Light scattering by randomly oriented bispheres

Michael I. Mishchenko

NASA Goddard Institute for Space Studies/Hughes STX Corporation, 2880 Broadway, New York, New York 10025

Daniel W. Mackowski

Department of Mechanical Engineering, Auburn University, Auburn, Alabama 36849-5341

Received June 6, 1994

We describe how the T-matrix approach can be used to compute analytically the Stokes scattering matrix for randomly oriented bispheres with touching or separated components. Computations for randomly oriented bispheres with touching components are compared with those for volume-equivalent randomly oriented prolate spheroids with an aspect ratio of 2 and for a single volume-equivalent sphere. We show that cooperative (multiple-scattering) effects can make bispheres more efficient depolarizers than spheroids in the backscattering direction.

Computations of light scattering by closely packed multiple-sphere configurations are important in a number of fields, e.g., atmospheric optics and the theory of weak localization of photons. In most cases of practical interest, scattering particles are distributed over orientations rather than perfectly aligned. However, since computations for randomly oriented clusters have been difficult and computationally intensive, practically all numerical data published so far pertain to clusters in a fixed orientation. Therefore it is the aim of this Letter to describe how the T-matrix approach can be efficiently used in computations of the Stokes scattering matrix for randomly oriented bispheres (two-sphere clusters) with touching or separated components.

In the framework of the T-matrix method[1] the electric fields incident on (subscript i) and scattered by (subscript s) a particle are expanded in a series of vector spherical harmonics as follows:

$$\mathbf{E}_i(\mathbf{r}) = \sum_{nm}\Big[a_{mn} Rg\mathbf{M}_{mn}(k\mathbf{r}) + b_{mn} Rg\mathbf{N}_{mn}(k\mathbf{r})\Big], \quad (1)$$

$$\mathbf{E}_s(\mathbf{r}) = \sum_{nm}\Big[p_{mn}\mathbf{M}_{mn}(k\mathbf{r}) + q_{mn}\mathbf{N}_{mn}(k\mathbf{r})\Big], \quad (2)$$

where $k = 2\pi/\lambda$ and λ is a free-space wavelength. The harmonics $Rg\mathbf{M}_{mn}$ and $Rg\mathbf{N}_{mn}$ have a Bessel-function radial dependence and are regular at the origin, whereas the functions $\mathbf{M}_{mn}$ and $\mathbf{N}_{mn}$ have a Hankel-function radial dependence and vanish at infinity. The expansion coefficients of the incident plane wave, a_{mn} and b_{mn}, are given by simple analytical expressions, whereas the expansion coefficients of the scattered field, p_{mn} and q_{mn}, are initially unknown. Because of the linearity of Maxwell's equations the relation between the expansion coefficients of the incident and scattered fields is linear and is given by a transition matrix $\mathbf{T}$:

$$\mathbf{p} = \mathbf{T}\mathbf{a}, \quad (3)$$

where we use compact matrix notation and denote the column of the expansion coefficients of the incident field by $\mathbf{a}$ and that of the scattered field by $\mathbf{p}$. If the T matrix for a given scatterer is known, Eqs. (2) and (3) can be used to compute the scattered field.

It was shown recently that the T-matrix approach can be used analytically to compute the Stokes scattering matrix for randomly oriented rotationally symmetric particles.[2] The major advantage of the analytical formulation is that first the elements of the scattering matrix are expanded in so-called generalized spherical functions,[3] and then the corresponding expansion coefficients are directly expressed in the elements of the T matrix computed in the natural reference frame with the z axis along the axis of particle symmetry. As a result, computation of the complicated angular structure of light scattered by a nonspherical particle in a fixed orientation with further numerical integration over particle orientations is avoided, thus making the analytical method accurate and computationally fast.

So far the T-matrix method has been used mainly in computations for simple isolated nonspherical scatterers. However, the T-matrix concept is quite general and can be applied to any composite scatterers, particularly bispheres. Since bispheres are bodies of revolution, the analytical method can be directly employed for efficient computations of the orientationally averaged Stokes scattering matrix, provided that the T matrix for a bisphere is computed in the natural reference frame with the z axis connecting the sphere centers.

To compute the T matrix, we use the method described in detail in Refs. 4 and 5 (see also Ref. 6). In brief, the scattered field from a two-sphere cluster can be given as the superposition of individual fields scattered from each sphere: $\mathbf{E}_s = \mathbf{E}_s{}^1 + \mathbf{E}_s{}^2$ (note, however, that these individual fields are interdependent because of electromagnetic interaction between the spheres). The external electric field illuminating the cluster and the individual fields scattered by the

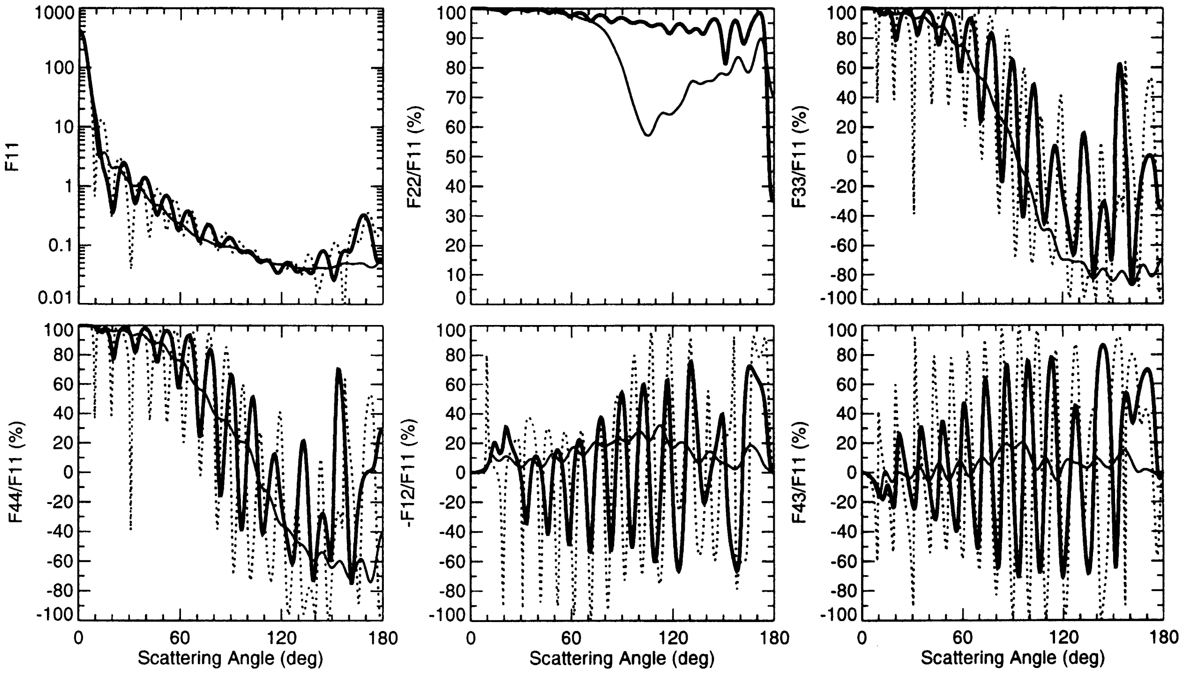

Fig. 1. Elements of the Stokes scattering matrix for randomly oriented bispheres with touching components (thick solid curves), randomly oriented prolate spheroids with aspect ratio 2 (thin solid curves), and a single sphere (dotted curves).

spheres can be expanded in vector spherical harmonics with origins at the sphere centers:

$$\mathbf{E}_i{}^j(\mathbf{r}^j) = \sum_{nm} \left[a_{mn}{}^j Rg\mathbf{M}_{mn}(k\mathbf{r}^j) + b_{mn}{}^j Rg\mathbf{N}_{mn}(k\mathbf{r}^j) \right], \quad j = 1, 2, \quad (4)$$

$$\mathbf{E}_s{}^j(\mathbf{r}^j) = \sum_{nm} \left[p^j \mathbf{M}_{mn}(k\mathbf{r}^j) + q_{mn}{}^j \mathbf{N}_{mn}(k\mathbf{r}^j) \right], \quad j = 1, 2, \quad (5)$$

where the index j numbers the spheres and vector $\mathbf{r}^j$ originates at the center of the jth sphere. To exploit the orthogonality of the harmonics in the sphere boundary conditions, we need to employ addition theorems in which a harmonic about one sphere origin can be reexpanded in harmonics about a second origin. This process ultimately leads to a system of linear equations for the scattered-field expansion coefficients $\mathbf{p}^j$ for each sphere j:

$$\mathbf{p}^j + \mathbf{A}^j \mathbf{H}^{jj'} \mathbf{p}^{j'} = \mathbf{A}^j \mathbf{a}^j, \qquad j, j' = 1, 2, \; j \neq j'. \quad (6)$$

Here diagonal matrices $\mathbf{A}^j$ represent the standard Lorenz–Mie scattering coefficients for each sphere and are functions solely of sphere size parameters and refractive indices. On the other hand, matrices $\mathbf{H}^{jj'}$ account for the electromagnetic coupling between the spheres and depend on only the distance and direction of travel between the spheres. We considerably reduce the complexity of the equations by exploiting the axial symmetry of the two-sphere cluster that results in a decoupling of different m modes. Inversion of Eq. (6) gives sphere-centered transition matrices $\mathbf{T}^{jj'}$ that transform the expansion coefficients of the incident field into the expansion coefficients of the individual scattered fields:

$$\mathbf{p}^j = \sum_{j'=1}^{2} \mathbf{T}^{jj'} \mathbf{a}^{j'}. \quad (7)$$

Realize that the matrices $\mathbf{T}^{jj'}$, as defined above, are based on the two origins of the spheres. In the far-field region the scattered-field expansions from the individual spheres can be transformed into a single expansion based on a single origin of the cluster. Employing again the addition theorem for vector harmonics, we find that the expansion coefficients based on the single cluster origin are

$$\mathbf{p} = \sum_{j=1}^{2} \mathbf{J}^j \mathbf{p}^j = \sum_{j=1}^{2} \sum_{j'=1}^{2} \mathbf{J}^j \mathbf{T}^{jj'} \mathbf{a}^{j'} = \sum_{j=1}^{2} \sum_{j'=1}^{2} \mathbf{J}^j \mathbf{T}^{jj'} \mathbf{J}^{j'} \mathbf{a} = \mathbf{T}\mathbf{a}, \quad (8)$$

in which the matrices $\mathbf{J}^j$ are similar in form to the $\mathbf{H}$ matrices in Eq. (6). The matrix $\mathbf{T}$ defined in the last term of Eq. (8) is the T matrix that we seek [see Eq. (3)] and can be used directly in computing the orientationally averaged scattering matrix for the cluster.

Calculation results presented here correspond to two identical touching spheres in random orientation. The refractive index and size parameter of each sphere are 1.5+0.02i and 15.874, respectively. The equal-volume-sphere size parameter for the two-sphere cluster is thus equal to 20. Computation of the T matrix for the two-sphere cluster required 11.6 s of CPU time on an IBM RISC Model 37T workstation, followed by 32.6 s to compute the expansion coefficients appearing in the generalized spherical function expansions of the scattering matrix elements and 0.3 s to compute the scattering matrix for 361 scattering angles. Thus the time for computing the orientationally averaged light scattering characteristics is comparable with that for computing the natural T matrix for the bisphere. This demonstrates once again the efficiency of the analytical averaging method. And, to the best of our knowledge, these are the first rigorous computations of the full scattering matrix for randomly oriented bispheres.

Results of the calculations appear in Fig. 1, in which the (normalized) elements of the Stokes scattering matrix, as defined by Eq. (2.9) of Ref. 7, are presented versus scattering angle. For comparison, Fig. 1 also presents results for randomly oriented volume-equivalent prolate spheroids with an aspect ratio of 2 and for a single volume-equivalent sphere. The bisphere–spheroid differences seen in Fig. 1 result mainly from the fact that the elements of the scattering matrix for the bisphere are strongly oscillating functions of the scattering angle, whereas for the spheroid the curves are much smoother. Thus, even when randomly oriented, bispheres preserve most of the interference structure characteristic of single monodisperse spherical particles,[7] whereas for spheroids this interference structure is more noticeably suppressed by particle nonsphericity and orientational averaging. Therefore averaging over sizes for bispheres is even more necessary than for spheroids to make the elements of the scattering matrix smooth enough that conclusions about the effects of particle shape on light scattering become meaningful (see Ref. 8).

The most interesting behavior is exhibited by the (2,2) and (4,4) elements of the scattering matrix. For single spheres, $F_{22}/F_{11} \equiv 1$, whereas for randomly oriented nonspherical particles this ratio may, in general, deviate from unity. Figure 1 shows that, for bispheres, the ratio F_{22}/F_{11} is in most cases closer to unity than for spheroids. However, at exactly the backscattering direction the deviation from unity is much bigger for bispheres, making the backscattering linear depolarization ratio $(F_{11} - F_{22})/(F_{11} + F_{22})$ calculated at the 180° scattering angle equal to 0.481, as opposed to 0.175 for spheroids. Similarly, the backscattering circular depolarization ratio $(F_{11} + F_{44})/(F_{11} - F_{44})$ for bispheres (1.851) is much larger than that for spheroids (0.425). For single spheres, both ratios are equal to zero (no depolarization). It is well known that two factors, namely, particle nonsphericity and multiple scattering, can cause significant depolarization. Thus apparently it is multiple (cooperative) scattering of light between two spheres that makes bispheres more efficient depolarizers than volume-equivalent spheroids.

Finally, we note that different parts of our code and the whole code have been extensively tested against numerical data,[9,10] and in all cases the quantitative agreement was excellent. Also, we have found that our computations for randomly oriented bispheres are in full agreement with the general equalities[11–13] $F_{12}(0) = F_{12}(\pi) = F_{34}(0) = F_{34}(\pi) = 0$, $F_{22}(0) = F_{33}(0)$, $F_{22}(\pi) = -F_{33}(\pi)$, $F_{11}(\pi) - F_{22}(\pi) = F_{44}(\pi) - F_{33}(\pi)$, and $F_{11}(0) - F_{22}(0) - F_{33}(0) + F_{44}(0) = 0$ as well as with general inequalities derived by van der Mee and Hovenier.[14]

In summary, we have demonstrated that the T-matrix approach in combination with the analytical averaging procedure can be efficiently used to compute light scattering by randomly oriented bispheres. The generalization of this method to linear chains consisting of an arbitrary number of spheres is rather straightforward and is the subject of our current research.

We are grateful to P. J. Flatau and V. P. Tishkovets for communicating the data that were used in testing our computer code. M. I. Mishchenko thanks B. E. Carlson, A. A. Lacis, and L. D. Travis for many useful discussions and acknowledges partial support from the NASA Office of Mission to Planet Earth, the NASA EOS Project, and the Department of Energy Interagency Agreement under the Atmospheric Radiation Measurement Program.

References

1. P. C. Waterman, Phys. Rev. D **3,** 825 (1971).
2. M. I. Mishchenko, J. Opt. Soc. Am. A **8,** 871 (1991); Appl. Opt. **32,** 4652 (1993).
3. J. W. Hovenier and C. V. M. van der Mee, Astron. Astrophys. **128,** 1 (1983).
4. D. W. Mackowski, Proc. R. Soc. London Ser. A **433,** 599 (1991).
5. D. W. Mackowski, J. Opt. Soc. Am. A **11,** 2851 (1994).
6. K. A. Fuller, Appl. Opt. **30,** 4716 (1991).
7. J. E. Hansen and L. D. Travis, Space Sci. Rev. **16,** 527 (1974).
8. M. I. Mishchenko and L. D. Travis, J. Quant. Spectrosc. Radiat. Transfer **51,** 759 (1994).
9. K. A. Fuller, G. W. Kattawar, and R. T. Wang, Appl. Opt. **25,** 2521 (1986).
10. V. P. Tishkovets, Kinem. Fiz. Nebes. Tel. **10**(2), 58 (1994).
11. H. C. van de Hulst, *Light Scattering by Small Particles* (Wiley, New York, 1957).
12. C.-R. Hu, G. W. Kattawar, M. E. Parkin, and P. Herb, Appl. Opt. **26,** 4159 (1987).
13. M. I. Mishchenko and L. D. Travis, Appl Opt. **33,** 7206 (1994).
14. C. V. M. van der Mee and J. W. Hovenier, Astron. Astrophys. **228,** 559 (1990).