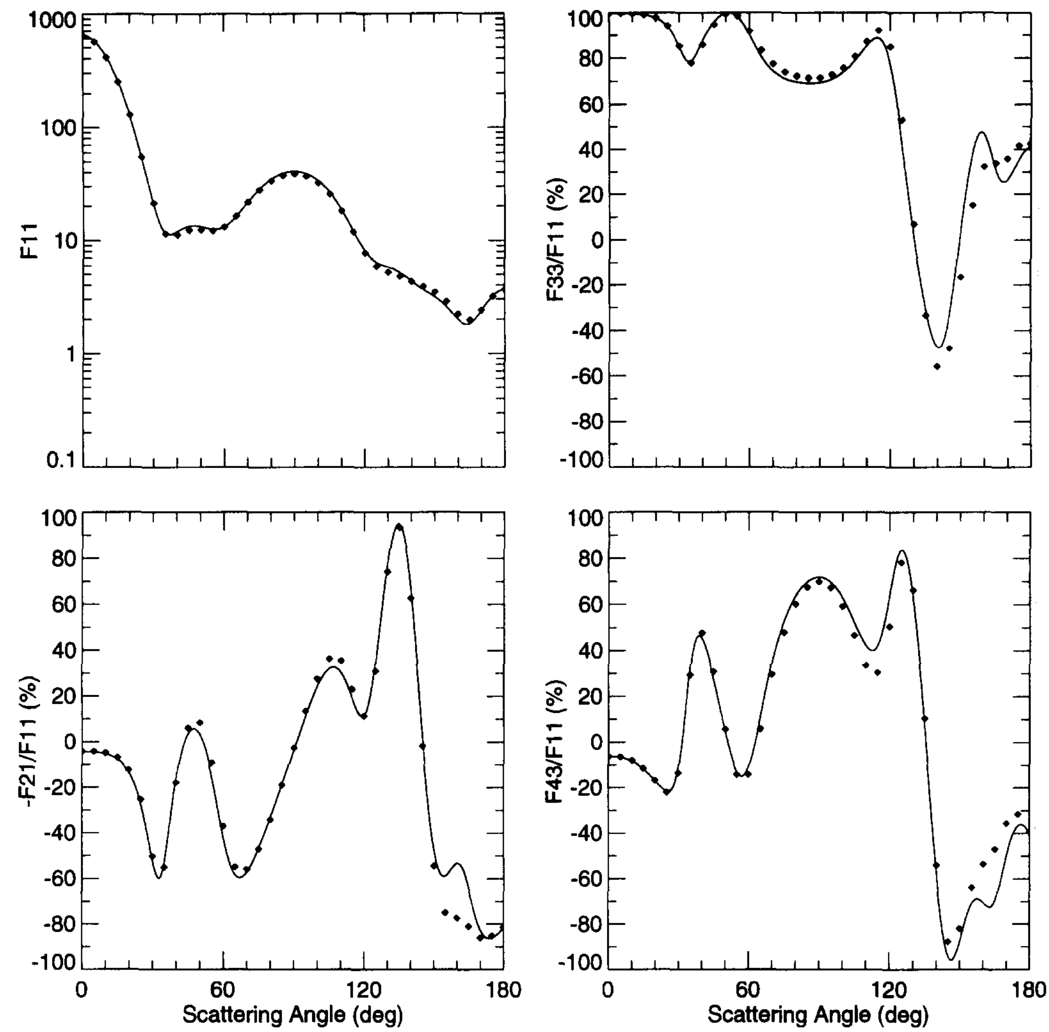

Fig. 2. The element F_{11} and the ratios of elements of the scattering matrix F_{33}/F_{11}, $-F_{21}/F_{11}$ and F_{43}/F_{11} as functions of the scattering angle in degrees for the prolate spheroid as described in the text. The dots represent DDA results. The curves are based on TMM and SVM results.

The intersections perpendicular to the rotation axis of our four particles can not be strictly circular in DDA computations. This requires a rotational averaging about that axis. The intersections have roughly an octagonal symmetry which limits the need for rotations about angles in the interval 0–45°. We found that stable results were obtained with four rotation averages, i.e., rotation angles of 0, 15, 30 and 45°.

The DDA is computationally much more laborious than the TMM and SVM, but, in principle, it can handle scatterers that are arbitrarily inhomogeneous and irregular.

4. COMPUTATIONAL RESULTS

The elements F_{11}, F_{21}, F_{33} and F_{43} of the scattering matrix are tabulated in Tables 1–4 for scattering angles, Θ, in the range 0–180° with a step size of 5°.

The results for the prolate (Table 1) and oblate (Table 2) spheroids were obtained with the T-matrix method and the separation of variables method. Both methods gave the same numbers within all six decimals given. This is a very gratifying result, since the methods are quite different and the scattering problems involved are not trivial.

The numbers in Tables 3 and 4 for the cylinder and bisphere, respectively, have been calculated with the (superposition) T-matrix method. Since the pertinent computer code has been thoroughly checked, the numbers in Tables 3 and 4 are expected to be correct within one unit of the last digit given.

The results of the computations with the discrete dipole approximation are shown as dots in Figs. 2–5. These figures show the element F_{11} and the ratios $-F_{21}/F_{11}$, F_{33}/F_{11}, and F_{43}/F_{11} as functions of scattering angle for the four scattering problems under consideration. The ratio $-F_{21}/F_{11}$ represents the degree of linear polarization of the scattered light for incident unpolarized light. The curves in Figs. 2–5 are based on the results of Tables 1–4. It is evident from Figs. 2–5 that the DDA produces the general features of the curves very well and that the deviations are generally small enough to be of little or no importance in many practical situations (see also Ref. 31).

5. DISCUSSION

It should be noted that the matrix elements F_{11}, F_{21}, F_{33} and F_{43} are not independent since in the cases considered in this paper

$$F_{11} = \{F_{21}^2 + F_{33}^2 + F_{43}^2\}^{1/2} \tag{3}$$

for all scattering angles. This is due to the fact that the amplitude matrix, which transforms the electric field components, is diagonal.[32]

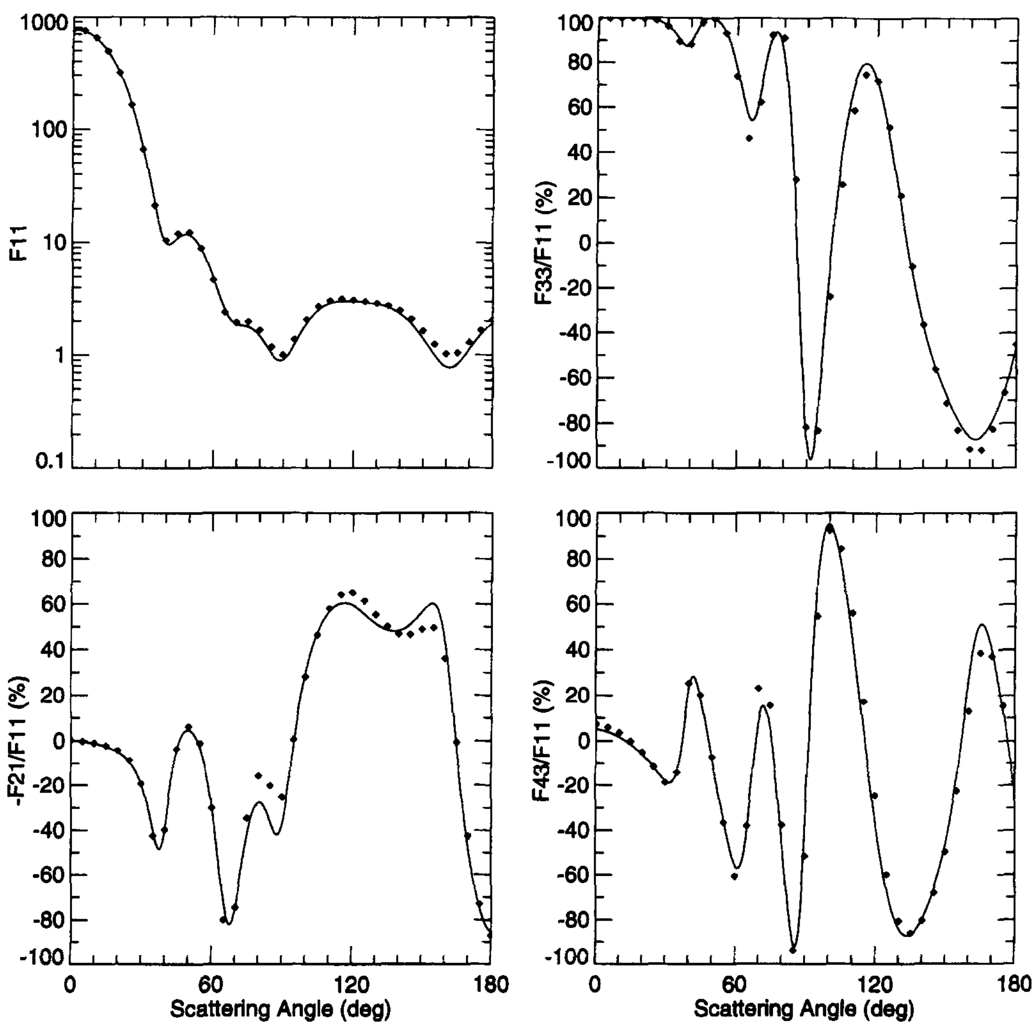

Fig. 3. As Fig. 2, but for an oblate spheroid.

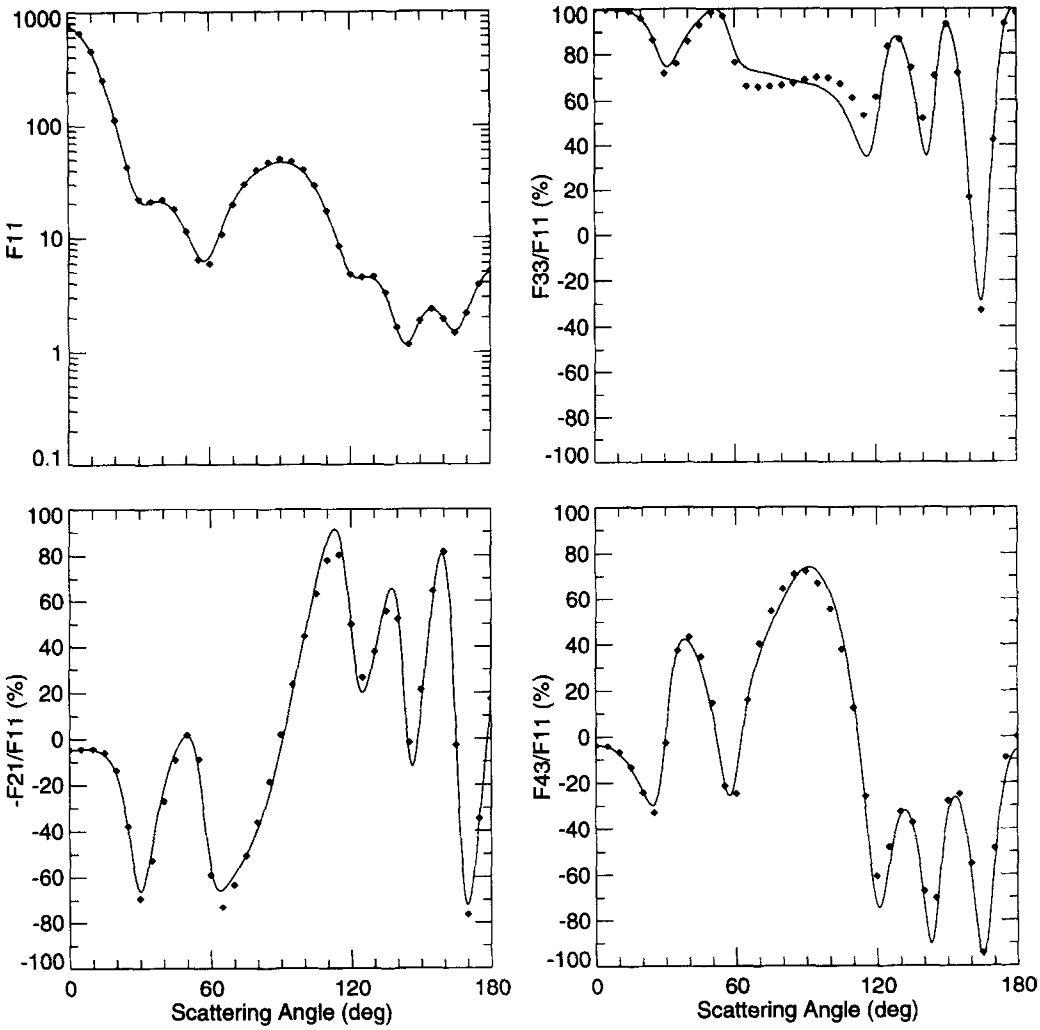

Fig. 4. As Fig. 2, but for a cylinder and curves based on TMM results.

Evidently, because of their high accuracy and reliability the numbers of Tables 1–4 may be viewed as benchmark results, which can be used to check an arbitrary computational method for light scattering by non-spherical particles. The high accuracy of the tables also reveals some subtle details. For instance, although for all four particle shapes F_{11} is very close to F_{33} for a scattering angle of 0°, these two matrix elements are never exactly equal for scattering in the strictly forward direction. This can not be seen in the top right panels of Figs. 2–5, but it is evident from the lower panels in conjunction with Eq. (3).

By comparing Figs. 2–5 with each other we can easily notice the shape effects of the scattering patterns. Apparently, there are a number of similarities. For example, each of the four curves representing $-F_{21}/F_{11}$ vs scattering angle has at least 5 minima. Also, going from one particle to another, the angular distributions of corresponding matrix elements are strikingly similar for scattering angles smaller than about 50°, which is the direction of the rotation axis of each particle (see Fig. 1). Furthermore, the cylinder and the bisphere show a large overall similarity in the angular distributions of their scattering matrix elements. On the other hand, the deep minimum in F_{33}/F_{11} near a scattering angle of 90° is only shown by the oblate spheroid.

6. CONCLUDING REMARKS

Initially we thought that it would cost little time to implement the project, described in the Introduction of this paper, since all three computational methods had been well developed and each

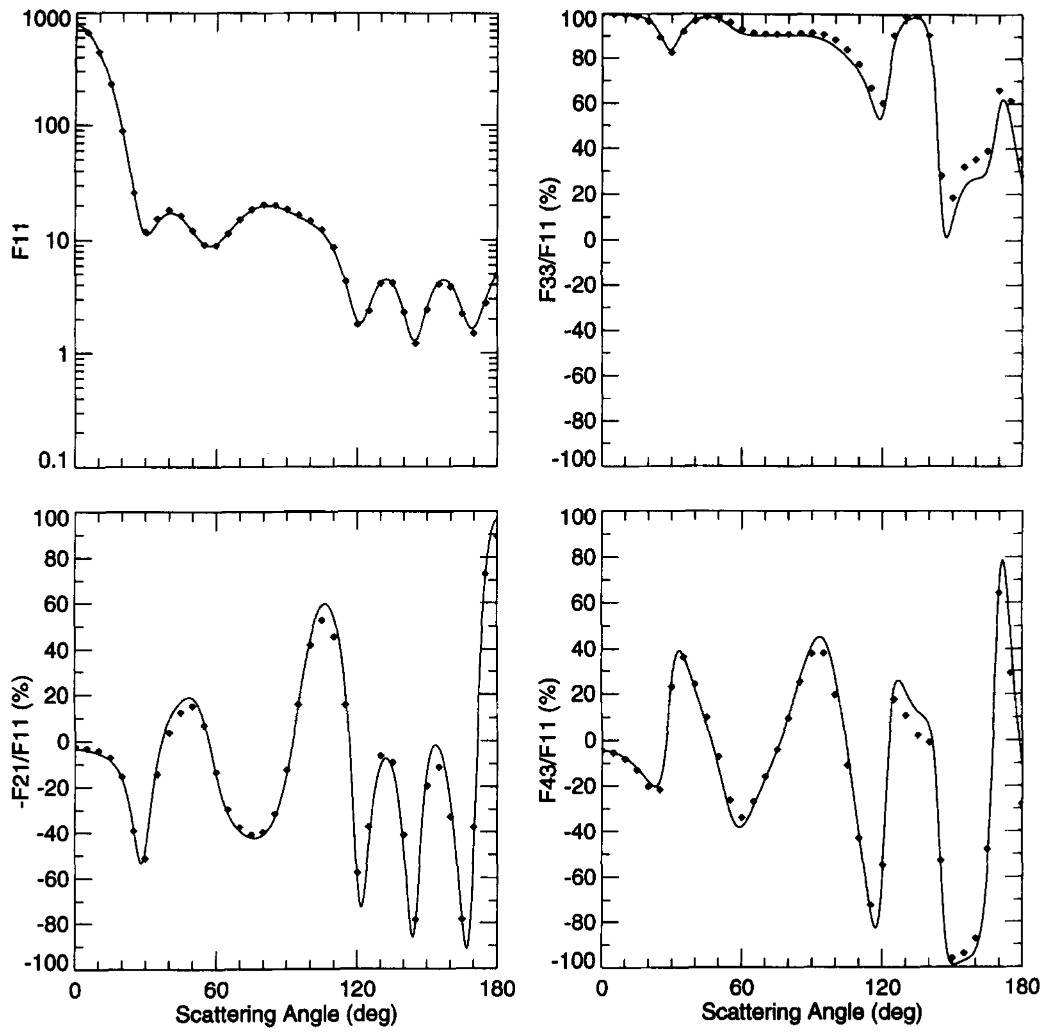

Fig. 5. As Fig. 2, but for a bisphere and curves based on TMM results.

advocate was very confident about his own method. Our first attempts, however, yielded widely different numbers. Continuing our efforts in a more modest mood, we checked the relevant methods and computer codes very carefully, until good agreement was obtained. In doing so, a lot was learned about the methods and several improvements in the implementation of the SVM and DDA were made. Apparently the original differences among the authors were due to misunderstandings about nomenclature and notation.

A question which is often posed in discussions on computations of light scattering by non-spherical particles is: "What is the best method?" Obviously, there is no unique answer to that question, since many factors are involved, such as the structure, size, shape and orientation of the particle, the scattering properties sought, the accuracy needed and the computer facilities which are (easily) available. The computations of this paper were limited to four scattering problems, which is not sufficient for a detailed comparison of the merits of the three methods used. We have, however, shown that all three of them are in good shape and deserve to be further developed.

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