Calculation of the T matrix and the scattering matrix for ensembles of spheres

Daniel W. Mackowski

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

Michael I. Mishchenko

State University of New York/Stony Brook, Stony Brook, New York, and NASA Goddard Institute for Space Studies, 2880 Broadway, New York, New York 10025

Received February 26, 1996; accepted April 18, 1996; revised manuscript received May 21, 1996

We present a method for determination of the random-orientation polarimetric scattering properties of an arbitrary, nonsymmetric cluster of spheres. The method is based on calculation of the cluster T matrix, from which the orientation-averaged scattering matrix and total cross sections can be analytically obtained. An efficient numerical method is developed for the T-matrix calculation, which is faster and requires less computer memory than the alternative approach based on matrix inversion. The method also allows calculation of the random-orientation scattering properties of a cluster in a fraction of the time required for numerical quadrature. Numerical results for the random-orientation scattering matrix are presented for sphere ensembles in the form of densely packed clusters and linear chains. © 1996 Optical Society of America.

1. INTRODUCTION

An understanding of the radiative properties of small particles is relevant to a wide variety of applications, which include optical diagnostics for industrial aerosol processes and combustion, environmental issues (e.g., visibility and haze problems), remote atmospheric sensing (lidar), and astronomical issues such as the effect of interstellar dust on propagation of stellar radiation. More often than not, particles that are formed in natural or technological processes will possess complicated morphologies. Frequently, however, the morphological complexity of small particles arises from aggregation of individual particles that, by themselves, possess a simple shape. Soot particles, for example, are commonly in the form of aggregates of small spherical monomers. Scavenging of soot by water or sulfate droplets results in a compound particle that, again, consists of several spherical subunits. Pigment particles that are produced in aerosol reactors (such as TiO_2), are, again, commonly in the form of clusters of nearly spherical individual particles.

As a whole, a cluster of small particles is profoundly nonspherical in shape, and because of this the usual light-scattering formulas based on regular particles (such as spheres) cannot, with reasonable accuracy, be directly applied to aggregated particles. However, if the individual particles making up the aggregate possess shapes that admit analytical solutions to the wave equations, it is possible to calculate exactly, by appropriate superposition techniques, the radiative properties of the aggregate.

This approach has been well established for clusters of spheres—for both cases of internal aggregation (spheres within spheres) and external spheres.[1–6] Even though the solution is analytical, application of the solution to large-scale clusters (in both the sizes and the numbers of spheres) is ultimately limited by numerical issues. The exact formulation leads to a system of linear equations for the multipole expansion coefficient that describe the scattered field from each sphere in the cluster. Large-scale clusters can easily result in systems containing of the order of 10^3 unknowns—for which direct solution techniques (i.e., matrix inversion) become intractable. Because of this, solution of the equations for the multipole coefficients is almost always accomplished with iterative methods.[6–8] A drawback of this approach is that it yields only the coefficients—and the resulting scattering and absorption properties of the cluster—for a particular fixed orientation and polarization of the incident field. Frequently, the random-orientation properties of a cluster are of primary interest, and to obtain these from an iterative solution scheme obviously requires numerical quadrature of the fixed-orientation results over a range of incident directions. Recent investigations of the polarimetric scattering properties of sphere clusters have shown that the depolarized scattering components can be highly sensitive to the orientation of the cluster with respect to the incident field.[9,10] Because of this, the accurate numerical calculation of polarimetric scattering properties of a cluster can require an extremely fine quadrature scheme.

The purpose of this paper is to describe an efficient computational scheme to calculate analytically the fixed and averaged scattering properties of a cluster of spheres. Our approach is to exploit the properties of a T-matrix description of scattering from the cluster—from which the random-orientation cross sections and scattering matrix can be obtained analytically. We have recently used this approach to examine the orientation-averaged scattering matrix of the axisymmetric bisphere cluster;[10] here we intend to generalize the formulation to arbitrary clusters. We begin by reviewing the transformation of the

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superimposed-field (or sphere-centered) representation of scattering from the cluster into a single T-matrix description and present the mathematical procedure for extracting the random-orientation scattering matrix elements. Next, we address the numerical issues regarding the calculation of the T matrix for an arbitrary cluster and offer a computational strategy that minimizes both memory and time requirements. We end by presenting calculation results for chains of spheres and packed sphere clusters and discuss some of the salient aspects of the effects of multiple scattering and cluster symmetry on the scattering matrix elements.

2. FORMULATION

A. Cluster T Matrix

The procedure for analytically calculating the T matrix for a cluster of spheres has been described in detail in Mackowski;[11] only an outline of the formulation will be presented here. As mentioned in Section 1, the approach involves a superposition solution to Maxwell's equations for the multiple spherical boundary domain. The scattered field from the cluster as a whole is resolved into partial fields scattered from each of the N_S spheres in the cluster, i.e.,

$$\mathbf{E}_s = \sum_{i=1}^{N_S} \mathbf{E}_{i,s}, \tag{1}$$

where each partial field $\mathbf{E}_{i,s}$ is represented by an expansion of vector spherical harmonics that are written about the origin of the ith sphere:

$$\mathbf{E}_{s,i} = \sum_{n=1}^{\infty} \sum_{m=-n}^{n} \sum_{p=1}^{2} a^i_{mnp} \mathbf{h}_{mnp}(\mathbf{r}_i). \tag{2}$$

In the above equation $\mathbf{h}$ denotes the outgoing-wave vector spherical harmonic of order n and degree m, and a^i are the corresponding scattered-field expansion coefficients for sphere i. The extra index p denotes mode, in which $p = 1$ and 2 refer to the TM and TE modes of the scattered field, respectively.

The field arriving at the surface of the ith sphere will consist of the incident field plus scattered fields that originate from all other spheres in the cluster. By use of the addition theorem for vector harmonics these interacting scattered fields can be transformed into expansions about the origin of sphere i, which makes possible an analytic formulation of the boundary conditions at the surface. After truncation of the expansions to $n = N_{O,i}$ orders one eventually arrives at a system of equations for the scattering coefficients, which take the form

$$a^i_{mnp} + \bar{a}^i_{np} \sum_{\substack{j=1 \\ j\neq i}}^{N_S} \sum_{l=1}^{N_{O,i}} \sum_{k=-l}^{l} \sum_{q=1}^{2} H^{ij}_{mnp\,klq} a^j_{klq} = \bar{a}^i_{np} p^i_{mnp}. \tag{3}$$

In the above equation H^{ij} denotes a matrix formed from the vector harmonic addition coefficients (based on the spherical Hankel functions) and depends solely on the distance and the direction of translation from origins j to i, $\bar{a}^i$ are the Lorenz/Mie multipole scattering coefficients for sphere i, which depend on the size parameter x_i ($= ka_i = 2\pi a_i/\lambda$) and the refractive index $m_i = n + ik$ of sphere i, and p^i denote the expansion coefficients for the plane, linearly polarized incident wave at the origin of i. Calculation of the addition coefficients is discussed in Appendix A.

Formal inversion of the system of equations in Eq. (3) identifies the sphere-centered T matrix of the cluster:

$$a^i_{mnp} = \sum_{j=1}^{N_S} \sum_{l=1}^{N_{O,i}} \sum_{k=-l}^{l} \sum_{q=1}^{2} T^{ij}_{mnp\,klq} p^j_{klq}. \tag{4}$$

The T^{ij} matrix is of limited use in describing the scattered field from the cluster [through Eqs. (1) and (2)] because it is based upon the multiple sphere origins of the cluster. Total cross sections in both fixed and random orientation can be obtained from relatively simple operations directly on T^{ij},[11] yet to describe the differential scattering cross sections (i.e., the scattering matrix), it is advantageous to transform the sphere-centered T^{ij} matrix into an equivalent cluster-centered T matrix that is based upon a single origin of the cluster. This transformation is of the form

$$T_{nl} = \sum_{i=1}^{N_S} \sum_{j=1}^{N_S} \sum_{n'=1}^{N_{O,i}} \sum_{l'=1}^{N_{O,i}} J^{0i}_{nn'} T^{ij}_{n'l'} J^{j0}_{l'l}. \tag{5}$$

The J^{0i} and J^{j0} matrices are formed from the addition coefficients based on the spherical Bessel function, and the subscripts n and l (and their primes) are shorthand for order, degree, and mode.

The T matrix given in the above equation is completely equivalent to those calculated with extended boundary-condition methods.[12,13] In particular, the orientation-averaged scattering matrix elements can be analytically obtained from the T matrix, following the procedures developed by Mishchenko[14] for axisymmetric scatterers and by Khlebtsov[15] for general scatterers. Because the nomenclature and the normalization are somewhat different here from those appearing in these previous works, we will describe in more detail the formulation of the fixed- and random-orientation scattering matrix elements.

B. Amplitude Matrix Elements

At large distances from the cluster the scattered field becomes a spherical transverse wave. Using the asymptotic limit of the Hankel function, we can express the components of the wave by[16]

$$E_{\theta s} = \frac{i}{kr} \exp(ikr) \sum_{n=1}^{N_O} \sum_{m=-n}^{n} \sum_{p=1}^{2} \times (-i)^{n+1} a_{mnp} \tau_{mnp}(\theta) \exp(im\phi), \tag{6}$$

$$E_{\phi s} = -\frac{1}{kr} \exp(ikr) \sum_{n=1}^{N_O} \sum_{m=-n}^{n} \sum_{p=1}^{2} \times (-i)^{n+1} a_{mnp} \tau_{mn3-p}(\theta) \exp(im\phi). \tag{7}$$

In the above equations a_{mnp} are the scattering coefficients that define the entire scattered field from the cluster, and N_O is the maximum retained order in the cluster T matrix (which will be discussed below). The scattering coefficients are obtained from the cluster T matrix through

$$a_{mnp} = \sum_{l=1}^{N_O} \sum_{k=-l}^{l} \sum_{q=1}^{2} T_{mnpklq} p_{klq}, \tag{8}$$

where p_{klq} represent the expansion coefficients for the incident wave defined at the cluster origin. The scattering functions τ_{mnp} are defined by

$$\tau_{mn1}(\theta) = \frac{\mathrm{d}}{\mathrm{d}\theta} P_n^m(\cos\theta), \tag{9}$$

$$\tau_{mn2}(\theta) = \frac{m}{\sin\theta} P_n^m(\cos\theta). \tag{10}$$

Equations (6) and (7) are not especially useful in describing the scattered field because the polar coordinates θ and ϕ are defined with respect to the cluster axes and not with respect to the direction and the polarization of the incident wave. This problem is rectified by rotating the scattered field to coincide with the incident field.[14,17] The incident field can be defined according to a Cartesian frame x', y', z' so that z' is the direction of propagation and Φ is the angle between the electric-field vector and the x' axis. Since this incident frame can be specified by rotating the cluster frame through appropriate Euler angles α, β, and γ, an identical rotation on the scattering functions will redefine the scattered field with respect to the incident field. The rotation transformation can be expressed[18] as

$$\tau_{mnp}(\theta)\exp(im\phi) = \exp(im\alpha)\sum_{k=-n}^{n} \mathscr{D}_{kn}^{m}(\beta) \times \exp(ik\gamma)\tau_{knp}(\theta')\exp(ik\phi'), \tag{11}$$

where the rotation functions $\mathscr{D}$ are equivalent (to a normalizing factor) to the generalized spherical functions.[14] Formulas for these functions appear in Appendix A. The polar angles θ' and ϕ' are now fixed with respect to the incident field; $\theta' = 0$ is the forward-scattering direction, and $\phi' = \Phi$ is the direction of the incident electric-field vector. By application of Eq. (11) to Eqs. (6) and (7), the scattering coefficients can be transformed so that θ' and ϕ' take the place of θ and ϕ. This transformation is

$$a'_{mnp} = \exp(im\gamma)\sum_{k=-n}^{n} \mathscr{D}_{mn}^{k}(\beta)\exp(ik\alpha)a_{knp}. \tag{12}$$

Since α, β, and γ represent the rotation of the cluster coordinate system to the incident-field system, then $-\gamma$, $-\beta$, and $-\alpha$ will rotate the incident field to the cluster coordinates. Consequently, the incident-field expansion coefficients can be described by

$$p_{mnp}(\alpha, \beta, \gamma) = \exp(-im\alpha)\sum_{k=-n}^{n} \mathscr{D}_{mn}^{k}(-\beta) \times \exp[-ik(\gamma + \Phi)]\, p'_{knp}(0, 0, 0), \tag{13}$$

where p' refers to the expansion coefficients for the plane, linearly polarized wave in the incident-field frame. Specifically,

$$p'_{1n1} = \frac{i^{n+1}}{2}\frac{2n+1}{n(n+1)}, \qquad p'_{-1n1} = -\frac{i^{n+1}}{2}(2n+1),$$
$$p'_{1n2} = p'_{1n1}, \qquad p'_{-1n2} = -p'_{-1n1},$$
$$p'_{mnp} = q'_{mnp} = 0, \qquad |m| \neq 1. \tag{14}$$

With the use of

$$\mathscr{D}_{mn}^{k}(-\beta) = (-1)^{k+m}\mathscr{D}_{-kn}^{-m}(\beta) \tag{15}$$

the T-matrix equation for the scattering coefficients becomes

$$a'_{mnp} = (-1)^{k+s}\exp[i(m-k)\gamma]\exp[i(t-s)\alpha] \times \mathscr{D}_{mn}^{t}(\beta)T_{tnpslq}\mathscr{D}_{-kl}^{-s}(\beta)\exp(-ik\Phi)p'_{klq}. \tag{16}$$

Above and in what follows we adopt a tensorial convention; summation over all indices not appearing on the left-hand side of the equation is implied.

To identify the elements of the amplitude scattering matrix, first we arbitrarily define the scattering plane by $\phi' = 0$ (i.e., the z'–x' plane). Components of the scattered electric field that are polarized parallel and perpendicular to this plane are given simply by $E_{\parallel s} = E_{\theta' s}$ and $E_{\perp s} = E_{\phi' s}$. Likewise, incident radiation polarized either parallel or perpendicular to the plane is defined by setting Φ in Eq. (16) to 0 or $\pi/2$, respectively. The four elements of the amplitude scattering matrix can then be expressed as

$$S_1 = \tau_{mn3-p}(\theta')(-i)^n a'^{2}_{mnp}, \tag{17}$$

$$S_2 = \tau_{mnp}(\theta')(-i)^{n+1} a'^{1}_{mnp}, \tag{18}$$

$$S_3 = \tau_{mnp}(\theta')(-i)^{n+1} a'^{2}_{mnp}, \tag{19}$$

$$S_4 = \tau_{mn3-p}(\theta')(-i)^n a'^{1}_{mnp}, \tag{20}$$

where the superscript 1 or 2 on a'_{mnp} denotes the scattering coefficients calculated for parallel or perpendicular incident polarization, respectively.

C. Random-Orientation Scattering Matrix

For a fixed orientation of the cluster with respect to the incident beam the most direct method for calculating the scattering matrix is first to calculate the scattering amplitude elements in Eqs. (17)–(20) and then apply the formulas for the scattering matrix elements in terms of products of the amplitude elements.[16] To calculate analytically the orientation-averaged scattering matrix, on the other hand, one must first form the products of the scattering amplitude elements and then integrate the products over all directions and polarizations of the incident field. Referring to Eq. (16), the T-matrix representation of a'_{mnp} contains the product of two rotation functions—and the products of the amplitude elements will correspondingly contain products of four rotation functions. Integration over all incident directions thus requires a linearization of the $\mathscr{D}$ product in Eq. (16). Borrowing from relationships developed for the coupling of angular momentum in quantum mechanics,[18] we can expand the product into

$$\mathscr{D}^{t}_{mn}(\beta)\mathscr{D}^{s}_{-kl}(\beta) = \sum_{w=|n-l|}^{n+l} (-1)^{n+l+w} \times \hat{C}^{w}_{tn,-sl}\hat{C}^{w}_{-mn,kl}\mathscr{D}^{t-s}_{m-kw}(\beta). \tag{21}$$

The $\hat{C}$ coefficients are equivalent (to a normalizing factor) to the vector-coupling coefficients; definitions and recurrence relations are given in Appendix A.

By use of the orthogonality properties of the rotation functions, i.e.,

$$\frac{1}{8\pi^2}\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi} \exp[i(u-u')\gamma] \times \exp[i(v-v')\alpha]\mathscr{D}^{v}_{uw}(\beta)\mathscr{D}^{v'}_{u'w'}(\beta)\sin\beta \mathrm{d}\beta\mathrm{d}\alpha\mathrm{d}\gamma = \frac{1}{2w+1}\frac{f_{vw}}{f_{uw}}\delta_{uu'}\delta_{vv'}\delta_{ww'}, \tag{22}$$

where

$$f_{vw} = \frac{(w+v)!}{(w-v)!}, \tag{23}$$

the orientation-averaged products of the scattering coefficients can be given as

$$i^{n'-n}(\langle a'^{1}_{u+knp}a'^{1*}_{u'+k'n'p'}\rangle + \langle a'^{2}_{u+knp}a'^{2*}_{u'+k'n'p'}\rangle) = 2D^{u}_{knpkn'p'}\delta_{uu'}\delta_{kk'}, \tag{24}$$

$$i^{n'-n}(\langle a'^{1}_{u+knp}a'^{1*}_{u'+k'n'p'}\rangle - \langle a'^{2}_{u+knp}a'^{2*}_{u'+k'n'p'}\rangle) = 2D^{u}_{knp-kn'p'}\delta_{uu'}\delta_{k-k'}, \tag{25}$$

$$i^{n'-n}\langle a'^{2}_{u+knp}a'^{1*}_{u'+k'n'p'}\rangle = i^{-k}D^{u}_{knpk'n'p'}\delta_{uu'}. \tag{26}$$

The matrix D is obtained from the following operations;

$$D^{u}_{knpk'n'p'} = \sum_{w=|n-n'|}^{n+n'} \frac{1}{2w+1} \times \sum_{v=-w}^{w} f_{vw}f_{-uw}B^{uvw}_{knp}B^{uvw*}_{k'n'p'}, \tag{27}$$

$$B^{uvw}_{knp} = \sum_{l=L_1}^{L_2} (-1)^{n+l}A^{vw}_{kl\,np}\hat{C}^{w}_{-u-kn,kl}, \tag{28}$$

$$A^{vw}_{kl\,np} = i^{-n}\sum_{t=T_1}^{T_2}\sum_{q=1}^{2} (-1)^{t}\hat{C}^{w}_{tn,v-tl}T_{tnpt-vlq}p'_{klq}. \tag{29}$$

The limits on l and t in the above equations are defined as

$$L_1 = \max(1, |w-n|), \qquad L_2 = \min(N_O, w+n),$$
$$T_1 = \max(-n, -l+v), \qquad T_2 = \min(n, l+v). \tag{30}$$

In addition, the indices k and k' in the above equations are limited to the values of -1 and 1.

Referring again to Eqs. (17)–(20), the angular dependence of the scattering matrix elements would be contained in products of the $\tau_{mnp}(\theta')$ scattering functions. When we use the same techniques discussed above, these products can be linearized into a single expansion of $\mathscr{D}^{m'}_{mn}(\theta')$ rotation functions, which provides a compact and computationally efficient representation of the scattering matrix.[14,17] To develop this expansion, we first note that the τ and $\mathscr{D}$ functions are related by

$$\tau_{mnp} = -\frac{1}{2}[n(n+1)\mathscr{D}^{m}_{1n} + (-1)^{p}\mathscr{D}^{m}_{-1n}]. \tag{31}$$

With the use of Eq. (21), along with the definitions of the scattering matrix[16] and a considerable amount of algebra, the scattering matrix elements can be given by

$$S_{11} = \mathrm{Re}\left[\sum_{w=0}^{2N_O}(a_{0,1,w} + a_{0,-1,w})\mathscr{D}^{0}_{0w}(\theta')\right], \tag{32}$$

$$S_{14} = \mathrm{Re}\left[\sum_{w=0}^{2N_O}(a_{0,1,w} - a_{0,-1,w})\mathscr{D}^{0}_{0w}(\theta')\right], \tag{33}$$

$$S_{44} = \mathrm{Re}\left[\sum_{w=0}^{2N_O}(b_{0,1,w} - b_{0,-1,w})\mathscr{D}^{0}_{0w}(\theta')\right], \tag{34}$$

$$S_{42} = \mathrm{Re}\left[\sum_{w=0}^{2N_O}(b_{0,1,w} + b_{0,-1,w})\mathscr{D}^{0}_{0w}(\theta')\right], \tag{35}$$

$$S_{12} = \mathrm{Re}\left[\sum_{w=2}^{2N_O}(a_{2,1,w} + a_{2,-1,w})\mathscr{D}^{2}_{0w}(\theta')\right], \tag{36}$$

$$S_{24} = \mathrm{Re}\left[\sum_{w=2}^{2N_O}(a_{2,1,w} - a_{2,-1,w})\mathscr{D}^{2}_{0w}(\theta')\right], \tag{37}$$

$$S_{34} = -\mathrm{Im}\left[\sum_{w=2}^{2N_O}(b_{2,1,w} - b_{2,-1,w})\mathscr{D}^{2}_{0w}(\theta')\right], \tag{38}$$

$$S_{31} = -\mathrm{Im}\left[\sum_{w=2}^{2N_O}(b_{2,1,w} + b_{2,-1,w})\mathscr{D}^{2}_{0w}(\theta')\right], \tag{39}$$

$$S_{13} = 2\,\mathrm{Im}\left[\sum_{w=2}^{2N_O}a_{2,0,w}\mathscr{D}^{2}_{0w}(\theta')\right], \tag{40}$$

$$S_{41} = 2\,\mathrm{Re}\left[\sum_{w=2}^{2N_O}b_{2,0,w}\mathscr{D}^{2}_{0w}(\theta')\right], \tag{41}$$

$$S_{22} = \mathrm{Re}\left\{\sum_{w=2}^{2N_O}[c_{20w}\mathscr{D}^{2}_{2w}(\theta') + c_{-20w}\mathscr{D}^{2}_{-2w}(\theta')]\right\}, \tag{42}$$

$$S_{23} = \mathrm{Im}\left\{\sum_{w=2}^{2N_O}[c_{20w}\mathscr{D}^{2}_{2w}(\theta') + c_{-20w}\mathscr{D}^{2}_{-2w}(\theta')]\right\}, \tag{43}$$

$$S_{33} = \mathrm{Re}\left\{\sum_{w=2}^{2N_O}[c_{20w}\mathscr{D}^{2}_{2w}(\theta') - c_{-20w}\mathscr{D}^{2}_{-2w}(\theta')]\right\}, \tag{44}$$

$$S_{32} = -\mathrm{Im}\left\{\sum_{w=2}^{2N_O}[c_{20w}\mathscr{D}^{2}_{2w}(\theta') - c_{-20w}\mathscr{D}^{2}_{-2w}(\theta')]\right\}. \tag{45}$$

The a, b, and c expansion coefficients appearing in the above equations are obtained from

$$a_{0,k,w} = \sum_{n,p,n'} f_{1n'} \hat{C}^{w}_{1n,-1n'} F^{w}_{knp\,kn'p'}, \tag{46}$$

$$b_{0,k,w} = \sum_{n,p,n'} f_{1n'} \hat{C}^{w}_{1n,-1n'} F^{w}_{knp\,kn'3-p'}, \tag{47}$$

$$a_{2,k,w} = \sum_{n,p,n'} (-1)^{p'} \hat{C}^{w}_{1n,1n'} F^{w}_{knp\,kn'p'}, \tag{48}$$

$$b_{2,k,w} = \sum_{n,p,n'} (-1)^{3-p'} \hat{C}^{w}_{1n,1n'} F^{w}_{knp\,kn'3-p'}, \tag{49}$$

$$a_{2,0,w} = \sum_{n,p,n'} f_{1n'} \hat{C}^{w}_{1n,-1n'} F^{w}_{1np-1n'p'}, \tag{50}$$

$$b_{2,0,w} = \sum_{n,p,n'} f_{1n'} \hat{C}^{w}_{1n,-1n'} F^{w}_{1np-1n'3-p'}, \tag{51}$$

$$c_{2,0,w} = f_{2w} \sum_{n,p,n',p'} (-1)^{p'} \hat{C}^{w}_{1n,1n'} F^{w}_{1np-1n'p'}, \tag{52}$$

$$c_{-2,0,w} = \sum_{n,p,n',p'} (-1)^{p+n+n'+w} \hat{C}^{w}_{1n,1n'} F^{w}_{1np-1n'p'}, \tag{53}$$

where the F matrix is given by

$$F^{w}_{knp\,k'n'p'} = 2\sum_{u} (-1)^{u} f_{u+k'n'} \times \hat{C}^{w}_{u+kn,-u-k'n'} D^{u}_{knp\,k'n'p'}. \tag{54}$$

The mode index p' in Eqs. (46–51) takes on values that depend on n, n', and w. Specifically, if $n + n' + w$ is even, then $p' = p$; otherwise $p' = 3 - p$. The range on w in the above equations is also dependent on n and n', with $|n - n'| \leq w \leq n + n'$. As above, the indices k and k' in the above equations take on only the values of -1 and 1.

The first two terms in Eq. (32) have a well-recognized physical significance. Specifically, the random-orientation total cross section of the cluster will be given by

$$\langle C_{\text{sca}} \rangle = \frac{2\pi}{k^2} \operatorname{Re}(a_{0,1,0} + a_{0,-1,0}). \tag{55}$$

Likewise, the second term in Eq. (32) corresponds to the asymmetry factor

$$\langle C_{\text{sca}} \cos \theta' \rangle = \frac{2\pi}{3k^2} \operatorname{Re}(a_{0,1,1} + a_{0,-1,1}). \tag{56}$$

Finally, the orientation-averaged extinction cross section can be obtained from

$$\langle C_{\text{ext}} \rangle = \frac{2\pi}{k^2} \operatorname{Re}\left(\sum_{n,m,p} T_{mnp\,mnp} \right). \tag{57}$$

Frequently, these three properties are of sole importance in radiative transfer applications. In such cases the given formulation for the orientation-averaged scattering properties can be optimized. First, the orientation-averaged scattering cross section can be obtained directly from the T matrix through

$$\langle C_{\text{sca}} \rangle = \frac{2\pi}{k^2} \sum_{n,m,p} \sum_{k,l,q} \frac{n(n+1)(2l+1) f_{mn}}{l(l+1)(2n+1) f_{kl}} |T_{mnp\,klq}|^2. \tag{58}$$

Second, note from Eq. (54) that $F^{1}_{knp\,kn'p'}$ would have $|n - n'| = 1$; i.e., the C's for $w = 1$ would be zero for other combinations of n and n'. Taking this over to Eq. (27), it is seen that the index n' can be constrained to limits of $n' = n \pm 1$ if only $a_{0,k,1}$ is required.

D. Numerical Aspects

The formal procedure for calculating the cluster T matrix would first require the inversion of the system of equations in Eq. (3) to produce the sphere-centered T^{ij} matrix, followed by the contraction of the T^{ij} into the cluster T matrix through Eq. (5). The random-orientation scattering matrix can then be obtained from calculation of the A, B, and D matrices in Eqs. (27)–(29) and the scattering matrix expansion coefficients in Eqs. (46)–(54).

The first two steps can easily become the most challenging numerical task of the calculations. Considering order, degree, and mode, the number of unknown scattering coefficients for each individual sphere, denoted M_i, will be $M_i = 2N_{O,i}(N_{O,i} + 2)$, where $N_{O,i}$ is the maximum order retained in the individual sphere scattered-field expansions. Typically, $N_{O,i}$ will be proportional to the size parameter x_i of the individual spheres. For example, a system of five spheres, each having a size parameter of 6 (and with the use of $N_{O,i} = 8$), will require (at most) inversion of an 800×800 complex-valued matrix. On the other hand, the maximum order of the cluster T matrix, denoted N_O, will be proportional to the size parameter of the smallest sphere that circumscribes the entire cluster. Referring to Eq. (5), this implies that the J^{0i} and J^{j0} matrices are not square—J^{0i} has M_i columns and $M = 2N_O(N_O + 2)$ rows, and vice-versa for J^{j0}. Putting it all together, calculation of the T^{ij} matrix will involve of the order of $(N_S M_i)^3$ operations, followed by $(N_S M_i)^2 M + N_S M_i M^2$ operations to contract the T^{ij} matrix into T.

This task can be considerably simplified if the cluster takes the configuration of a linear chain of spheres aligned along a common z axis, which results in a decoupling of the azimuthal degrees of the harmonics.[19] Borghese *et al.* have discussed the application of group theory techniques to reduce the size of Eq. (3) for clusters that possess more complicated symmetries.[20] Such methods, however, are not easily implemented, nor would they work for random, nonsymmetric clusters.

In the end, however, it is only the cluster-centered T matrix, and not the sphere-centered T^{ij} matrix, that is needed to calculate analytically the orientation-averaged scattering matrix. Because of this, a considerable amount of computer memory (and often time) can be saved by linking steps 1 and 2. To illustrate this process, we first note that the formal inversion of Eq. (3) can be described by the matrix equation

$$\begin{bmatrix} \mathbf{I} & \bar{\mathbf{a}}^1\mathbf{H}^{12} & \bar{\mathbf{a}}^1\mathbf{H}^{13} & \cdots \\ \bar{\mathbf{a}}^2\mathbf{H}^{21} & \mathbf{I} & \bar{\mathbf{a}}^2\mathbf{H}^{23} & \cdots \\ \bar{\mathbf{a}}^3\mathbf{H}^{31} & \bar{a}^3\mathbf{H}^{32} & \mathbf{I} & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{bmatrix} \begin{bmatrix} \mathbf{T}^{11} & \mathbf{T}^{12} & \mathbf{T}^{13} & \cdots \\ \mathbf{T}^{21} & \mathbf{T}^{22} & \mathbf{T}^{23} & \cdots \\ \mathbf{T}^{31} & \mathbf{T}^{32} & \mathbf{T}^{33} & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{bmatrix} = \begin{bmatrix} \mathbf{a}^1\mathbf{I} & 0 & 0 & \cdots \\ 0 & \bar{\mathbf{a}}^2\mathbf{I} & 0 & \cdots \\ 0 & 0 & \bar{\mathbf{a}}^3\mathbf{I} & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{bmatrix}, \tag{59}$$

where $\bar{\mathbf{a}}$ is a diagonal matrix representing the Lorenz/Mie coefficients and $\mathbf{H}$ is a matrix of the Hankel-function addition coefficients. The above system is now contracted by the operation

$$\begin{bmatrix} \mathbf{I} & \bar{\mathbf{a}}^1\mathbf{H}^{12} & \bar{\mathbf{a}}^1\mathbf{H}^{13} & \cdots \\ \bar{\mathbf{a}}^2\mathbf{H}^{21} & \mathbf{I} & \bar{\mathbf{a}}^2\mathbf{H}^{23} & \cdots \\ \bar{\mathbf{a}}^3\mathbf{H}^{31} & \bar{\mathbf{a}}^3\mathbf{H}^{32} & \mathbf{I} & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{bmatrix} \begin{bmatrix} \mathbf{T}^{11} & \mathbf{T}^{12} & \mathbf{T}^{13} & \cdots \\ \mathbf{T}^{21} & \mathbf{T}^{22} & \mathbf{T}^{23} & \cdots \\ \mathbf{T}^{31} & \mathbf{T}^{32} & \mathbf{T}^{33} & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{bmatrix} \times \begin{pmatrix} \mathbf{J}^{10} \\ \mathbf{J}^{20} \\ \mathbf{J}^{30} \\ \vdots \end{pmatrix} = \begin{bmatrix} \bar{\mathbf{a}}^1\mathbf{I} & 0 & 0 & \cdots \\ 0 & \bar{\mathbf{a}}^2\mathbf{I} & 0 & \cdots \\ 0 & 0 & \bar{\mathbf{a}}^3\mathbf{I} & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{bmatrix} \begin{pmatrix} \mathbf{J}^{10} \\ \mathbf{J}^{20} \\ \mathbf{J}^{30} \\ \vdots \end{pmatrix}, \tag{60}$$

where $\mathbf{J}$ is the matrix of Bessel-function addition coefficients as defined in Eq. (5). By defining

$$\mathbf{T}^i = \sum_{j=1}^{N_S} \mathbf{T}^{ij}\mathbf{J}^{j0}, \tag{61}$$

we obtain

$$\begin{bmatrix} \mathbf{I} & \bar{\mathbf{a}}^1\mathbf{H}^{12} & \bar{\mathbf{a}}^1\mathbf{H}^{13} & \cdots \\ \bar{\mathbf{a}}^2\mathbf{H}^{21} & \mathbf{I} & \bar{\mathbf{a}}^2\mathbf{H}^{23} & \cdots \\ \bar{\mathbf{a}}^3\mathbf{H}^{31} & \bar{\mathbf{a}}^3\mathbf{H}^{32} & \mathbf{I} & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{bmatrix} \begin{pmatrix} \mathbf{T}^1 \\ \mathbf{T}^2 \\ \mathbf{T}^3 \\ \vdots \end{pmatrix} = \begin{pmatrix} \bar{\mathbf{a}}^1\mathbf{J}^{10} \\ \bar{\mathbf{a}}^2\mathbf{J}^{20} \\ \bar{\mathbf{a}}^3\mathbf{J}^{30} \\ \vdots \end{pmatrix}. \tag{62}$$

Returning to the tensorial form of notation, the system in Eq. (62) can be written as

$$T^i_{mnp\ klq} = \bar{a}^i_{np} J^{i0}_{mnp\ klq} - \bar{a}^i_{np} H^{ij}_{mnp\ m'n'p'} T^j_{m'n'p'\ klq}. \tag{63}$$

As above, summation over all indices not appearing on the left-hand side is implied, and it is also noted that $H^{ij}_{mnp\ klq} \equiv 0$ and $J^{ij}_{mnp\ klq} = \delta_{nl}\delta_{mk}\delta_{pq}$ for $i = j$. Upon solution of the above system for the T^i matrices the cluster-centered T matrix is obtained from the second contraction:

$$T_{mnp\ klq} = J^{0i}_{mnp\ m'n'p'} T^i_{m'n'p'\ klq}. \tag{64}$$

Our procedure for calculating the T matrix of the cluster is as follows. For successive values of the column indices l, k, and q we solve Eq. (63) for the column vectors of T^i by using an iteration method. Next, the column vectors of T^i are multiplied into J^{0i}, and the resulting vector is added to the T matrix as described in Eq. (64). It is therefore not necessary in this process to store in memory the complete T^i matrices; rather, the columns of T^i are calculated sequentially and then discarded once their contribution to the T matrix has been determined. In addition, the matrix multiplication in Eq. (64) is carried out only for row orders n up to and including the current value of the column order l. Values of the T matrix for $n > l$ are obtained from the symmetry relation

$$T_{mnp\ klq} = (-1)^{m+k} \frac{l(l+1)(2n+1)}{n(n+1)(2l+1)} T_{-klq\ -mnp}. \tag{65}$$

In implementing the method, we must establish two separate truncation limits on harmonic order for each sphere. The maximum value of the row order n in $T^i_{mnp\ klq}$, denoted as $N_{O,i}$, can usually be fixed *a priori* from the number of Lorenz/Mie terms required to represent the single-sphere properties of sphere i. It should be noted, however, that for certain cases the required value of $N_{O,i}$ for contacting spheres can significantly exceed that required for isolated spheres, especially when $x_i \ll 1$ and the real and imaginary parts of the refractive index are large.[11,21] The truncation limit on the column order l in $T^i_{mnp\ klq}$, again for sphere i, can be determined from the convergence of the orientation-averaged extinction efficiency of sphere i. This quantity, which is given by[11]

$$\langle Q_{\mathrm{ext},i} \rangle = \frac{2}{x_i^2} \mathrm{Re}\left(\sum_{n,m,p} \sum_{l,k,q} J^{0i}_{klq\ mnp} T^i_{mnp\ klq} \right), \tag{66}$$

can be easily calculated during the course of performing Eq. (64). The maximum value of l is fixed when the relative contribution to Eq. (66) for successive values of l decreases below a given tolerance.

It should be emphasized that each T^i matrix does not need to be calculated to a column order l equal to the maximum order N_O of the cluster T matrix—it only needs to be calculated out to a value of l that results in an acceptable convergence of Eq. (66). Once the T^i matrix converges, it can be removed from the equations, and because of this the number of equations to be solved in Eq. (63) will decrease as the entire T matrix approaches convergence. Spheres that are closer to the cluster origin will have their T^i matrices converge in a smaller number of orders than the more distant spheres.

An additional property that can be obtained directly from the procedure is the random-orientation absorption efficiency of the individual spheres,[11] which is given by

$$\langle Q_{\mathrm{abs},i} \rangle = \frac{2}{x_i^2} \sum_{n,m,p} \sum_{l,k,q} \bar{d}^i_{np} \frac{n(n+1)(2l+1)f_{mn}}{l(l+1)(2n+1)f_{kl}} \times T^i_{mnp\ klq} T^{i*}_{mnp\ klq}, \tag{67}$$

where $\bar{d}_{np}$ is a real-valued quantity that is related to the Lorenz/Mie coefficient of sphere i.

As mentioned above, an advantage of this scheme is that it obviates the full storage in memory of the T^{ij} matrix, which is unavoidable when direct methods (i.e., Gaussian elimination) are used to solve Eq. (59). In addition, the method allows one to exploit directly the factorization of the H and J matrices into rotational and axial translation parts, i.e.,[3]

$$\mathbf{H}^{ij} = H_{mnp\,klq}(r_{ij}, \theta_{ij}, \phi_{ij}) = (-1)^m \exp[i(k-m)\phi_{ij}] \sum_{m'} (-1)^{m'} \times \mathscr{D}^{m'}_{mn}(\theta_{ij}) H_{m'np\,m'lq}(r_{ij}, 0, 0) \mathscr{D}^{k}_{m'}(\theta_{ij}), \tag{68}$$

where r_{ij}, θ_{ij}, and ϕ_{ij} denote the polar coordinates that describe the origin of i relative to j. An identical expansion can be obtained for the J^{0i} matrix. The number of operations required to perform a multiplication of H^{ij} with a vector will be of the order of $N^3_{O,i}$ for the factored matrix, as opposed to $N^4_{O,i}$ for the unfactored matrix. Memory requirements for storage of the factored matrices are also reduced in a proportional manner.

Nevertheless, the most critical factor in the execution time is the number of iterations required to solve Eq. (63) for a given right-hand-side vector. An *a priori* estimate of the minimum number of iterations is not obvious—suffice it to say that the proximity of the spheres to each other, and the resonant behavior of the spheres, can strongly influence the convergence rate.[7,22] Empirically, however, we have found that the conjugate gradient method provides the most rapid convergence to the solution, which is consistent with findings for the discrete-dipole representation of particle scattering.[23]

Calculation of the A, B, and D matrices in Eqs. (27)–(29) involves a numerical effort that can become comparable with that required in calculation of the T matrix. Considering that the indices w, v, and u run from 0 to $2N_O$, $-w$ to w, and $-N_O - 1$ to $N_O + 1$, respectively, calculation of A, B, and D each involves of the order of ${N_O}^5$ operations (not including calculation of the $\hat{C}$'s). In addition, complete storage of the A and B matrices will require of the order of $8{N_O}^4$ locations, which is greater than that required for the T matrix itself. In practice, we do not store the A and B matrices for all values of w and v prior to calculating the D matrix from Eq. (27). Rather, the operations in Eqs. (27)–(29) are performed sequentially within a common, nested loop over v and w. For the special case of axially symmetric sphere configurations—for which $T_{mnp\,klq}$ is nonzero only for $m = k$—the A and B matrices in Eqs. (28) and (29) will be zero for all v except $v = 0$.

3. RESULTS AND DISCUSSION

A. Numerical Performance

The veracity of the formulation and the code under which it is implemented were proven by several tests. Total cross sections and scattering matrix values for fixed orientation were calculated from the generated T matrix and were found to be in perfect agreement with results obtained from direct iterative solution of Eq. (3). Random-orientation cross sections and scattering matrix elements for linear chains were found to be independent of the configuration of the chain in the cluster frame (i.e., aligned along the x, y, or z axes) and were identical to our previously published values.[10] Orientation-averaged scattering matrix elements were shown to obey the relations $S_{22}(0) = S_{33}(0)$, $S_{22}(\pi) = -S_{33}(\pi)$, and $S_{11}(\pi) - 2S_{22}(\pi) = S_{44}(\pi)$.[24] In addition, the random-orientation matrix elements for clusters possessing a plane of symmetry (not necessarily axisymmetric chains) were nonzero only for S_{11}, S_{22}, S_{33}, and S_{44} and for S_{12} and S_{34} and their transposes.

We found that scheme presented in Eqs. (63) and (64) can shorten considerably the execution time required to calculate the T matrix and the random-orientation properties of a cluster. To provide a relative indication, we note that calculation of the T matrix for a close-packed cluster of 11 spheres, each sphere having a size parameter of 4 (with $N_{O,i} = 7$) and a refractive index of $1.5 + 0.005i$ required 38 min of CPU time on a SUN SPARC 20 workstation. For this case the maximum order of the T matrix was $N_O = 21$. Calculation of the orientation-averaged scattering matrix expansion coefficients required an additional 8 min. In comparison, direct calculation of the T^{ij} matrix required 38 min, followed by 31 min to contract T^{ij} into T. As another measure, we calculated the T matrix of a fractal-like cluster of 250 spheres (i.e., a soot aggregate) with sphere properties of $x = 0.3$ and $m = 1.6 + 0.6i$.[21] One harmonic order (including both the electric and magnetic dipole terms) were used in the sphere expansions, and the T matrix was truncated at $N_O = 13$ orders. Calculation of the T matrix by means of the present method required 28 min, followed by 1 min to calculate the random-orientation scattering matrix coefficients. Direct calculation of the T^{ij} matrix, on the other hand, required 160 min. These two examples bear out our general conclusion that the present method offers the greatest improvement over the direct method when the size of the T matrix is significantly less than the size of the T^{ij} matrix (i.e., $M \ll N_S M_i$).

As a further point of comparison, calculation of the orientation-averaged matrix elements by numerical quadrature of the fixed-orientation values typically required overnight runs for the clusters described above. We consistently found that ~4-digit accuracy in the matrix elements was obtained when the total number of quadrature points—and the total number of right-hand sides to be solved by iteration—was always well in excess of the number of right-hand sides in Eq. (63). In view of this, the scheme developed here can be viewed as the optimum iterative method to finding the orientation-averaged properties of the cluster.

The basic formulation and approach developed here would also have direct application to the discrete-dipole model of scattering from nonspherical particles,[8,23] which can be obtained from the general, multipole formulation in the limit of small sphere size parameter. Indeed, the T-matrix formulation presented here, and the associated methods for calculating the random-orientation scattering matrix elements, can be viewed as a multipole generalization of the dipole-based formulations developed by Singham *et al.*[25] and McClain and Ghoul.[26] The advantage of our method is that it would provide an efficient means of calculating the T matrix of the particle from the discrete-dipole procedure—in that it would obviate the direct numerical inversion of the coefficient matrix. Judging from our calculations on finite-sized clusters of spheres, we would expect that this approach provides a significantly faster method of obtaining the random-

orientation properties of the discretized particle than the traditional method of numerical quadrature.

There certainly is ample potential for further optimization of the mathematical formulations and the numerical procedures. For example, exploitation of symmetry properties of the cluster (beyond the obvious axisymmetric configuration) could significantly reduce the computational times.[20] Optimized iteration strategies for solution of Eq. (63) also need to be developed. In addition, the method can have serious drawbacks for disperse clusters of widely separated spheres. As has been noted in previous work,[11,27] such systems do not lend themselves well to a single, cluster-centered T-matrix description, simply because the number of T-matrix orders that are required to expand the partial fields about a common origin can become excessive. For these systems it would be advantageous to retain a multiple-origin representation of the scattering from the cluster (i.e., the T^{ij} matrix), which would require a completely different formulation of the orientation-averaged scattering matrix elements and expansion coefficients.

B. Calculation Results

We will present a comprehensive examination of the scattering properties of sphere clusters in a forthcoming paper. For the present purposes we wish to present a small sample of results and point out some salient features of scattering from clusters. In Figs. 1 and 2 we plot the scattering matrix elements, as a function of scattering angle θ', for two distinctly different types of clusters. In Fig. 1 the cluster is a straight chain, whereas in Fig. 2 the spheres are packed into a hexagonal lattice. These two configurations represent extrema in packing density of the spheres. In both plots the number of spheres in the cluster, N_S, ranges from 1 to 5, and the size parameter and the refractive index of the individual spheres are 5

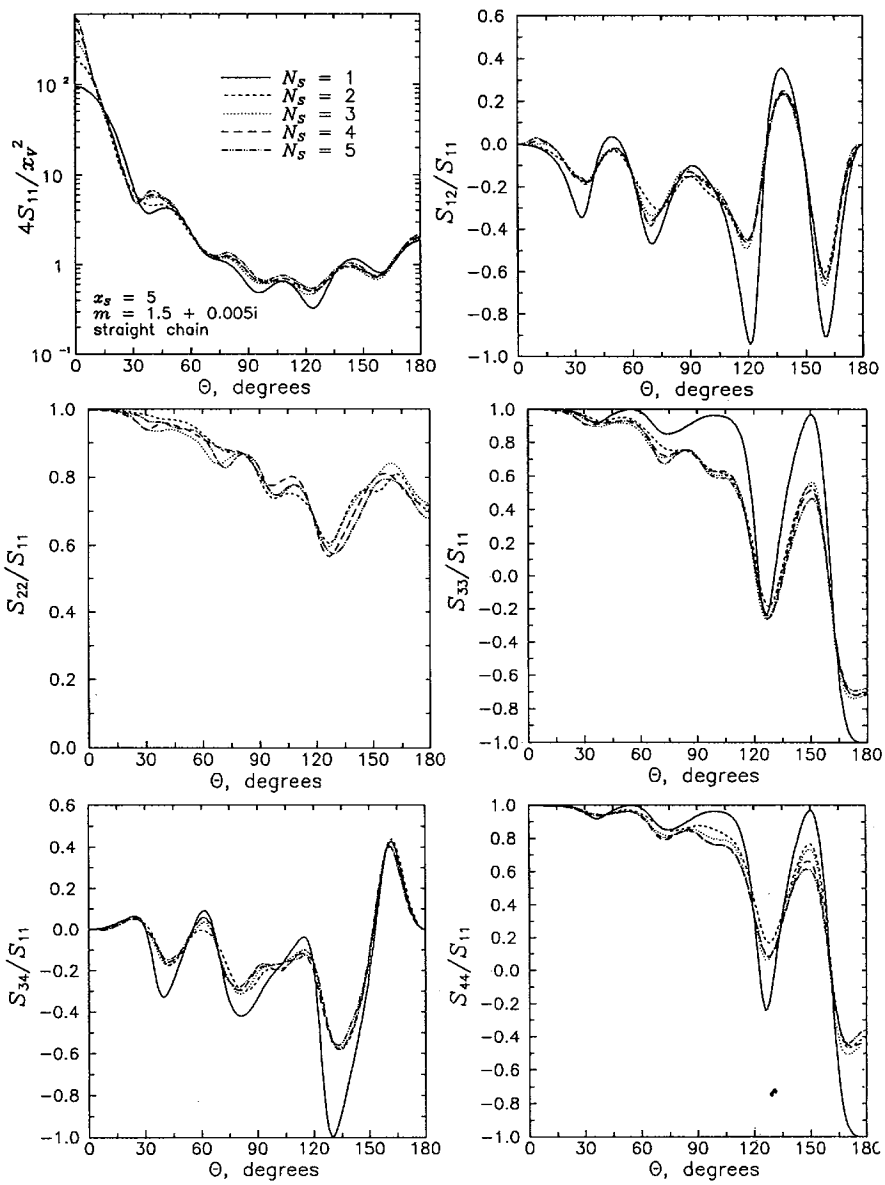

Fig. 1. Orientation-averaged scattering matrix elements for a linear chain of spheres.

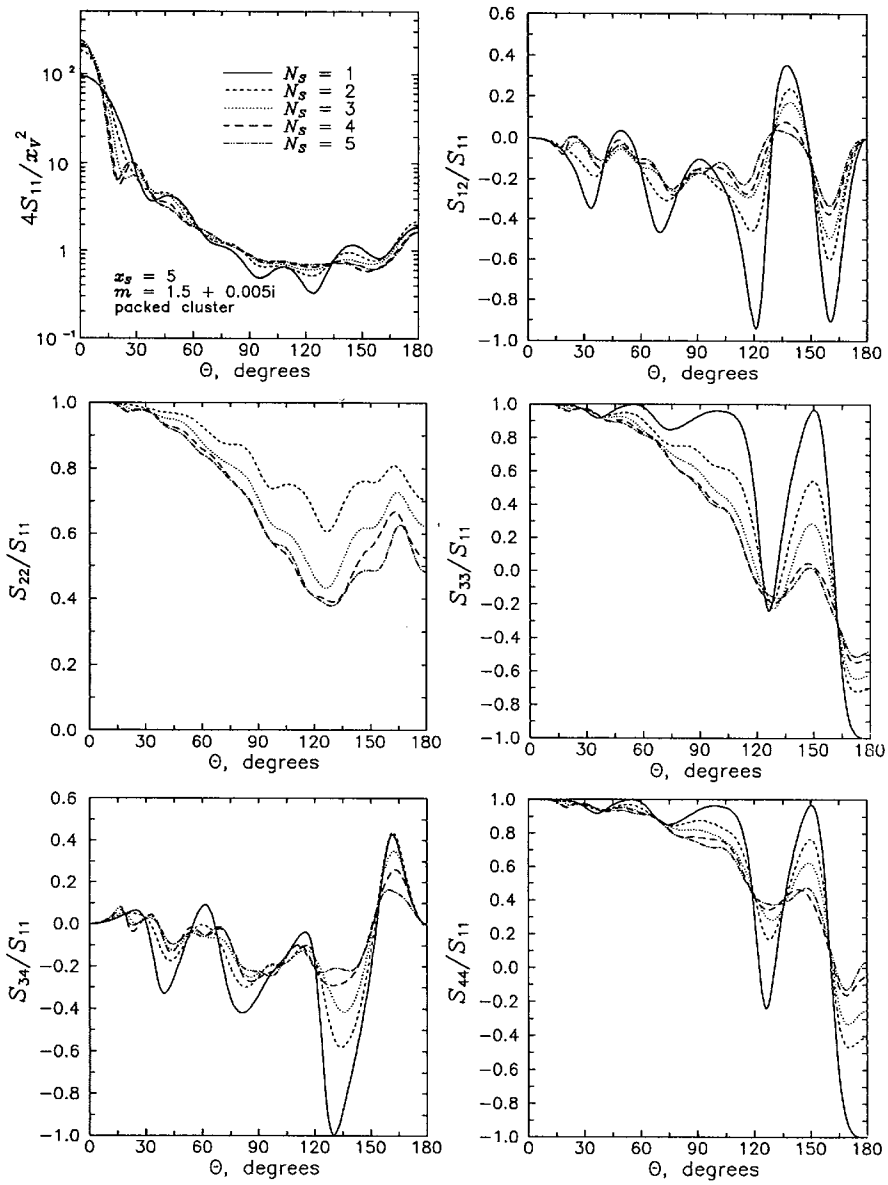

Fig. 2. Orientation-averaged scattering matrix elements for a packed cluster of spheres.

and 1.5 + 0.005i, respectively. The matrix element S_{11} has been normalized with $4/x_V{}^2$, where x_V is the volume-mean size parameter, and the other elements have been normalized with S_{11}.

A quick glance at both figures reveals that the configuration of the spheres can have a significant effect on the scattering properties of the cluster. Aside from the increase in the forward-direction value of S_{11}, the matrix elements for the chain (Fig. 1) attain a form that is nearly independent of N_S for $N_S \geq 2$. Clustering results in a damping of the oscillations in the matrix elements compared with those in the single sphere, yet the locations of extrema in the oscillations for the chain are nearly identical to those for the single sphere—with the exception, of course, of S_{22}/S_{11}, which is identically unity for the sphere. On the other hand, the matrix elements for the packed cluster are significantly altered by increasing N_S and appear to approach a saturation level in which all oscillations are damped out. The effect of clustering for this configuration is especially noticeable in the backward-direction values of S_{22}, S_{33}, and S_{44}.

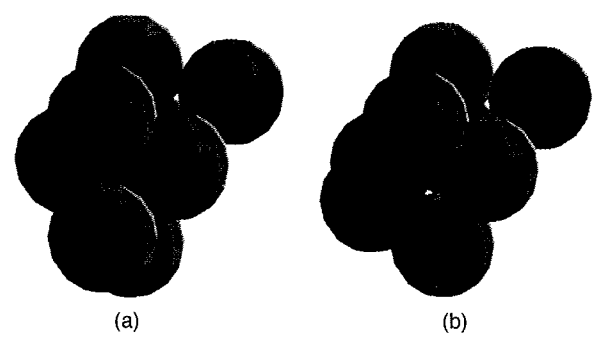

Fig. 3. (a) Asymmetric and (b) symmetric tetrahedral-lattice clusters of eight spheres.

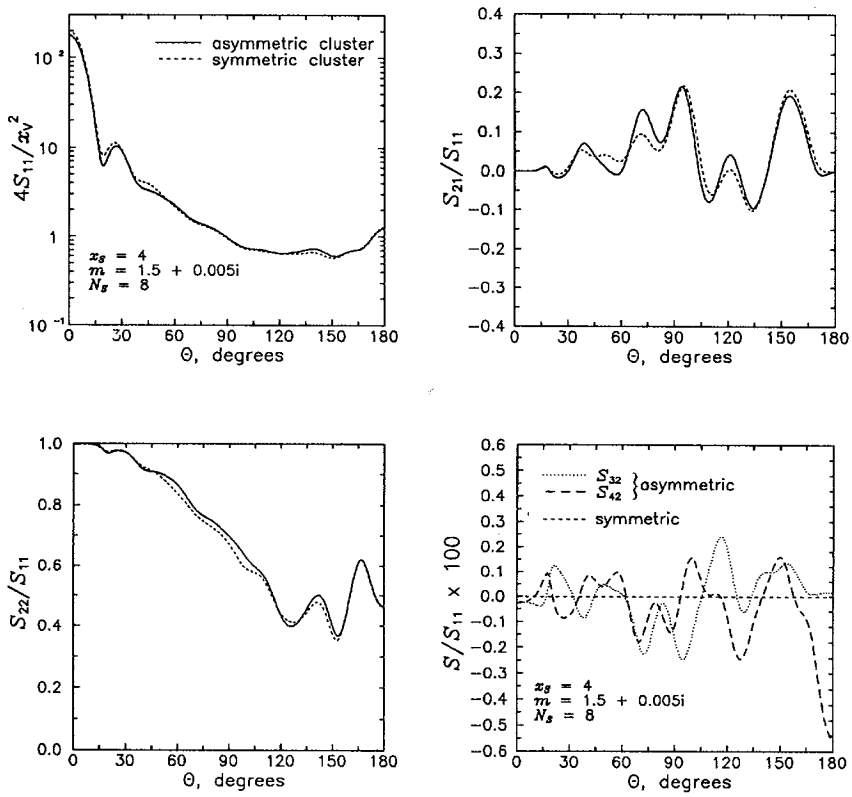

Fig. 4. Orientation-averaged scattering matrix elements for the symmetric and asymmetric clusters.

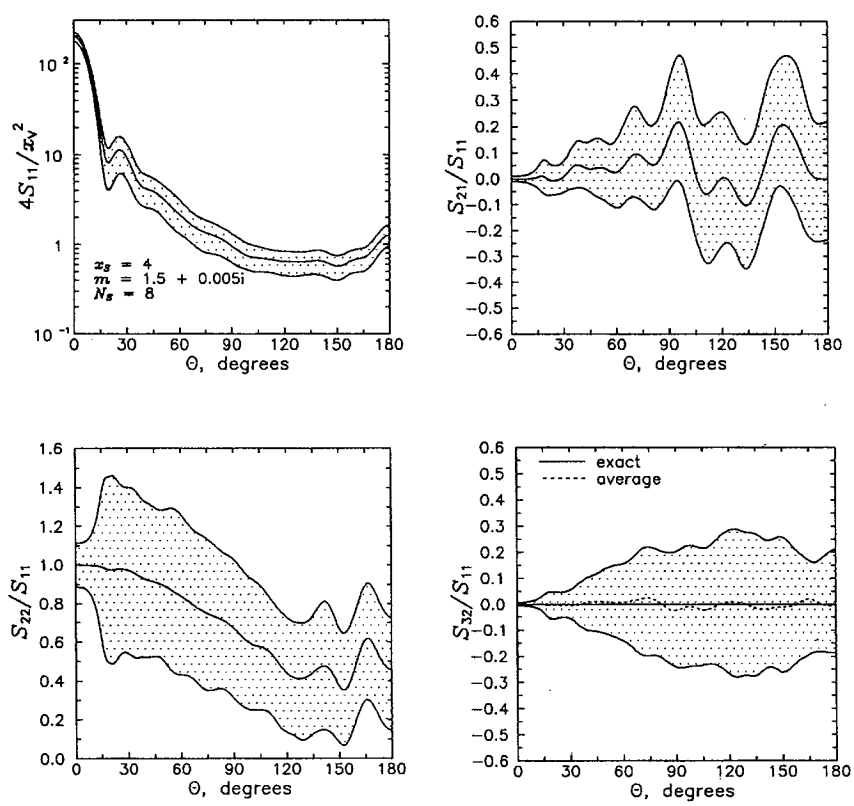

Fig. 5. Variation in fixed-orientation matrix elements for the eight-sphere symmetric cluster. The dotted area represents 1 standard deviation.

To explain the differences in scattering between these two configurations, we first note that clustering will affect scattering by the two mechanisms of far-field wave interference and near-field interactions (or, equivalently, multiple scattering). When the size parameters of the spheres are of the order of unity or greater (as is the case here), orientation averaging acts to zero out the effects of interference in all but the forwardmost scattering directions. The differences between the single-sphere and orientation-averaged multiple-sphere scattering patterns are therefore an effect primarily of multiple scattering. Multiple scattering for the chain configuration occurs primarily between neighboring spheres on the chain, and because of this the matrix elements do not change appreciably as N_S increases—except for the effect of interference on $S_{11}(\theta' \sim 0)$. On the other hand, the packed-cluster configuration offers a much higher opportunity for multiple scattering among the spheres—and therefore results in a greater difference in the random-orientation elements relative to the single-sphere values.

The configurations used in Figs. 1 and 2 all possess a plane of symmetry, and because of this the elements S_{23}, S_{24}, S_{13}, and S_{14} and their transposes were zero. To examine the magnitude of these terms for nonsymmetric clusters—and also to test the veracity of the code—we performed calculations on two configurations of eight spheres. The configurations represent two isomers of a hexagonal lattice and are shown in Fig. 3. Note that the only difference between the two configurations is the placement of one sphere, with the cluster in (a) forming an asymmetric configuration and the cluster in (b) being symmetric. Orientation-averaged scattering matrix results for the two configurations are presented in Fig. 4, where we show the S_{11}, S_{21}, S_{22}, S_{32}, and S_{42} elements. The sphere refractive index is the same as that above, and the sphere size parameter is 4. The major elements S_{11}, S_{21}, and S_{22} are nearly identical for the two configurations, which is expected, since the configurations are nearly identical. The asymmetric cluster, however, has small ($\sim 10^{-3} S_{11}$) yet distinctly nonzero values of S_{32} and S_{42}.

To give an indication of the variation in matrix elements over cluster orientation, we calculated matrix elements for the eight-sphere, symmetric cluster for a large number of fixed, randomly chosen orientations. The results of these calculations are given in Fig. 5. The dotted area in the plots represents a 1-standard-deviation spread in the calculated matrix elements for 1000 random orientations, and the solid curve in the center of the dotted area represents the exact, orientation-averaged results. We have also presented, for S_{32}, the mean of the fixed-orientation values. The relatively large spread in the results indicates that the matrix elements can be highly sensitive to orientation, which was observed for bispheres in Ref. 10. This behavior has bearing on the validity of a random-orientation model for scattering by clusters. It would be expected that actual particles suspended in gases or liquids attain a somewhat biased orientation because of differences in the gravitational and hydrodynamic centers of the particles. Experimentally measured matrix elements of such particles could be significantly different from those predicted from a random-orientation assumption, especially for the higher-order elements such as S_{32}.

APPENDIX A: ADDITION COEFFICIENTS, ROTATION FUNCTIONS, AND VECTOR COUPLING COEFFICIENTS

Relations for calculation of the addition coefficients are presented in Refs. 3 and 21. The first reference contains errors in the axial-translation formulas. Corrected formulas are given here. The vector addition coefficients for axial translation over a positive distance z_{ij} can be obtained from the scalar addition coefficients $C^{ij}_{mn\,ml}$:

$$H^{ij}_{mnp\,mlp} = z_{ij}\left[\frac{n+m+1}{(n+1)(2n+3)} C^{ij}_{mn+1\,ml} + \frac{n-m}{n(2n-1)} C^{ij}_{mn-1\,ml}\right] + C^{ij}_{mn\,ml}, \quad \text{(A1)}$$

$$H^{ij}_{mnp\,ml3-p} = z_{ij}\frac{im}{n(n+1)} C^{ij}_{mn\,ml}. \quad \text{(A2)}$$

The scalar coefficient (again for positive axial translation) can be obtained by recurrence relations[3,21] or by the direct formula

$$C^{ij}_{mn\,ml} = (-1)^m i^{n-l}(2n+1) \times \sum_w i^w a(m, l; -m, n; w) h_w(r_{ij}), \quad \text{(A3)}$$

where h_w is the spherical Hankel function and $a(m, l; -m, n; p)$ are Gaunt coefficients, which are defined by the linearization

$$P_l^m(\cos\theta) P_n^{-m}(\cos\theta) = \sum_w a(m, l; -m, n; w) P_w(\cos\theta), \quad \text{(A4)}$$

where w takes on the values of $w = |n-l|, |n-l| + 2, \ldots, n+l$. Recognizing that $P_n^m = \mathscr{D}_{0n}^m$ and using Eq. (21), we see that the Gaunt coefficients are equivalent to

$$a(m, l; -m, n; w) = (-1)^{n+l+w} \hat{C}^w_{ml,-mn} \hat{C}^w_{0l,0n}. \quad \text{(A5)}$$

The formulas for the J^{ij} coefficients are identical to Eqs. (A1) and (A2), except that the C^{ij} coefficients are now based on the spherical Bessel function in Eq. (A3). For the case of a negative axial translation Eqs. (A1) and (A2) would be multiplied by $(-1)^{n+l}$ and $(-1)^{n+l+1}$, respectively.

The rotation coefficients $\mathscr{D}_{lm}^m$ are related to the generalized spherical functions[14,18] $d_{km}^{(n)}$ by

$$\mathscr{D}_{kn}^m = (-1)^{m+k}\left[\frac{(n-k)!(n+m)!}{(n+k)!(n-m)!}\right]^{1/2} d_{km}^{(n)}. \quad \text{(A6)}$$

A recurrence relation for calculating the functions upward in m is

$$\mathscr{D}_{kn}^0(\beta) = P_n^{-k}(\cos\beta), \quad \text{(A7)}$$

$$\mathscr{D}_{kn}^{m+1} = \cos^2(\beta/2)\mathscr{D}_{k-1\,n}^{m} - (n-k)(n+k+1) \times \sin^2(\beta/2)\mathscr{D}_{k+1\,n}^{m} - k(\sin\beta)\mathscr{D}_{kn}^{m}. \quad \text{(A8)}$$

Values for negative m are obtained from $\mathscr{D}_{kn}^{-m} = (-1)^{m+k} f_{-mn} f_{-kn} \mathscr{D}_{-kn}^{m}$ and $\mathscr{D}_{-kn}^{-m} = \mathscr{D}_{mn}^{k}$.

The $\hat{C}$ coefficients appearing in this work are related to the vector coupling coefficients[14,18] by

$$\hat{C}_{mn,kl}^{w} = \left(\frac{f_{mn} f_{kl}}{f_{m+k\,w}}\right)^{1/2} C_{mn,kl}^{m+k\,w}.$$

The $\hat{C}$ coefficients can be calculated by a relatively simple procedure. First define the auxiliary coefficient S by

$$\hat{C}_{mn,kl}^{w} = g_{nlw}(n+m)!(l+k)!(w-m-k)!S_{mn,kl}^{w}, \quad \text{(A9)}$$

where

$$g_{nlw} = \left[\frac{(2w+1)(n+l-w)!(w+n-l)!(w+l-n)!}{(n+l+w+1)!}\right]^{1/2}. \quad \text{(A10)}$$

The S coefficients, in turn, obey the three-term downward recurrence relation

$$S_{mn,kl}^{w-1} = b_w S_{mn,kl}^{w} + c_w S_{mn,kl}^{w+1}, \quad \text{(A11)}$$

with starting values of

$$b_w = \frac{(2w+1)\{(m-k)w(w+1) - (m+k)[n(n+1) - l(l+1)]\}}{(w+1)(n+l-w+1)(n+l+w+1)}, \quad \text{(A12)}$$

$$c_w = \frac{w(w+n-l+1)(w+l-n+1)(w+m+k+1)(w-m-k+1)}{(w+1)(n+l-w+1)(n+l+w+1)}, \quad \text{(A13)}$$

$$S_{mn,kl}^{n+l+1} = 0,$$

$$S_{mn,kl}^{n+l} = \frac{1}{(n-m)!(l+k)!(n+m)!(l-k)!}. \quad \text{(A14)}$$

The minimum value of w will be the larger of $|n-l|$ and $|m+k|$. Recurrence formulas for S in the n and l indices can be obtained from the formulas

$$\begin{aligned} S_{mn,kl}^{w} &= (-1)^{w+n+k} S_{m+kw,-kl}^{n} \\ &= (-1)^{w+n+k} S_{kl,-m-k\,w}^{n} \\ &= (-1)^{n+m} S_{mn,-m-k\,w}^{l} \\ &= (-1)^{n+m} S_{m+k\,w,-mn}^{l}. \end{aligned} \quad \text{(A15)}$$

ACKNOWLEDGMENTS

The authors acknowledge the support of a DuPont Educational Assistance Grant (D. W. Mackowski) and of the NASA Office of Mission to Planet Earth (M. I. Mishchenko).

Address all correspondence to Daniel W. Mackowski; e-mail: dmckwski@eng.auburn.edu.

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