PHYSICAL REVIEW B VOLUME 22, NUMBER 8 15 OCTOBER 1980

Theory of resonances in the electromagnetic scattering by macroscopic bodies

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(Received 28 January 1980)

The electromagnetic scattering resonances of a collection of macroscopic bodies with uniform electric properties are used to construct a spectral representation for the scattered field. The resonances and their weights are found by solving for the eigenvalues and eigenstates of a non-Hermitian, linear integral operator Γ. A scheme is developed for doing this by diagonalizing a matrix that represents Γ by the set of individual grain eigenstates—the diagonal elements are individual grain eigenvalues while the off-diagonal elements are overlap integrals of eigenstates from two different grains. For a system of spherical scatterers, this scheme leads to a reasonable method of calculating numerically the scattered field in cases where the multiple scattering is important. As an example, the scattering by a pair of identical spheres is worked out analytically for a limiting case. Sum rules for the weights in the spectral representation are derived and discussed.

I. INTRODUCTION

There are many important situations where electromagnetic waves are scattered by a collection of particles each of which is large enough to be described as a homogeneous body with a definite shape. The scattering is caused by the fact that these bodies are made of a material whose complex dielectric constant κ_1 and magnetic permeability μ_1 differ from those of the surrounding medium κ_2, μ_2. In the last several years, this classical problem has undergone a revival of interest. This interest is produced in part by practical considerations, for example, the possibility of using a system of such particles as a selective absorber of solar radiation, and partly because of the fundamental questions that are raised. The propagation of electromagnetic radiation among a collection of particles is, in fact, formally related to the propagation of electrons among collections of atoms. Among recent studies,[1] we may mention several mean-field treatments of electromagnetic propagation in inhomogeneous media,[2,3] as well as a multiple-scattering approach, applicable especially to periodic arrays of spheres, which proceeds by analogy with Green's-function treatments of electron propagation in ordered solids.[4]

In this article we discuss the scattering problem in the case when $\mu_1 = \mu_2 = 1$, and when all the scatterers have the same value of $\kappa_1 \neq \kappa_2$. We show that a spectral representation can be set up for the scattered field as a function of κ_1/κ_2, in terms of certain resonances which are special values of κ_1/κ_2, obtained by solving an eigenvalue problem.

The spectral representation is useful if a convenient scheme can be found for calculating the eigenvalues and the eigenfunctions (the latter are needed in order to determine the residues in the spectral representation). We propose such a scheme where one first has to find the eigenstates of each isolated scattering body or grain. The eigenstates of the entire scattering system, as well as the scattered field, can then be expanded in these individual grain eigenstates. The mathematical problem to be solved thus becomes a system of linear algebraic equations for the expansion coefficients. The matrix of this system has diagonal elements which are individual grain eigenvalues, and off-diagonal elements which are overlap integrals between eigenstates of different grains. This scheme is especially useful if the individual grains have sufficiently simple shapes so that their eigenstates, as well as the overlap integrals, can be calculated analytically or nearly so. That is the case for spherical grains, which we discuss as an example in this article.

Other features of the spectral representation are that it enables us to derive exact sum rules for the residues of the scattering resonances. The direct calculation of a resonance, where the scattered field diverges, is, of course, a much better procedure than to try to calculate the scattered field itself there, since any approximate method for calculating the latter quantity will break down sufficiently close to the resonance. Finally, the spectral representation achieves a kind of separation of the physics of the scattering problem from its microscopic geometry: the positions of the resonances depend only on the wave vector of the incident wave and on the microscopic geometry of the scatterer. The residues also depend on the direction or the point of observation. But both types of quantities are independent of the intrinsic physical properties of the

scatterer, i.e., the value of κ_1/κ_2. Thus, once we have found the eigenstates, we have solved for the dependence on the geometry, and we can substitute any value we desire for the physical parameter κ_1/κ_2.

The approach taken in this article to the scattering problem of electromagnetic waves follows ideas which are similar to those applied by one of the authors (D. J. B.) to the problem of the effective static dielectric constant of a composite material.[5,6] As far as we know, such an approach has never been applied before to the scattering problem.

In Sec. II we describe the general method for a system of macroscopic scattering grains, and develop the necessary formalism. In Sec. III we specialize the discussion to a system of scattering spheres, and set up the necessary equipment for performing a calculation for any number of interacting spheres. We do this particularly for the long-wavelength limit when all the radii, as well as the particle separations, are small compared to the wavelength outside the scatterers. We get an explicit expression for the field scattered by two identical spheres whose separation vector $\vec{b}$ is parallel to the incident wave vector $\vec{k}_0$, and whose radius a is much smaller than $|\vec{b}|$.

In Sec. IV we discuss the advantages of our method, as well as its relationship to existing discussions of the electromagnetic-scattering problem.[7,8] Possible applications are suggested.

In Appendix A we calculate the eigenfunctions and eigenvalues of an isolated sphere, in particular detail for the case $k_0 a \ll 1$, and use them to obtain an expression for the field scattered by a single sphere in the long-wavelength limit. In Appendix B we calculate expressions for the matrix elements between eigenstates of two spheres, again in particular detail for the case where the radii as well as the separation are much smaller than the incident wavelength.

II. GENERAL FORMALISM AND BASIC PROPERTIES

We consider a homogeneous, source-free host medium characterized by material parameters $\epsilon_2, \sigma_2, \mu_2$ into which some inclusions of a different homogeneous material $\epsilon_1, \sigma_1, \mu_1$ have been introduced. The inclusions occupy a finite, limited volume. We will assume that $\mu_1 = \mu_2 = 1$, and attempt to calculate the total electric field $\vec{E}$ (i.e., incident plus scattered) that arises when the incident field is $\vec{E}_0 \sim e^{-i\omega t}$. The conductivity σ and the dielectric constant ϵ will be lumped together in the usual way in a complex, frequency-dependent dielectric constant κ that can have two different values at different points in space. We represent it in the following form:

$$\kappa(\vec{r}) = \kappa_1\Theta_1(\vec{r}) + \kappa_2\Theta_2(\vec{r}) = \kappa_2[1 - u\Theta_1(\vec{r})],$$
$$\kappa_i \equiv \epsilon_i - 4\pi\sigma_i/i\omega, \quad i = 1, 2 \tag{2.1}$$
$$u \equiv 1 - \kappa_1/\kappa_2,$$

where $\Theta_2(\vec{r})$ $[\Theta_1(\vec{r})]$ is a step function set equal to 1 for $\vec{r}$ inside the host medium (inclusions), and set equal to 0 outside it. Maxwell's source-free equations then become

$$\vec{\nabla}\cdot\kappa\vec{E} = 0, \quad \vec{\nabla}\times\vec{E} = \frac{i\omega}{c}\vec{H},$$
$$\vec{\nabla}\cdot\vec{H} = 0, \quad \vec{\nabla}\times\vec{H} = -\frac{i\omega}{c}\kappa\vec{E}, \tag{2.2}$$

and from these equations we can obtain the following equation for $\vec{E}$:

$$-\vec{\nabla}\times(\vec{\nabla}\times\vec{E}) + k^2\vec{E} = uk^2\Theta_1\vec{E},$$
$$k^2 \equiv \omega^2\kappa_2/c^2. \tag{2.3}$$

In order to find $\vec{E}(\vec{r})$ we must solve this equation together with the usual boundary condition for scattering, i.e., that at large distances $\vec{E}(\vec{r})$ must be equal to the incident field $\vec{E}_0(\vec{r})$ plus an outgoing field $\sim e^{ikr}/r$.

In order to transform this problem into an integral equation, we need to use the tensor Green's function $\vec{G}(\vec{r} - \vec{r}', k)$ defined by

$$-\vec{\nabla}\times(\vec{\nabla}\times\vec{G}) + k^2\vec{G} = k^2\delta^3(\vec{r} - \vec{r}')\vec{I}, \tag{2.4}$$

where $\vec{I}$ is the unit tensor and where $\vec{G}$ must be an outgoing wave at large distances. The solution for $\vec{G}$ is just

$$G_{\alpha\beta}(\vec{R}, k) = (k^2\delta_{\alpha\beta} + \nabla_\alpha\nabla_\beta)e^{ikR}/(-4\pi R). \tag{2.5}$$

When this is used to "solve" Eq. (2.3) by treating its right-hand side (rhs) as if it were known, the following integral equation is obtained:

$$\vec{E}(\vec{r}) = \vec{E}_0(\vec{r}) + u\int d^3r'\Theta_1(\vec{r}')\vec{G}(\vec{r} - \vec{r}', k)\cdot\vec{E}(\vec{r}'), \tag{2.6}$$

where the appearance of $\vec{E}_0$ ensures that $\vec{E}$ satisfies the correct boundary conditions. We will represent the linear integral operator appearing in this equation by Γ, and the equation will then be written symbolically as

$$E = E_0 + u\Gamma E, \tag{2.7}$$

where

$$\Gamma E \equiv \int d^3r'\Theta_1(\vec{r}')\vec{G}(\vec{r} - \vec{r}', k)\cdot\vec{E}(\vec{r}') \tag{2.8}$$

and where we have dispensed with the explicit vector notation.

In order to make further progress we would like, by analogy with the static case treated in Refs. 5 and 6, to introduce eigenstates of Γ and use them to expand the solution of the scattering problem. Although the scalar product of two fields $\vec{E}_1$ and $\vec{E}_2$ can be defined by analogy with the static case, i.e.,

$$(E_1, E_2) \equiv \int d^3r \Theta_1(\vec{r}) \vec{E}_1^*(\vec{r}) \cdot \vec{E}_2(\vec{r}), \tag{2.9}$$

the operator Γ is not Hermitian, even though it is symmetric. Because of this, the eigenfunctions of Γ in general form a bi-orthogonal instead of a regular-orthogonal system.[9] Because of its symmetry, Γ satisfies

$$\Gamma^\dagger = \Gamma^*; \tag{2.10}$$

therefore the left eigenfunctions of Γ are just the complex conjugates of the right eigenfunctions, i.e.,

$$\begin{aligned} &\Gamma|t\rangle = t|t\rangle, \\ &\langle t^*|\Gamma = (\Gamma^*|t^*\rangle)^\dagger = (t^*|t^*\rangle)^\dagger = \langle t^*|t, \end{aligned} \tag{2.11}$$

where we have introduced a bra-ket notation. These eigenfunctions satisfy the bi-orthogonality relations

$$\langle t^*|s\rangle = 0 \quad \text{for} \quad t \neq s. \tag{2.12}$$

When there is a degeneracy of eigenvalues, we can always choose the eigenfunctions so that they are mutually bi-orthogonal. The only property which we cannot in general ensure is normalizability, i.e.,

$$\langle t^*|t\rangle \neq 0. \tag{2.13}$$

We will therefore have to assume that this property holds for all the eigenstates, enabling us to normalize them to unity. This property must be verified in every particular case. When it holds, the operator Γ can be expanded in terms of its eigenfunctions

$$\Gamma = \sum_t |t\rangle t \langle t^*|. \tag{2.14}$$

The formal solution of Eq. (2.7), namely

$$E = \frac{1}{1-u\Gamma} E_0 = E_0 + \frac{\Gamma}{1/u - \Gamma} E_0 \equiv E_0 + E_{sc}, \tag{2.15}$$

can be written in a spectral representation by expanding E_{sc} in the eigenfunctions of Γ to yield

$$\begin{aligned} &|E_{sc}\rangle = \sum_t |t\rangle \frac{t}{s-t} \langle t^*|E_0\rangle, \\ &s \equiv \frac{1}{u} = \frac{\kappa_2}{\kappa_2 - \kappa_1}. \end{aligned} \tag{2.16}$$

The eigenfunctions $|t\rangle$ all behave as outgoing waves $\sim e^{ikr}/r$ for large r, and hence this leads to a spectral representation for the scattering amplitude. In this representation, the microscopic geometry of the scatterers and the incident wave number k completely determine the eigenfunctions and the eigenvalues. The scalar product $\langle t^*|E_0\rangle$ depends also on the incident wave (e.g., for a plane wave it depends on the incident wave vector $\vec{k}_0$). The physical properties of the scattering system, i.e., the values of κ_1 and κ_2, enter only through the variable s which appears in Eq. (2.16). Thus the spectral representation clearly separates the geometrical aspects of the problem from the material or physical aspects.

As usual, with a spectral representation we can always associate a sum rule. In this case, the sum rule is obtained by letting $s \to \infty$, i.e., $\kappa_1 \to \kappa_2$, in Eq. (2.16). In this way we get

$$\lim_{s\to\infty} s|E_{sc}\rangle \equiv \sum_t |t\rangle t \langle t^*|E_0\rangle = \Gamma|E_0\rangle, \tag{2.17}$$

which is just the coefficient of u^1 in the expansion of $|E\rangle$ in powers of u. In physical terms, this is just the first Born approximation for the scattered field.

The eigenstates of Γ fall into two classes:

(1) Longitudinal eigenstates $\vec{E} = \vec{\nabla}\phi$. For these states the eigenvalue must be $u = 1$, and $\vec{E}$ must vanish outside the volume of κ_1 material [see Eq. (2.3)]. Inside the κ_1 volume $\phi(\vec{r})$ is arbitrary, and the equations are satisfied in a trivial way. The only constraint on ϕ is that it must obey a certain condition at the κ_1, κ_2 boundary: because the tangential component of $\vec{E}$ is continuous, we must have $\phi = \text{const}$ over every connected piece of that boundary. Obviously, $\vec{H}$ vanishes everywhere for these states.

(2) All the eigenstates for which $u \neq 1$ *(these are transverse fields).* From Eq. (2.3) $\vec{\nabla} \cdot \vec{E} = 0$ both inside κ_1 and inside κ_2 regions. These states must obey the equations [either (2.3) or (2.11)] in a nontrivial way.

It can be shown that only the class (2) eigenstates are needed to expand the scattering problem. If $\vec{E}$ is a solution of the scattering problem, while $\vec{E}_1$ is a $u = 1$ eigenfunction, we can write

$$\begin{aligned} \langle E_1^*|E\rangle &= \langle E_1^*|E_0\rangle + u\langle E_1^*|\Gamma|E\rangle \\ &= \langle E_1^*|E_0\rangle + u\langle E_1^*|E\rangle, \end{aligned} \tag{2.18}$$

where we used Eqs. (2.7) and (2.11). The scalar product of $|E_1^*\rangle$ and the incident field $|E_0\rangle$ can be shown to vanish as follows:

$$\begin{aligned} \langle E_1^*|E_0\rangle &= \int dV \Theta_1 \vec{E}_1 \cdot \vec{E}_0 = \int_{V_1} dV \vec{\nabla}\phi \cdot \vec{E}_0 \\ &= \int_{V_1} dV [\vec{\nabla} \cdot (\phi \vec{E}_0) - \phi \vec{\nabla} \cdot \vec{E}_0]. \end{aligned} \tag{2.19}$$

The second term under the last integral vanishes because the incident field is a transverse field. The first term can be transformed to a surface integral (over the κ_1-κ_2 boundary), where $\phi = \phi_0 \equiv$ const, and then back again to a volume integral, leaving ϕ_0 outside the integral. Thus we get

$$\langle E_1^* | E_0 \rangle = \phi_0 \int_{V_1} dV \, \vec{\nabla} \cdot \vec{E}_0 = 0 . \tag{2.20}$$

We have thus shown that

$$\langle E_1^* | E \rangle = u \langle E_1^* | E \rangle , \tag{2.21}$$

and hence $\langle E_1^* | E \rangle = 0$ for $u \neq 1$.

The physical significance of the class (2) eigenstates is that at special values of κ_1/κ_2 (the eigenvalues), an outgoing wave can arise in the system spuriously, i. e., without the presence of an incident wave. If we assume that κ_2 is real and positive, which means that k is also real, such an eigenstate represents a constant net energy flow out to infinity due to the outgoing wave character of the field at large distances. Since there is no gain or loss of energy in the host medium for purely real κ_2, this requires that there be a source of energy inside the κ_1 scatterer. This means that we must have $\mathrm{Im}\kappa_1 < 0$ for any eigenstate, i. e., $\mathrm{Im}u > 0$ for any eigenvalue u. This is, of course, unphysical, which means that we can never actually encounter a resonance in a real system. However, resonances for which $|\mathrm{Im}u|$ is very small can sometimes be approached very closely. This is what occurs in a Fabry-Perot interferometer when an incident wave gets "trapped" and undergoes many internal reflections before finally getting out again.

In order for the spectral representation to be useful, we must find a convenient scheme for calculating the eigenstates $|t\rangle$. This is achieved by noting that, as in the static case,[5,6] Γ can be written as a sum of individual grain operators, following the analogous property of $\Theta_1(\vec{r})$:

$$\begin{aligned} &\Theta_1(\vec{r}) = \sum_a \Theta_a(\vec{r}) , \\ &\Gamma = \sum_a \Gamma_a , \end{aligned} \tag{2.22}$$

where the sums are over distinct, nontouching grains a. The scheme consists of first finding the eigenstates $\vec{E}_{a\alpha}(\vec{r})$ of every isolated grain, and then using these states to expand the solution of the entire problem.

$$\begin{aligned} &\Gamma_a E_{a\alpha} = s_{a\alpha} E_{a\alpha} , \\ &\Theta_1^\dagger(\vec{r}) \vec{E}(\vec{r}) = \sum_{a\alpha} A_{a\alpha} \Theta_a^\dagger(\vec{r}) \vec{E}_{a\alpha}(\vec{r}) . \end{aligned} \tag{2.23}$$

The functions $\Theta_1^\dagger, \Theta_a^\dagger$ are nearly the same as Θ_1, Θ_a. They differ from them in that they are equal to 1 over a volume that is infinitesimally larger than that of Θ_1, Θ_a. This is required in order to avoid spurious singularities when calculating scalar products. Note that the expansion represents $\vec{E}(\vec{r})$ only inside the κ_1 material, and that in each grain only the eigenfunctions arising from that grain are used.

In this scheme, the operator Γ is represented by a matrix between individual grain eigenstates, which can be represented as follows in the form of an overlap integral:

$$\begin{aligned} \langle \Theta_a^\dagger E_{a\alpha}^* | \Gamma | \Theta_b^\dagger E_{b\beta} \rangle &= s_{b\beta} \langle \Theta_a^\dagger E_{a\alpha}^* | E_{b\beta} \rangle \\ &= s_{b\beta} \int d^3r \, \Theta_a (\vec{E}_{a\alpha} \cdot \vec{E}_{b\beta}) . \end{aligned} \tag{2.24}$$

Note that the integration volume includes only the grain a. The problem of finding the eigenstates of Γ thus becomes a problem of finding the eigenvectors of this non-Hermitian matrix. Once the eigenvalues t and eigenvectors $\langle \Theta_a^\dagger E_{a\alpha}^* | t \rangle$ are known, we can expand the scattered field $\vec{E}$ in a series of individual grain eigenfunctions $\vec{E}_{a\alpha}$.

Some care must be exercised when doing this because the usual decomposition of the unit operator I, based on completeness,

$$I = \sum_\alpha | E_{a\alpha} \rangle \langle E_{a\alpha}^* | , \tag{2.25}$$

is only valid inside the grain a. Even if we sum this expression over all grains a, we still get a decomposition of the unit operator that is valid only inside κ_1 material. However, we would like to expand $\vec{E}_{sc}$ also (or even mainly) outside the scattering grains. In order to achieve this, we note that in order for Γ to operate on one of its eigenfunctions $\vec{E}_t$, only values of $\vec{E}_t$ inside the κ_1 material are needed. As a result of this operation, however, we obtain the values of $\vec{E}_t$ everywhere. Therefore, we can write

$$\begin{aligned} | E_t \rangle = \frac{1}{t} \Gamma | \Theta_1^\dagger E_t \rangle &= \sum_{a\alpha} \frac{1}{t} \Gamma | \Theta_a^\dagger E_{a\alpha} \rangle \langle \Theta_a^\dagger E_{a\alpha}^* | \Theta_1^\dagger E_t \rangle \\ &= \sum_{a\alpha} \frac{s_{a\alpha}}{t} | E_{a\alpha} \rangle \langle \Theta_a^\dagger E_{a\alpha}^* | E_t \rangle . \end{aligned} \tag{2.26}$$

Here we have used Eqs. (2.22) and (2.23) to express the result of Γ operating on an isolated grain eigenfunction $\Theta_a^\dagger \vec{E}_{a\alpha}$. The result of Eq. (2.26) is an expansion of the eigenfunction $\vec{E}_t$ which is valid also outside the κ_1 material. (In fact, inside the κ_1 material it is not a very useful expansion because even inside a particular grain a eigenfunctions from other grains also contribute.)

Using these results, we can now rewrite Eq.

(2.16) as a sum over the individual grain eigenfunctions $\vec{E}_{a\alpha}$ with coefficients that are given in terms of the eigenvectors $\langle\Theta_a^\dagger E_{a\alpha}^*|t\rangle$ and both the perturbed and unperturbed eigenvalues t and $s_{a\alpha}$:

$$|E_{sc}\rangle = \sum_{a\alpha,b\beta,t} s_{a\alpha}|E_{a\alpha}\rangle\langle\Theta_a^\dagger E_{a\alpha}^*|t\rangle \frac{1}{s-t} \times\langle t^*|\Theta_b^\dagger E_{b\beta}\rangle\langle\Theta_b^\dagger E_{b\beta}^*|E_0\rangle . \quad (2.27)$$

The sum rule of Eq. (2.17) becomes, in this scheme

$$\sum_{a\alpha,b\beta,t} s_{a\alpha}|E_{a\alpha}\rangle\langle\Theta_a^\dagger E_{a\alpha}^*|t\rangle\langle t^*|\Theta_b^\dagger E_{b\beta}\rangle\langle\Theta_b^\dagger E_{b\beta}^*|E_0\rangle = \sum_{a\alpha}|E_{a\alpha}\rangle\langle\Theta_a^\dagger E_{a\alpha}^*|E_0\rangle . \quad (2.28)$$

A somewhat more useful sum rule can be obtained by deleting the summation over $a\alpha$ in the last equation. Even though the states $\vec{E}_{a\alpha}$ appearing in the expansion are not independent, since each state has not been restricted to its own grain, we can nevertheless equate the coefficients on both sides of Eq. (2.28): This follows from the fact that both sets of states $\{t\}$ and $\{\Theta_a^\dagger\vec{E}_{a\alpha}\}$ are independent and complete over the volume of κ_1 material. We thus get the following sum rule for the residues of the partial scattering amplitudes of Eq. (2.27):

$$\sum_{b\beta t}\langle\Theta_a^\dagger E_{a\alpha}^*|t\rangle\langle t^*|\Theta_b^\dagger E_{b\beta}\rangle\langle\Theta_b^\dagger E_{b\beta}^*|E_0\rangle = \langle\Theta_a^\dagger E_{a\alpha}^*|E_0\rangle . \quad (2.29)$$

III. SCATTERING BY A COLLECTION OF SPHERES

The first step is to find the eigenstates of a single, isolated sphere. This is most easily done by returning to the formulation of the problem as a partial differential equation, as in Eq. (2.3), together with the boundary condition of outgoing waves at large distances (there is no incident wave in the eigenvalue problem). As we found in the general case, the eigenstates of a single sphere fall into two classes, and we will only need the transverse eigenstates, for which $u \neq 1$. Because of the spherical and inversion symmetry, the eigenstates of Γ are also eigenstates of the angular momentum operators J_z and J^2 and of parity P, and they can be constructed from the usual transverse electron (TE) and transverse magnetic (TM) vector spherical harmonics (VSH). This is done in detail in Appendix A. Because J_z, J^2, and P are Hermitian, therefore eigenstates differing in their TE or TM character, or in their angular momentum quantum numbers l, m are orthogonal in the usual sense. Bi-orthogonality must only be invoked for eigenstates that differ only in the radial quantum number n. Therefore the complex conjugation that must be applied to a right eigenfunction of Γ in order to change it into a right eigenfunction of $\Gamma^\dagger = \Gamma^*$ is only applied to the radial functions. This operation is denoted by C; thus

$$\Gamma|E_{lmn}^{(F)}\rangle = s_{ln}^{(F)}|E_{lmn}^{(F)}\rangle , \qquad \Gamma^\dagger|CE_{lmn}^{(F)}\rangle = s_{ln}^{(F)*}|CE_{lmn}^{(F)}\rangle , \quad (3.1)$$

where F stands for either E (a TM state) or M (a TE state).

For a single sphere of radius a, the scattered field is calculated in Appendix A in the long wavelength limit $ka \ll 1$, in the standard form of a spectral representation as a function of s [see Eq. (A26)]. The solution is obtained in a form that is applicable for any values of κ_1 and κ_2—one merely has to substitute the appropriate value for $s = \kappa_2/(\kappa_2 - \kappa_1)$. For example, if the sphere is a perfect conductor then $s = 0$. With the help of the sum rule[10] on the zeros x_{ln} of the spherical Bessel functions $j_l(x)$,

$$\sum_{n=1}^{\infty}\frac{1}{x_{ln}^2} = \frac{1}{2(2l+3)} , \quad (3.2)$$

we recover the well-known results for this problem.[7]

In order to discuss the scattering by more than one sphere, we need the matrix elements of Γ between eigenstates of different spheres with radii a, a', centered at $\vec{b}, \vec{b}'$,

$$\langle\Theta_b^\dagger CE_{blmn}^{(F)}|\Gamma|\Theta_{b'}^\dagger E_{b'l'm'n'}^{(F')}\rangle = s_{b'l'n'}^{(F')}\langle\Theta_b^\dagger CE_{blmn}^{(F)}|E_{b'l'm'n'}^{(F')}\rangle = s_{b'l'n'}^{(F')}\int_{|\vec{r}-\vec{b}|<a} d^3r\,[C\vec{E}_{lmn}^{(F)}(\vec{r}-\vec{b})\cdot\vec{E}_{l'm'n'}^{(F')}(\vec{r}-\vec{b}')] . \quad (3.3)$$

These matrix elements are evaluated in Appendix B, and in particular detail for the long-wavelength limit $ka \ll 1$, $ka' \ll 1$, $k|\vec{b}'-\vec{b}| \ll 1$. The right eigenvalues t and the right eigenvectors $\langle\Theta_b^\dagger CE_{blmn}^{(F)}|t\rangle$ of this matrix must be found. Using them, we can write the spectral representation for the scattered field,

$$|E_{sc}\rangle = \sum_{Fblmn F'b'l'm'n',t} s_{bln}^{(F)}|E_{blmn}^{(F)}\rangle\langle\Theta_b^\dagger CE_{blmn}^{(F)}|t\rangle \frac{1}{s-t} \times\langle Ct|\Theta_{b'}^\dagger E_{b'l'm'n'}^{(F')}\rangle \times\langle\Theta_{b'}^\dagger CE_{b'l'm'n'}^{(F')}|E_0\rangle . \quad (3.4)$$

As is shown in Appendix B, in the long-wavelength limit we only need to consider the approximately separate subspaces of $E^{(M)}$ states (with eigenvalues denoted by $t^{(M)}$) and of $E^{(E)}$ states with $n=0$ (with eigenvalues $t^{(E)}$). In that case the relative components of the eigenvectors are of order $O(k^0)$. Consequently, the order of magnitude of the residue of any pole in Eq. (3.4) is determined by the $Fl=F'l'=M1$ or $Fln=F'l'n'=E10$ terms in the sum. This order of magnitude is determined by the product

$$s^{(F)}_{bln}\,|\,E^{(F)}_{blmn}\rangle\langle\Theta^{+}_{b'}CE^{(F)}_{b'lm'n'}\,|\,E_0\rangle \sim \frac{e^{ikr}}{kr}\times\begin{cases}(ka)^3, & F_n=E_0\\ (ka)^5, & F=M\end{cases}. \quad (3.5)$$

One should not conclude from this result that the TE states are unimportant compared to the TM states, since although the residues are smaller, the eigenvalues $t^{(M)}$ are also smaller than the $t^{(E)}$ eigenvalues by the same type of $O(k^2a^2)$ factor.

As an example, we apply the procedure developed above to the problem of the scattering by a pair of identical spheres of radius a, whose centers are at $\vec{b}$ and $\vec{b}'$. In this case, the symmetry of the scatterer leads us to choose a coordinate system in such a way that the z axis lies along $\vec{b}'-\vec{b}$. The incident wave vector $\vec{k}_0$ is chosen to lie in the x-z plane, and is given in spherical coordinates by $(k,\Theta_0,0)$. From this choice it follows that only states with $m=m'$ can lead to nonvanishing matrix elements of Γ [see Eqs. (B5)–(B8)]. We also assume that $|\vec{b}'-\vec{b}|\gg a$, in which case the off-diagonal matrix elements are usually much smaller than the differences between the unperturbed isolated sphere eigenvalues. We therefore restrict ourselves to considering only the electric and magnetic dipole states of each sphere, and we ignore all interactions except when they couple a pair of states with the same unperturbed eigenvalues. We are thus left with the problem of diagonalizing a large number of 2×2 matrices of the form

$$(t^{(F)}_{mn})_{bb'}\equiv\langle\Theta^{+}_{b}CE^{(F)}_{blmn}\,|\,\Gamma\,|\,\Theta^{+}_{b'}E^{(F)}_{b'lmn}\rangle, \quad (3.6)$$

with the matrix elements given by Eqs. (B11) and (B12). The matrices $(t^{(F)}_{mn})_{bb'}$ have the form

$$t^{(F)}_{mn}=\begin{pmatrix}t_0 & t_1\\ t_1 & t_0\end{pmatrix}, \quad (3.7)$$

and therefore the eigenvalues and eigenvectors are

$$t_{\pm}=t_0\pm t_1,\qquad |t_{\pm}\rangle=\begin{pmatrix}\frac{1}{\sqrt{2}}\\ \pm\frac{1}{\sqrt{2}}\end{pmatrix}. \quad (3.8)$$

The quantities $(t^{(F)}_{mn})_0$ and $(t^{(F)}_{mn})_1$ are given by

$$(t^{(M)}_{mn})_0=-\left(\frac{ka}{x_{0n}}\right)^2,$$

$$(t^{(M)}_{mn})_1=\frac{1}{3\sqrt{5}}\frac{(ka)^2}{x^4_{0n}}\left(\frac{a}{|\vec{b}'-\vec{b}|}\right)^3\begin{cases}(-2) & \text{for } m=0\\ 1 & \text{for } m=\pm1\end{cases},$$

$$(t^{(E)}_{m0})_0=\tfrac{1}{3}, \quad (3.9)$$

$$(t^{(E)}_{m0})_1=\frac{1}{45}\left(\frac{a}{|\vec{b}'-\vec{b}|}\right)^3\begin{cases}(-2) & \text{for } m=0\\ 1 & \text{for } m=\pm1\end{cases},$$

$$x_{0n}=n\pi.$$

Since these quantities are all real, the matrix of Eq. (3.7) is Hermitian, and the right eigenvectors of Eq. (3.8) are also the left eigenvectors:

$$|Ct_{\pm}\rangle=|t_{\pm}\rangle. \quad (3.10)$$

When these results, as well as the scalar products $\langle CE^{(F)}_{lmn}\,|\,R(\Theta_0)E_0\rangle$ from Eq. (A25), are substituted into the spectral expansion, we obtain the following result for the scattered field in the asymptotic region $kr\gg1$.

$$\vec{E}_{sc}(\vec{r})\cong\frac{e^{ikr}}{kr}\sum_m[(\vec{n}\times\vec{X}_{1m})T^{(E)}_{1m}-i\vec{X}_{1m}T^{(M)}_{1m}], \quad (3.11)$$

where the partial-wave-scattering amplitudes $T^{(F)}_{lm}$ that appear are given by

$$T^{(E)}_{1m}\cong\mp4E_0i(12\pi)^{1/2}\frac{(ka)^3}{9}\frac{d^{(1)}_{\pm1m}(\Theta_0)}{s-\frac{1}{3}-(t^{(E)}_{m0})_1},$$

$$T^{(M)}_{1m}\cong-4E_0i(12\pi)^{1/2}(ka)^5d^{(1)}_{\pm1m}(\Theta_0)\times\sum_{n=1}^{\infty}\frac{1}{x^2_{0n}}\frac{1}{sx^2_{0n}+(ka)^2-x^2_{0n}(t^{(M)}_{mn})_1}. \quad (3.12)$$

The quantities $d^{(1)}_{\pm1m}(\Theta_0)$ in these equations are Wigner's rotation matrices,[11] which appear because the incident wave vector does not lie along the z axis (see Appendix A). By setting $\Theta_0=0$ in these results [note that $d^{(1)}_{m'm}(0)=\delta_{m'm}$] and comparing with Eq. (A26), it is easy to see that the scattering amplitude reduces, as it should, to the sum of the scattering amplitudes of two isolated spheres in the limit $|\vec{b}'-\vec{b}|\gg a$. To leading order in $a/|\vec{b}'-\vec{b}|$, the only change brought about by including the interactions is a small shift in each of the isolated sphere eigenvalues, with the shift depending also on the azimuthal quantum number m. Note that only one of the perturbed eigenvalues from each pair $t_{\pm}$ contributes to the spectral sum with a nonzero weight, namely

$$(t^{(F)}_{mn})_{+}=(t^{(F)}_{mn})_0+(t^{(F)}_{mn})_1. \quad (3.13)$$

This is in agreement with a similar result found previously in the static limit.[5] Note also that a discussion of electrostatic resonances in the two-sphere system has recently been given that has

certain points of similarity with the present discussion.[12] However, M states do not appear in that discussion, which is confined to the strict static limit.

IV. DISCUSSION AND CONCLUSIONS

We have presented a new approach to the exact calculation of electromagnetic scattering by macroscopic scatterers including the multiple scattering. The approach is based on a calculation of a complete set of scattering resonances, i.e., outgoing fields which can arise spuriously without the presence of an incident field at special values of κ_1/κ_2. When this approach is applied to an isolated sphere, the known results are rederived, albeit more tediously than by the usual phase-shift analysis.[7] In particular, the expression for the scattered field includes summation, not only over the angular momentum quantum numbers l and m, but over an infinite set of radial quantum numbers n as well. The practical advantages of our approach appear only when more complicated problems are considered: The phase-shift analysis of the scattering by even two spheres is already very complicated, owing to the loss of spherical symmetry. In our approach, by contrast, all that is needed is to include the two-sphere interaction terms in the matrix of Γ, which must then be diagonalized. Thus, the scattering by a small number of spheres is not a much more difficult problem than the scattering by a single sphere.

We should mention that the scattering resonances of a single sphere were investigated by Debye in his thesis, and are described in Ref. 8 (p. 154). Nevertheless, apparently no use was made of these resonances to discuss the scattering by more than one sphere. Also, there was no general theory of such resonances for an arbitrarily shaped scattering system.

It would seem that our approach should be useful for discussing the scattering by dense systems of strong scatterers (i.e., κ_1/κ_2 very different from 1) where multiple scattering is important. Such systems are metal-insulator granular composites of various types (e.g., cermets or metal smokes). In these systems the scattering is not confined to a small region of space, but is present everywhere. The problem is then to calculate the propagation of a wave (i.e., its dispersion law and its absorption) through the composite scattering medium. It would be interesting to apply our approach to this problem.

ACKNOWLEDGMENTS

We would like to acknowledge useful conversations with W. Lamb. This research was supported in part by the United States-Israel Binational Science Foundation under Grant No. 2006/79 and in part by NSF Grant DMR78-11770.

APPENDIX A: THE SCATTERING PROBLEM OF AN ISOLATED SPHERE

The basic transverse vector spherical harmonic (VSH) is defined by

$$\vec{X}_{lm} \equiv \frac{\vec{L}Y_{lm}}{[l(l+1)]^{1/2}} . \tag{A1}$$

The eigenstates of an isolated sphere of radius a are constructed from the two basic types of VSH—the TE (or electric multipole) field and the TM (or magnetic multipole) field as

$$\begin{aligned} &\text{TE: } \vec{E}^{(M)}_{lmn}(\vec{r}) = f^{(M)}_{ln}(r)\vec{X}_{lm}(\Omega) , \\ &\text{TM: } \vec{E}^{(E)}_{lmn}(\vec{r}) = \frac{i}{k[1-u^{(E)}_{ln}\Theta_1(r)]}[\vec{\nabla}\times f^{(E)}_{ln}(r)\vec{X}_{lm}(\Omega)] . \end{aligned} \tag{A2}$$

The radial functions have the following form for both cases $F=M$ and $F=E$:

$$f^{(F)}_{ln}(r) = \begin{cases} A^{(F)}_{ln} j_l(k(1-u^{(F)}_{ln})^{1/2} r) & \text{for } r<a \\ B^{(F)}_{ln} h^{(1)}_l(kr) & \text{for } r>a \end{cases} , \tag{A3}$$

where j_l and $h^{(1)}_l$ are the spherical Bessel functions.[7] The eigenvalues (F) as well as the coefficients A and B are determined by continuity conditions at $r=a$. Note that, beginning with Eq. (A2), and throughout Appendix A, $\Theta_1(r)$ is nonzero only within a sphere of radius a centered at the origin.

The tangential component of $\vec{E}$ as well as that of $\vec{H}$ must be continuous at $r=a$ (the latter requirement follows from the assumption that $\mu=1$ everywhere). It follows that the following radial functions must be continuous there:

$$\begin{aligned} &f^{(M)}_{ln}, \quad \frac{df^{(M)}_{ln}}{dr} \\ &f^{(E)}_{ln}, \quad \frac{1}{1-u^{(E)}_{ln}\Theta_1}\frac{d(rf^{(E)}_{ln})}{dr} . \end{aligned} \tag{A4}$$

We thus get the following equations for determining the eigenvalues:

$$x^{(F)}_{ln} \equiv k(1-u^{(F)}_{ln})^{1/2} a , \tag{A5}$$

$$\left.\frac{xj_l'(x)}{j_l(x)}\right|_{x=x^{(M)}_{ln}} = \left.\frac{xh^{(1)\prime}_l(x)}{h^{(1)}_l(x)}\right|_{x=ka} , \tag{A6}$$

$$\left.\frac{1}{x^2}\left(1+\frac{xj_l'(x)}{j_l(x)}\right)\right|_{x=x^{(E)}_{ln}} = \left.\frac{1}{x^2}\left(1+\frac{xh^{(1)\prime}_l(x)}{h^{(1)}_l(x)}\right)\right|_{x=ka} . \tag{A7}$$

For given l and F, we get an infinite sequence of solutions $x^{(F)}_{ln}$ which are enumerated by the index (or "radial quantum number") n. The eigenvalues $x^{(F)}_{ln}$ or $u^{(F)}_{ln}$ are, in general, complex owing to the nature of the rhs of Eqs. (A6) and (A7), reflecting the non-Hermitian character of Γ. They are also

evidently independent of m, and this degeneracy is due to the fact that the total angular momentum operator of the em field $\vec{J}$ commutes with Γ. Since $\vec{J}$ is a regular Hermitian operator its eigenstates, namely the VSH, satisfy the usual orthogonality relations when integrated over the angular variables. It is only the states with the same angular momentum and parity but different radial quantum number n, i.e., the radial (spherical Bessel) functions, that satisfy instead the bi-orthogonality relations. Therefore instead of taking the complex conjugate of the entire isolated sphere right eigenfunction, we only have to conjugate the radial part in order to get a left eigenfunction. This operation will be denoted by a capital C. Thus, instead of $|t^*\rangle$ we have

$$C\vec{E}^{(M)}_{lmn} \equiv f^{(M)*}_{ln}(r)\vec{X}_{lm}(\Omega)\,,$$
$$C\vec{E}^{(E)}_{lmn} \equiv \left(\frac{i}{k(1-u^{(E)}_{ln}\Theta_1)}\right)^{*}[\vec{\nabla}\times f^{(E)*}_{ln}(r)\vec{X}_{lm}(\Omega)]\,. \tag{A8}$$

The coefficients $A^{(F)}_{ln}$ are determined from the normalization condition on $E^{(F)}_{lmn}$:

$$1=\langle CE^{(M)}_{lmn}\,|\,E^{(M)}_{lmn}\rangle = \int_{r<a} d^3r[A^{(M)}_{ln}j_l(k(1-u^M_{ln})^{1/2}r)]^2\vec{X}^*_{lm}\cdot\vec{X}_{lm} = \frac{(A^{(M)}_{ln})^2a^3}{(x^{(M)}_{ln})^3}\int_0^{x^{(M)}_{ln}} dx[xj_l(x)]^2\,, \tag{A9}$$

$$1=\langle CE^{(E)}_{lmn}\,|\,E^{(E)}_{lmn}\rangle = \int_{r<a} d^3r\left(\frac{iA^{(E)}_{ln}}{k(1-u^{(E)}_{ln})}\right)^2[\vec{\nabla}\times j_l(k(1-u^{(E)}_{ln})^{1/2}r)\vec{X}^*_{lm}]\cdot[\vec{\nabla}\times j_l(k(1-u^{(E)}_{ln})^{1/2}r)\vec{X}_{lm}]$$
$$=-(A^{(E)}_{ln})^2a^3\left[\frac{(ka)^2}{(x^{(E)}_{ln})^5}\int_0^{x^{(E)}_{ln}} dx[xj_l(x)]^2+\left(\frac{j_l(x^{(E)}_{ln})}{x^{(E)}_{ln}}\right)^2\left(1+ka\frac{h^{(1)\prime}_l(ka)}{h^{(1)}_l(ka)}\right)\right]. \tag{A10}$$

In order to get the last equation we had to use the integral formula

$$\int_{r<a} d^3r[\vec{\nabla}\times f_l(\alpha r)\vec{X}^*_{lm}]\cdot[\vec{\nabla}\times g_{l'}(\beta r)\vec{X}_{l'm'}] = \frac{\delta_{ll'}\delta_{mm'}}{\beta}\left\{\int_0^{\beta a} dx\,x^2 f_l\left(\frac{\alpha}{\beta}x\right)g_l(x)+\left[xf_l(\alpha a)g_l(x)\left(1+\frac{xg'_l(x)}{g_l(x)}\right)\right]_0^{\beta a}\right\}, \tag{A11}$$

which is valid when $g_{l'}(x)$ is any spherical Bessel function, as well as Eq. (A7). The other coefficients $B^{(F)}_{ln}$ are determined from $A^{(F)}_{ln}$ by using the continuity conditions

$$B^{(F)}_{ln}=A^{(F)}_{ln}\frac{j_l(x^{(F)}_{ln})}{h^{(1)}_l(ka)}\,. \tag{A12}$$

An important special case that is both instructive and useful occurs when $ka\ll 1$. In that limit, Eqs. (A6) and (A7) become

$$\left.\frac{xj_{l-1}(x)}{j_l(x)}\right|_{x=x^{(M)}_{ln}} = O(k^2a^2)\,, \tag{A13}$$

$$\left.\left(\frac{j_{l-1}(x)}{xj_l(x)}-\frac{l}{x^2}\right)\right|_{x=x^{(E)}_{ln}} = -\frac{l}{k^2a^2}+\frac{1}{2l-1}+O(k^2a^2)\,, \tag{A14}$$

respectively, where we have used the relation

$$f'_l(x)=-\frac{l+1}{x}f_l(x)+f_{l-1}(x)\,, \tag{A15}$$

valid for any spherical Bessel function, and the asymptotic form of $h^{(1)}_l(x)$ for small x

$$h^{(1)}_l(x)\cong -i\frac{(2l-1)!!}{x^{l+1}}\,. \tag{A16}$$

Equation (A14) has one solution with a very small $x^{(E)}_{ln}=O(ka)$, to which we assign the radial quantum number $n=0$:

$$(x^{(E)}_{l0})^2=-k^2a^2\frac{l+1}{l}+O(k^4a^4)\,,$$
$$u^{(E)}_{l0}\cong\frac{2l+1}{l}\,. \tag{A17}$$

Comparing with Refs. 5 and 6, we see that these are the eigenstates that reduce to the electrostatic multipole resonances in the limit $k=0$. All other solutions of Eqs. (A13) and (A14) are $O(1)$, and they are given, to lowest order, by the zeros x_{ln}, $n=1,2,\ldots$, of the spherical Bessel functions $j_l(x)$:

$$x^{(M)}_{ln}=x_{l-1n}+O(k^2a^2)\,,\quad u^{(M)}_{ln}\cong-\left(\frac{x_{l-1n}}{ka}\right)^2\,,\quad n\geq 1$$
$$x^{(E)}_{ln}=x_{ln}+O(k^2a^2)\,,\quad u^{(E)}_{ln}\cong-\left(\frac{x_{ln}}{ka}\right)^2\,,\quad n\geq 1\,. \tag{A18}$$

In the strict electrostatic limit, $u^{(E)}_{ln}$ (as well as $u^{(M)}_{ln}$), $n\geq 1$ are all infinite, and such states are then not needed when expanding the static electric field, as was shown in Ref. 6. As we will show later, a similar conclusion holds approximately for the $E^{(E)}$ states in the limit $ka\ll 1$. We now proceed to calculate all the necessary ingredients for solving the scattering problem of a single sphere in that limit explicitly in terms of the zeros x_{ln} of the functions $j_l(x)$.

By using the result

$$\int_0^a dr\, r^2 j_l(\alpha r) j_l(\beta r) = \frac{a^2}{\alpha^2-\beta^2}[\beta j_l(\alpha a) j_{l-1}(\beta a) - \alpha j_{l-1}(\alpha a) j_l(\beta a)], \tag{A19}$$

it is straightforward to calculate the normalization integrals in Eqs. (A9) and (A10). Using the asymptotic form of $j_l(x)$ for small x

$$j_l(x) \cong \frac{x^l}{(2l+1)!!}, \tag{A20}$$

we thus find

$$A_{ln}^{(M)} \cong \left(\frac{2}{a^3}\right)^{1/2} \frac{1}{j_l(x_{l-1n})}, \quad B_{ln}^{(M)} \cong i\left(\frac{2}{a^3}\right)^{1/2} \frac{(ka)^{l+1}}{(2l-1)!!},$$

$$A_{ln}^{(E)} \cong i\left(\frac{2}{a^3}\right)^{1/2} \frac{x_{ln}}{kaj_{l-1}(x_{ln})}, \quad B_{ln}^{(E)} \cong \left(\frac{2}{a^3}\right)^{1/2} \frac{(ka)^{l+2}}{l(2l-1)!!}, \quad n \geq 1 \tag{A21}$$

$$A_{l0}^{(E)} \cong \left(\frac{1}{la^3}\right)^{1/2} \frac{(2l+1)!!}{(ika)^{l-1}} \left(\frac{l}{l+1}\right)^{(l-1)/2}, \quad B_{l0}^{(E)} \cong -\left(\frac{l+1}{a^3}\right)^{1/2} \frac{(ka)^{l+2}}{l(2l-1)!!}.$$

The remaining quantities which must be calculated are the scalar products $\langle CE_{lmn}^{(F)} | E_0 \rangle$. We will take $\vec{E}_0$ to be a circularly polarized plane wave with a wave vector $\vec{k}_0$ whose spherical components are $(k, \theta_0, 0)$, i.e., it lies in the x-z plane and makes an angle θ_0 with the z axis. The reason for not taking the z axis along $\vec{k}_0$ is that in cases where the scattering system has an intrinsic axis of symmetry, it is usually better to choose that as the z axis. Such a case occurs in the scattering problem of two spheres, which is discussed in Sec. III.

In order to expand such a field in VSH, we start from the expansion of a circularly polarized plane wave traveling in the z direction[7]

$$\vec{E}_0(\vec{r}) \equiv E_0 \vec{e}_\pm e^{ikz} = E_0 \sum_{l=1}^{\infty} i^{l-1}[4\pi(2l+1)]^{1/2} \left(ij_l(kr)\vec{X}_{l\pm1} \pm \frac{i}{k}[\vec{\nabla} \times j_l(kr)\vec{X}_{l\pm1}] \right),$$
$$\vec{e}_\pm \equiv \vec{e}_x \pm i\vec{e}_y. \tag{A22}$$

We then rotate the coordinate axes by the angle $(-\theta_0)$ around the y axis, and use the fact that the E-type and M-type VSH with a given l are the components of an irreducible spherical tensor in order to represent that rotation by Wigner's $d_{mm'}^{(l)}(\theta_0)$ matrices.[11] The field $\vec{E}_0(\vec{r})$ as a function of the rotated coordinate axes will be denoted by $R(\theta_0)\vec{E}_0(\vec{r})$, i.e., we use the same symbols $\vec{r} = (r, \Omega)$ to denote the new coordinate of a given field point. In the rotated coordinate system, this field, which now has a wave vector $\vec{k}_0$ pointing in the right direction, has the following expansion:

$$R(\theta_0)\vec{E}_0 = E_0 \sum_{l=1}^{\infty} \sum_{m=-l}^{l} i^{l-1}[4\pi(2l+1)]^{1/2} d_{\pm1m}^{(l)}(\theta_0) \{ ij_l(kr)\vec{X}_{lm}(\Omega) \pm (i/k)[\vec{\nabla} \times j_l(kr)\vec{X}_{lm}(\Omega)] \}. \tag{A23}$$

This result reduces to Eq. (A22) when $\theta_0 = 0$ because the rotation matrices satisfy

$$d_{mm'}^{(l)}(0) = \delta_{mm'}. \tag{A24}$$

The required scalar products are now easily calculated by using the orthogonality properties of the VSH as well as Eqs. (A11) and (A19). For the present case, i.e., $ka \ll 1$, we thus get

$$\langle CE_{lmn}^{(M)} | R(\theta_0)E_0 \rangle = d_{\pm1m}^{(l)}(\theta_0) E_0 \frac{i^l[8\pi(2l+1)a^3]^{1/2}}{(2l-1)!!} \frac{(ka)^l}{x_{l-1n}^2},$$

$$\langle CE_{lmn}^{(E)} | R(\theta_0)E_0 \rangle = \pm d_{\pm1m}^{(l)}(\theta_0) E_0 \frac{i^l[8\pi(2l+1)a^3]^{1/2}}{l(2l-1)!!} \frac{(ka)^{l+1}}{x_{ln}^2}, \quad n \geq 1 \tag{A25}$$

$$\langle CE_{lm0}^{(E)} | R(\theta_0)E_0 \rangle = \pm d_{\pm1m}^{(l)}(\theta_0) E_0 \frac{i^l[4\pi(2l+1)a^3)^{1/2}}{(2l+1)!!} (l+1)^{1/2}(ka)^{l-1}.$$

In order to calculate the field scattered by a single sphere of radius a and arbitrary κ_1 in the long-wavelength limit $ka \ll 1$, we choose $\Theta_0 = 0$. The scattered field is thus given by

$$\vec{E}_{sc} \cong E_0 \sum_{l=1}^{\infty} i^l [4\pi(2l+1)]^{1/2} \Bigg[i h_l^{(1)}(kr)\vec{X}_{l\pm1} \frac{-2(ka)^{2l+1}}{[(2l-1)!!]^2} \sum_{n=1}^{\infty} \frac{(ka/x_{l-1n})^2}{(ka)^2 + s x_{ln}^2}$$

$$\pm \frac{i}{k}[\vec{\nabla} \times h_l^{(1)}(kr)\vec{X}_{l\pm1}]$$

$$\times \left(\frac{-(l+1)(ka)^{2l+1}}{[(2l+1)!!]^2} \frac{1}{s - l/(2l+1)} + \frac{-2(ka)^{2l+3}}{l^2[(2l-1)!!]^2} \sum_{n=1}^{\infty} \frac{(ka/x_{ln})^2}{(ka)^2 + s x_{ln}^2} \right) \Bigg] . \tag{A26}$$

It is immediately clear from this equation that the electric partial wave $(\vec{\nabla} \times h_l^{(1)}\vec{X}_{l\pm1})$ of the scattered field arises, to lowest order in ka, only from the $n=0$ radial eigenstate. On the other hand, in the magnetic partial wave $h_l^{(1)}\vec{X}_{l\pm1}$ all the radial eigenstates $n \geq 1$ contribute to the scattered field to lowest order in ka. The ka dependence of the isolated sphere eigenvalues and other quantities calculated in this appendix are extremely useful when one attempts to calculate the scattering by a collection of spheres in the limit where all the radii a_i satisfy $ka_i \ll 1$.

APPENDIX B: MATRIX ELEMENTS OF TWO-SPHERE INTERACTIONS

In order to calculate the matrix elements of Γ between two eigenstates belonging to different spheres, according to Eq. (2.24), we must calculate an overlap integral of the two eigenfunctions over one of the spheres. To facilitate this, we will expand the eigenstates of one sphere in VSH centered around the other sphere.

In this appendix, we use the following definition and notation for the VSH (Ref. 13):

$$\vec{Y}_{Jlm}(\Omega) \equiv \sum_{mq} Y_{lm}(\Omega)\vec{e}_q (lm\,|\,q\,|\,JM), \tag{B1}$$

where $\vec{e}_q$ are the spherical unit vectors

$$\vec{e}_1 = -\frac{1}{\sqrt{2}}(\vec{e}_x + i\vec{e}_y),$$

$$\vec{e}_0 = \vec{e}_z, \tag{B2}$$

$$\vec{e}_{-1} = -\vec{e}_1^{\,*},$$

and where $(lm\,|\,q\,|\,JM)$ is a Clebsch-Gordan vector coupling coefficient. The connection between this definition and the TE and TM VSH which are so useful in electromagnetism is

$$\vec{X}_{lm} = \vec{Y}_{ll\,m},$$

$$\frac{i}{k}[\vec{\nabla} \times f_l(kr)\vec{X}_{lm}] = \left(\frac{l}{2l+1}\right)^{1/2} f_{l+1}(kr)\vec{Y}_{l\,l+1\,m} \tag{B3}$$

$$- \left(\frac{l+1}{2l+1}\right)^{1/2} f_{l-1}(kr)\vec{Y}_{l\,l-1\,m},$$

where $f_l(x)$ is any spherical Bessel function. According to Ref. 14 the required expansion is

$$f_l(k\,|\vec{r} - \vec{b}\,|)\vec{Y}_{Jl\,M}(\Omega_{r-b}) = (4\pi)^{1/2} \sum_{l'J'M',\lambda\mu} j_{l'}(kr)\vec{Y}_{J'l'M'}(\Omega_r)$$

$$\times f_\lambda(kb) Y_{\lambda\mu}(\Omega_b) i^{l'-l+\lambda}(-1)^{1+J+J'-M} \begin{pmatrix} \lambda & l' & l \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} \lambda & J' & J \\ \mu & M' & -M \end{pmatrix}$$

$$\times \begin{Bmatrix} \lambda & J' & J \\ 1 & l & l' \end{Bmatrix} \text{ for } |\vec{b}| > |\vec{r}| \text{ and } |\vec{r} - \vec{b}| > |\vec{r}|, \tag{B4}$$

where 3-j and 6-j symbols appear as coefficients.

Using this expansion, it is a straightforward matter to evaluate the required overlap integrals between a state of the sphere at $\vec{b}$ with a radius a and a sphere at $\vec{b}'$ with a radius a':

$$\langle \Theta_b^* C E_{bl\,mn}^{(M)} \,|\, E_{b'l'm'n'}^{(M)} \rangle = A_{ln}^{(M)} B_{l'n'}^{(M)} (4\pi)^{1/2} i^{l-l'} (-1)^{1+l+l'-m'} I_l(a, a_{ln}^{(M)})$$

$$\times \sum_{\lambda\mu} i^\lambda h_\lambda^{(1)}(k\,|\vec{b}' - \vec{b}\,|) Y_{\lambda\mu}(\Omega_{b'-b}) \begin{pmatrix} \lambda & l & l' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} \lambda & l & l' \\ \mu & m & -m' \end{pmatrix} \begin{Bmatrix} \lambda & l & l' \\ 1 & l' & l \end{Bmatrix}, \tag{B5}$$

$$\langle \Theta_b^+ C E^{(E)}_{bl\,mn} \,|\, E^{(E)}_{b'l'm'n'} \rangle$$

$$= \frac{A^{(E)}_{ln} B^{(E)}_{l'n'}}{(1-u^{(E)}_{ln})^{1/2}} i^{l-l'} (-1)^{l+l'-m'} \left(\frac{4\pi}{(2l+1)(2l'+1)}\right)^{1/2}$$

$$\times \sum_{\lambda\mu} i^\lambda h^{(1)}_\lambda(k|\vec{b}'-\vec{b}|) Y_{\lambda\mu}(\Omega_{b'-b}) \begin{pmatrix} \lambda & l & l' \\ \mu & m & -m' \end{pmatrix}$$

$$\times \left(\left[(ll')^{1/2} \begin{pmatrix} \lambda & l+1 & l'+1 \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} \lambda & l & l' \\ 1 & l'+1 & l+1 \end{Bmatrix} + [l(l'+1)]^{1/2} \begin{pmatrix} \lambda & l+1 & l'-1 \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} \lambda & l & l' \\ 1 & l'-1 & l+1 \end{Bmatrix} \right] I_{l+1}(a, u^{(E)}_{ln}) \right.$$

$$\left. + \left[[(l+1)l']^{1/2} \begin{pmatrix} \lambda & l-1 & l'+1 \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} \lambda & l & l' \\ 1 & l'+1 & l-1 \end{Bmatrix} + [(l+1)(l'+1)]^{1/2} \begin{pmatrix} \lambda & l-1 & l'-1 \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} \lambda & l & l' \\ 1 & l'-1 & l-1 \end{Bmatrix} \right] \right.$$

$$\left. \times I_{l-1}(a, u^{(E)}_{ln}) \right), \tag{B6}$$

$$\langle \Theta_b^+ C E^{(M)}_{bl\,mn} \,|\, E^{(E)}_{b'l'm'n'} \rangle = A^{(M)}_{ln} B^{(E)}_{l'n'} (4\pi)^{1/2} i^{l-l'-1} (-1)^{1+l+l'-m'}$$

$$\times \sum_{\lambda\mu} i^\lambda h^{(1)}_\lambda(k|\vec{b}'-\vec{b}|) Y_{\lambda\mu}(\Omega_{b'-b}) \begin{pmatrix} \lambda & l & l' \\ \mu & m & -m' \end{pmatrix}$$

$$\times \left[\left(\frac{l'}{2l'+1}\right)^{1/2} \begin{pmatrix} \lambda & l & l'+1 \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} \lambda & l & l' \\ 1 & l'+1 & l \end{Bmatrix} + \left(\frac{l'+1}{2l'+1}\right)^{1/2} \begin{pmatrix} \lambda & l & l'-1 \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} \lambda & l & l' \\ 1 & l'-1 & l \end{Bmatrix} \right] I_l(a, u^{(M)}_{ln}), \tag{B7}$$

$$\langle \Theta_b^+ C E^{(E)}_{bl\,mn} \,|\, E^{(M)}_{b'l'm'n'} \rangle = \frac{A^{(E)}_{ln} B^{(M)}_{l'n'}}{(1-u^{(E)}_{ln})^{1/2}} (4\pi)^{1/2} i^{l-l'+1} (-1)^{1+l+l'-m'}$$

$$\times \sum_{\lambda\mu} i^\lambda h^{(1)}_\lambda(k|\vec{b}'-\vec{b}|) Y_{\lambda\mu}(\Omega_{b'-b}) \begin{pmatrix} \lambda & l & l' \\ \mu & m & -m' \end{pmatrix}$$

$$\times \left[\left(\frac{l}{2l+1}\right)^{1/2} \begin{pmatrix} \lambda & l+1 & l' \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} \lambda & l & l' \\ 1 & l' & l+1 \end{Bmatrix} I_{l+1}(a, u^{(E)}_{ln}) \right.$$

$$\left. + \left(\frac{l+1}{2l+1}\right)^{1/2} \begin{pmatrix} \lambda & l-1 & l' \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} \lambda & l & l' \\ 1 & l' & l-1 \end{Bmatrix} I_{l-1}(a, u^{(E)}_{ln}) \right]. \tag{B8}$$

The function $I_l(a, u)$ in these equations is defined by

$$I_l(a, u) \equiv \int_0^a dr\, r^2 j_l(k(1-u)^{1/2} r) j_l(kr) = -\frac{a^3}{kau} \left(j_l(x) j_{l-1}(ka) - \frac{x}{ka} j_{l-1}(x) j_l(ka) \right)_{x=k(1-u)^{1/2}a} \tag{B9}$$

for $l \geq 0$ [note in this connection that $j_{-1}(x) = n_0(x)$, the singular spherical Bessel function]. For the case when $ka \ll 1$, $I_l(a, u)$ can be evaluated approximately for $u = u^{(M)}_{ln}$ or $u = u^{(E)}_{ln}$ by using the appropriate expansion of $j_l(ka)$, as well as Eqs. (A17) and (A18):

$$I_l(a, u) \cong \frac{a^3 (ka)^{l-2}}{u(2l+1)!!} [x j_{l-1}(x) - (2l+1) j_l(x)]_{x=k(1-u)^{1/2}a}$$

$$= -\frac{a^3 (ka)^{l-1}}{(2l+1)!!} \frac{(1-u)^{1/2}}{u} j_{l+1}(k(1-u)^{1/2}a) \quad \text{for } ka \ll 1,$$

$$I_l(a, u^{(M)}_{ln}) \cong \frac{a^3 (ka)^l j_l(x_{l-1,n})}{(2l-1)!! x^2_{l-1,n}}, \tag{B10}$$

$$I_l(a, u^{(E)}_{ln}) \cong -\frac{a^3 (ka)^{l+2} j_{l-1}(x_{ln})}{l(2l-1)!! x^3_{ln}}, \quad n \geq 1$$

$$I_l(a, u^{(E)}_{l0}) \cong \frac{a^3 i^l (ka)^{2l} (l+1)}{(2l+1)(2l+1)!!(2l+3)!!} \left(\frac{l+1}{l}\right)^{1/2}.$$

In the long-wavelength limit $ka \ll 1$, $ka' \ll 1$, $k|\vec{b}-\vec{b}'| \ll 1$, only the largest value of λ needs to be kept in

the sums. If we use the asymptotic form of $h_\lambda^{(1)}$, as well as the approximate expressions for the A and B coefficients and the eigenvalues u obtained in Appendix A and the above expressions for I_l, we find for the matrix elements between states on different grains

$$\langle \Theta_b^+ C E^{(M)}_{b\,l\,mn} | \Gamma | \Theta_{b'}^+ E^{(M)}_{b'l'm'n'} \rangle \cong (-1)^{l'+m} \frac{(ka)(ka')}{x^2_{l\,-1n} x^2_{l'-1n'}} \times \frac{2}{2l+2l'+1} \left(\frac{4\pi ll'}{(l+1)(2l+1)(l'+1)(2l'+1)} \right)^{1/2} \begin{bmatrix} l+l'+m-m' \\ l+m \end{bmatrix}^{1/2} \begin{bmatrix} l+l'-m+m' \\ l-m \end{bmatrix}^{1/2} \times \left(\frac{a}{|\vec{b}'-\vec{b}|} \right)^{l+1/2} \left(\frac{a'}{|\vec{b}'-\vec{b}|} \right)^{l'+1/2} Y_{l'+l,m'-m}(\Omega_{b'-b}) = O(k^2), \tag{B11}$$

$$\langle \Theta_b^+ C E^{(E)}_{b\,l\,mn=0} | \Gamma | \Theta_{b'}^+ E^{(E)}_{b'l'm'n'=0} \rangle \cong (-1)^{l'+m} \frac{1}{(2l+1)(2l'+1)(2l+2l'+1)} \times \left(\frac{4\pi ll'}{(2l-1)(2l'+3)} \right)^{1/2} \begin{bmatrix} l+l'+m-m' \\ l+m \end{bmatrix}^{1/2} \begin{bmatrix} l+l'-m+m' \\ l-m \end{bmatrix}^{1/2} \times \left(\frac{a}{|\vec{b}'-\vec{b}|} \right)^{l+1/2} \left(\frac{a'}{|\vec{b}'-\vec{b}|} \right)^{l'+1/2} Y_{l'+l,m'-m}(\Omega_{b'-b}) = O(k^0), \tag{B12}$$

$$\langle \Theta_b^+ C E^{(E)}_{b\,l\,mn} | \Gamma | \Theta_{b'}^+ E^{(E)}_{b'l'm'n'} \rangle = \begin{cases} O(k^4) & \text{for } n \neq 0 \text{ and } n' \neq 0 \\ O(k^2) & \text{for } n = 0 \text{ and } n' \neq 0 \text{ or } n \neq 0 \text{ and } n' = 0, \end{cases} \tag{B13}$$

$$\langle \Theta_b^+ C E^{(M)}_{b\,l\,mn} | \Gamma | \Theta_{b'}^+ E^{(E)}_{b'l'm'n'} \rangle = \begin{cases} O(k^4) & \text{for } n' \neq 0 \\ O(k^2) & \text{for } n' = 0, \end{cases} \tag{B14}$$

$$\langle \Theta_b^+ C E^{(E)}_{b\,l\,mn} | \Gamma | \Theta_{b'}^+ E^{(M)}_{b'l'm'n'} \rangle = \begin{cases} O(k^4) & \text{for } n \neq 0 \\ O(k^2) & \text{for } n = 0. \end{cases} \tag{B15}$$

In Eqs. (B13)–(B15) we have not reproduced the complete results since we will now argue that they are not needed for a calculation of the scattered field in the long-wavelength limit.

The diagonal matrix elements of Γ are just the isolated sphere eigenvalues $s^{(E)}_{ln}$ and $s^{(M)}_{ln}$. From the results of Appendix A, these eigenvalues exhibit the following behavior in the long-wavelength limit:

$$s^{(M)}_{ln} = \frac{1}{u^{(M)}_{ln}} \cong -\left(\frac{ka}{x_{l\,-1n}} \right)^2 = O(k^2),$$

$$s^{(E)}_{ln} = \frac{1}{u^{(E)}_{ln}} \cong -\left(\frac{ka}{x_{ln}} \right)^2 = O(k^2), \quad n \geq 1 \tag{B16}$$

$$s^{(E)}_{l0} = \frac{1}{u^{(E)}_{l0}} \cong \frac{l}{2l+1} = O(k^0).$$

Consequently, it is easy to see that the subspace of $E^{(M)}$ states is unperturbed by the other states to lowest order, and the same is true of the subspace of $E^{(E)}$, $n=0$ states, and of the subspace of $E^{(E)}$, $n \neq 0$ states. Furthermore, the $E^{(E)}, n \neq 0$ states do not contribute in leading order to any of the multipole components in the expansion of the scattered field. Therefore, in discussing the long-wavelength scattering by a collection of spheres, we will have to diagonalize the Γ matrix only within the separate $E^{(M)}$ and $E^{(E)}, n=0$ subspaces. For this purpose only the matrix elements of Eqs. (B11) and (B12) are needed.

[1] For recent reviews and lists of references, see the articles in *Electrical Transport and Optical Properties of Inhomogeneous Media*, edited by J. C. Garland and D. B. Tanner (AIP, New York, 1978). The subject of electromagnetic scattering by small particles has also, of course, long been of intense interest in astrophysics and atmospheric physics, where there is an enormous literature, primarily on questions relating to scattering from isolated objects. A recent review relating to scattering by interstellar dust particles has been written by D. R. Huffman, Adv. Phys. 26, 129 (1977), which has numerous references.

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[9]See, e.g., P. M. Morse and H. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill, New York, 1953), p. 884.
[10]*Handbook of Mathematical Functions*, edited by M. Abramowitz and I. A. Stegun (Dover, New York, 1965), p. 370. The required sum rule follows from the representation of the spherical Bessel function $j_l(x)$ as an infinite product of factors $(1 - x^2/x_{ln}^2)$ when it is expanded in powers of x.
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