Absorption in multiple-scattering systems of coated spheres

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Received September 30, 2002; revised manuscript received January 27, 2003; accepted February 11, 2003

We derive formulas for rigorous transfer matrix calculations of absorption in multiple-coherent-scattering systems in which the scatterers are multiply coated spheres (not necessarily concentric). Any of the spherical coatings, cores, or host media may be composed of absorbing materials. For a nonabsorbing host media, the total absorption may be deduced from overall energy conservation. A more detailed description of the absorption is obtained through formulas yielding the absorption within individual scatterers and/or coatings. We present some illustrative applications of these formulas to the design of heterogeneous coated-sphere media exhibiting enhanced absorption. © 2003 Optical Society of America

OCIS codes: 290.4010, 290.4020, 160.4670, 290.1990, 290.5850.

1. INTRODUCTION

Theoretical treatments of multiple scattering of systems have undergone considerable progress in recent years.[1–8] These developments have been fueled in part by the increasing interest in exploiting multiple-scattering effects for the fabrication of inhomogeneous media with tailor-made electromagnetic properties. The range of properties obtainable with such media is, however, limited by the electromagnetic characteristics of available scattering materials. Singly and multiply coated scatterers are a means of enlarging this range by extending the characteristics of the scatterers. In this paper we shall consider scatterers consisting of concentric and nonconcentric multilayer spheres.

We shall derive procedures and formulas for evaluating the absorption in individual scatterers and coatings in multiple-scattering systems composed of coated spheres. Although the total absorption for absorbing scatterers immersed in nonabsorbing media may be deduced from the total cross sections,[3] the absorption within individual spheres can provide us with a more complete picture of the absorption. This additional information will prove particularly important in treating systems where absorption is present in both the scatterers and the host media.

Our derivation of the transfer matrix of coated spheres was chosen for its particularly stable numerical behavior in treating spheres with multiple coatings. The multiple-scattering aspects of the problem are treated by inserting the transfer matrices of coated spheres into a reliable recursive method for calculating the system transfer matrix.[1,4] A particularly useful property of the transfer matrix formulation is that it contains all scattering information for arbitrary incident waves. Among other advantages, this characteristic permits analytic calculations of orientation averages of a multiple-scattering system.

The outline of the paper is as follows. In Section 2 we briefly review a compact notation for field developments on a spherical partial-wave basis. In Section 3 we derive a formula for the transfer matrix of multiply coated spherical scatterers. Section 4 is devoted to the special case of concentric spheres. We present therein a normalization procedure of the field coefficients that further enhances the numeric stability of transfer matrix calculations of multiply coated scatterers.

We treat the multiple-scattering aspects of the problem in Section 5 by invoking a reliable recursive transfer matrix procedure. We treat absorption within individual

scatterers of an N-particle cluster in Section 6. We conclude the paper in Section 7 by presenting some illustrative calculations of the effective absorption length in multiple-scattering systems of coated spheres.

2. VECTOR PARTIAL-WAVE EXPANSIONS

This work is carried out in a time-harmonic framework with $\exp(-i\omega t)$ time dependence. An arbitrary incident field in an external (host) medium, denoted medium 0, can be developed in terms of regular partial waves developed about some chosen origin, $\mathbf{0}$, of the system:

$$\begin{aligned}\mathbf{E}_{\text{inc}}(\mathbf{r}) &= E\sum_{n=1}^{\infty}\sum_{m=-n}^{n}\left(\mathcal{R}g\{\mathbf{M}_{nm}(k_0\mathbf{r})\}[a]^{\mathbf{M}}_{nm}\right.\\ &\quad\left.+\ \mathcal{R}g\{\mathbf{N}_{nm}(k_0\mathbf{r})\}[a]^{\mathbf{N}}_{nm}\right)\\ &= E(\mathcal{R}g\{\mathbf{M}(k_0\mathbf{r})\},\ \mathcal{R}g\{\mathbf{N}(k_0\mathbf{r})\})\begin{pmatrix}a^{\mathbf{M}}\\ a^{\mathbf{N}}\end{pmatrix}\\ &\equiv E\mathcal{R}g\{\Xi^t(k_0\mathbf{r})\}a. \end{aligned}\tag{1}$$

In the last line, we introduce a condensed notation inspired by Chew,[8] in which a is an infinite-dimensional column vector composed of the coefficients of the incident-wave development. The elements of the abstract row vector $\Xi^t(k\mathbf{r})$ are the *normalized* basis set of *outgoing* vector partial waves $\mathbf{M}_{n,m}(k\mathbf{r})$ and $\mathbf{N}_{n,m}(k\mathbf{r})$,

$$\begin{aligned}\Xi^t(k\mathbf{r}) \equiv (&\mathbf{M}_{1,-1}(k\mathbf{r}),\ \mathbf{M}_{1,0}(k\mathbf{r}),\ldots,\\ &\mathbf{N}_{1,-1}(k\mathbf{r}),\ \mathbf{N}_{1,0}(k\mathbf{r}),\ldots),\end{aligned}\tag{2}$$

where the superscript t denotes the transpose.

The $\mathbf{M}_{nm}(k\mathbf{r})$ and $\mathbf{N}_{nm}(k\mathbf{r})$ functions can be conveniently expressed,

$$\begin{aligned}\mathbf{M}_{nm}(k\mathbf{r}) &\equiv -h_n(kr)\mathbf{X}_{nm}(\hat{\mathbf{r}}),\\ \mathbf{N}_{nm}(k\mathbf{r}) &\equiv \frac{1}{kr}\{\sqrt{n(n+1)}h_n(kr)\mathbf{Y}_{nm}(\hat{\mathbf{r}})\\ &\quad+[krh_n(kr)]'\mathbf{Z}_{nm}(\hat{\mathbf{r}})\},\end{aligned}\tag{3}$$

where $\mathbf{X}_{nm}(\hat{\mathbf{r}})$, $\mathbf{Y}_{nm}(\hat{\mathbf{r}})$, and $\mathbf{Z}_{nm}(\hat{\mathbf{r}})$ are the normalized vector spherical harmonics (see Appendix A). The h_n are the spherical Hankel functions of the first kind. The notation $\mathcal{R}g\{\ \}$ in Eq. (1) means "take the regular part of" and corresponds to replacing the spherical Hankel functions h_n in Eq. (3) with spherical Bessel functions, j_n.[8,9]

The scalar constant E in Eq. (1) is defined such that $E^2 \equiv 2I(\mathbf{0})[(\mu_0\mu_v)/(\varepsilon_0\epsilon_v)]^{1/2}$, where $I(\mathbf{0})$ is the incident irradiance (i.e., the norm of the incident Poynting vector) at the origin, and ϵ_v, μ_v are the vacuum permittivity and permeability, respectively. This convention for E imposes that the incident wave coefficients $[a]^{\mathbf{A}}_{nm}$ must be normalized such that[1] $\Sigma^1_{m=-1}[a^*]^{\mathbf{N}}_{1,m}[a]^{\mathbf{M}}_{1,m} = 6\pi$. For a homogeneous plane wave in a nonabsorbing medium, the above definition for E corresponds to the electric-field amplitude of the incident field, $E_{\text{p.w.}} = |\mathbf{E}_{\text{inc}}|$.

3. INDIVIDUAL TRANSFER MATRICES FOR COATED SPHERES

A. Boundary Conditions for Spheres with a Single Coating

In this section we derive the transfer matrix of a scatterer with a single spherical coating (see Fig. 1). A spherical region of radius R_1, centered on $\mathbf{x}_1$, is immersed in a host medium, denoted region 0. A second spherical region of radius R_2 and centered on $\mathbf{x}_2$ is immersed in region 1. Each of the homogeneous regions is characterized by complex relative constitutive parameters $\varepsilon_l(\omega)$ and $\mu_l(\omega)$, $l = 0, 1, 2$.

Labeling the coated sphere as the jth particle in an N-particle system, we may express the *total* electric field in medium 0, $\mathbf{E}_0$, as the sum of an excitation field on particle j, $\mathbf{E}_0^{(j),e}$, and the field scattered by this particle, $\mathbf{E}_0^{(j),s}$. At the surface of the scatterer, $\mathbf{E}_0^{(j),e}$ and $\mathbf{E}_0^{(j),s}$ may be developed in terms of spherical waves centered on $\mathbf{x}_1^{(j)}$,

$$\begin{aligned}\mathbf{E}_0 &= \mathbf{E}_0^{(j),e} + \mathbf{E}_0^{(j),s},\\ \mathbf{E}_0 &= E\mathcal{R}g\{\Xi^t(k_0\mathbf{r}_1^{(j)})\}e_0^{(j)}\\ &\quad+ E\Xi^t(k_0\mathbf{r}_1^{(j)})f_0^{(j)},\end{aligned}\tag{4}$$

where $\mathbf{r}_1^{(j)} \equiv \mathbf{r} - \mathbf{x}_1^{(j)}$.

In region 1, the field can be developed on a basis of regular spherical waves centered on $\mathbf{x}_1^{(j)}$ and outgoing spherical waves centered on $\mathbf{x}_2^{(j)}$,

$$\mathbf{E}_1^{(j)} = E\mathcal{R}g\{\Xi^t(k_1^{(j)}\mathbf{r}_1^{(j)})\}e_1^{(j)} + E\Xi^t(k_1\mathbf{r}_2^{(j)})f_1^{(j)}\tag{5a}$$

$$= E\mathcal{R}g\{\Xi^t(k_1^{(j)}\mathbf{r}_1^{(j)})\}e_1^{(j)} + E\Xi^t(k_1^{(j)}\mathbf{r}_1^{(j)})\beta^{[1,2]}_{k_1}f_1^{(j)}\tag{5b}$$

$$= E\mathcal{R}g\{\Xi^t(k_1^{(j)}\mathbf{r}_2^{(j)})\}\beta^{[2,1]}_{k_1}e_1^{(j)} + E\Xi^t(k_1^{(j)}\mathbf{r}_2^{(j)})f_1^{(j)},\tag{5c}$$

where Eqs. (5b) and (5c) are alternative developments centered respectively on $\mathbf{x}_1$ and $\mathbf{x}_2$. The $\beta^{[i,k]}_{k_l}$ are regular translation-addition matrices (see Appendix B) with the abbreviated notation $\beta^{[i,k]}_{k_l} \equiv \beta\,[k_l^{(j)}(\mathbf{x}_i^{(j)} - \mathbf{x}_k^{(j)})]$. Fields in the core (region 2) are developed in terms of regular waves centered on $\mathbf{x}_2^{(j)}$,

$$\mathbf{E}_2^{(j)} = E\mathcal{R}g\{\Xi^t(k_2^{(j)}\mathbf{r}_2^{(j)})\}e_2^{(j)},\tag{6}$$

where $e_2^{(j)}$ is the vector of excitation coefficients in the core. The magnetic fields in each region $l = 0, 1, 2$ are given by the Maxwell equation,

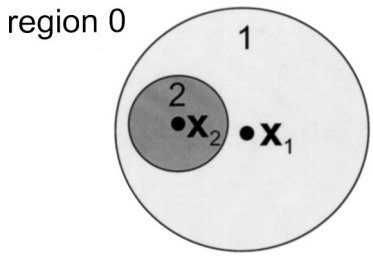

Fig. 1. Coated nonconcentric spherical scatterer.

$$i\omega\mu_l\mu_v\mathbf{H}_l(\mathbf{r}) = \nabla \times \mathbf{E}_l(\mathbf{r}), \tag{7}$$

where μ_v is the magnetic permeability of the vacuum.

The particle labels, superscript (j), used above are a nuisance as long as we are concerned with the scattering by a single particle. We will henceforth suppress the particle labels until Sections 5 and 6, in which we shall need to explicitly discuss the different particles in the system. With the particle labels suppressed, then, the continuity of the tangential electric field at the (0, 1) and (1, 2) interfaces yields the equations,

$$\begin{aligned}
&k_1\psi_n(k_0R_1)[e_0]^{\mathbf{M}}_{rm} + k_1\xi_n(k_0R_1)[f_0]^{\mathbf{M}}_{nm} \\
&\quad = k_0\psi_n(k_1R_1)[e_1]^{\mathbf{M}}_{nm} + k_0\xi_n(k_1R_1)[\beta^{[1,2]}_{k_1}f_1]^{\mathbf{M}}_{nm}, \\
&k_1\psi_n'(k_0R_1)[e_0]^{\mathbf{N}}_{nm} + k_1\xi_n'(k_0R_1)[f_0]^{\mathbf{N}}_{nm} \\
&\quad = k_0\psi_n'(k_1R_1)[e_1]^{\mathbf{N}}_{nm} + k_0\xi_n'(k_1R_1)[\beta^{[1,2]}_{k_1}f_1]^{\mathbf{N}}_{nm},
\end{aligned} \tag{8}$$

$$\begin{aligned}
&k_2\psi_n(k_1R_2)[\beta^{(2,1)}_{k_1}e_1]^{\mathbf{M}}_{nm} + k_2\xi_n(k_1R_2)[f_1]^{\mathbf{M}}_{nm} \\
&\quad = k_1\psi_n(k_2R_2)[e_2]^{\mathbf{M}}_{nm}, \\
&k_2\psi_n'(k_1R_2)[\beta^{(2,1)}_{k_1}e_1]^{\mathbf{N}}_{nm} + k_2\xi_n'(k_1R_2)[f_1]^{\mathbf{N}}_{nm} \\
&\quad = k_1\psi_n'(k_2R_2)[e_2]^{\mathbf{N}}_{nm},
\end{aligned} \tag{9}$$

where $\psi_n(x)$ and $\xi_n(x)$ are the Ricatti functions defined by $\psi_n(x) \equiv xj_n(x)$ and $\xi_n(x) \equiv xh_n(x)$.

The continuity of the tangential magnetic field at the interfaces then yields

$$\begin{aligned}
&\mu_1\psi_n'(k_0R_1)[e_0]^{\mathbf{M}}_{nm} + \mu_1\xi_n'(k_0R_1)[f_0]^{\mathbf{M}}_{nm} \\
&\quad = \mu_0\psi_n'(k_1R_1)[e_1]^{\mathbf{M}}_{nm} + \mu_0\xi_n'(k_1R_1)[\beta^{[1,2]}_{k_1}f_1]^{\mathbf{M}}_{nm}, \\
&\mu_1\psi_n(k_0R_1)[e_0]^{\mathbf{N}}_{nm} + \mu_1\xi_n(k_0R_1)[f_0]^{\mathbf{N}}_{nm} \\
&\quad = \mu_0\psi_n(k_1R_1)[e_1]^{\mathbf{N}}_{nm} + \mu_0\xi_n(k_1R_1)[\beta^{[1,2]}_{k_1}f_1]^{\mathbf{N}}_{nm},
\end{aligned} \tag{10}$$

$$\begin{aligned}
&\mu_2\psi_2'(k_1R_2)[\beta^{[2,1]}_{k_1}e_1]^{\mathbf{M}}_{nm} + \mu_2\xi_n'(k_1R_2)[f_1]^{\mathbf{M}}_{nm} \\
&\quad = \mu_1\psi_n'(k_2R_2)[e_2]^{\mathbf{M}}_{nm}, \\
&\mu_2\psi_n(k_1R_2)[\beta^{[2,1]}_{k_1}e_1]^{\mathbf{N}}_{nm} + \mu_2\xi_n(k_1R_2)[f_1]^{\mathbf{N}}_{nm} \\
&\quad = \mu_1\psi_n(k_2R_2)[e_2]^{\mathbf{N}}_{nm}.
\end{aligned} \tag{11}$$

B. Transfer Matrix for a Singly Coated Sphere

Equations (8)–(11) form a set of linear equations. One can therefore derive a linear relationship (transfer matrix) between the excitation coefficients e_0 and the scattering coefficients f_0 by eliminating all the other coefficients in the system. Numerical studies have shown[10–15] however, that it is preferable to solve these equations in a manner that emphasizes logarithmic derivatives and the ratios of Bessel functions.

Eliminating the coefficients e_1 from Eqs. (8) and (10) yields

$$[f_0]^{\mathbf{A}}_{nm} = [T_{10}]^{\mathbf{A}}_n[e_0]^{\mathbf{A}}_{nm} + [Q_{01}]^{\mathbf{A}}_n[\beta^{[1,2]}_{k_1}f_1]^{\mathbf{A}}_{nm}, \qquad \mathbf{A} = \mathbf{M},\ \mathbf{N}, \tag{12}$$

where $[T_{10}]^{\mathbf{A}}_n$ are the Mie coefficients[16–18] for a homogeneous sphere composed of medium 1 and immersed in medium 0 (i.e., taking $l = 0$ in the following expression),

$$\begin{aligned}
[T_{l+1,l}]^{\mathbf{M}}_n &= \frac{\psi_n(k_lR_{l+1})}{\xi_n(k_lR_{l+1})} \\
&\quad \times \frac{\dfrac{\mu_{l+1}}{\mu_l}\Phi_n(k_lR_{l+1}) - \rho_{l+1,l}\Phi_n(k_{l+1}R_{l+1})}{\rho_{l+1,l}\Phi_n(k_{l+1}R_{l+1}) - \dfrac{\mu_{l+1}}{\mu_l}\Psi_n(k_lR_{l+1})} \\
&\equiv \frac{\psi_n(k_lR_{l+1})}{\xi_n(k_lR_{l+1})} \times [\tilde{T}_{l+1,l}]^{\mathbf{M}}_n, \\
[T_{l+1,l}]^{\mathbf{N}}_n &= \frac{\psi_n(k_lR_{l+1})}{\xi_n(k_lR_{l+1})} \\
&\quad \times \frac{\dfrac{\mu_{l+1}}{\mu_l}\Phi_n(k_{l+1}R_{l+1}) - \rho_{l+1,l}\Phi_n(k_lR_{l+1})}{\rho_{l+1,l}\Psi_n(k_lR_{l+1}) - \dfrac{\mu_{l+1}}{\mu_l}\Phi_n(k_{l+1}R_{l+1})} \\
&\equiv \frac{\psi_n(k_lR_{l+1})}{\xi_n(k_lR_{l+1})} \times [\tilde{T}_{l+1,l}]^{\mathbf{N}}_n,
\end{aligned} \tag{13}$$

with $\rho_{l+1,l} \equiv k_{l+1}/k_l = [(\epsilon_{l+1}\mu_{l+1})/(\epsilon_l\mu_l)]^{1/2}$. The functions Φ and Ψ are the logarithmic derivatives of the Ricatti–Bessel functions: $\Phi_n(kr) \equiv \psi_n'(kr)/\psi_n(kr)$, $\Psi_n(kr) \equiv \xi_n'(kr)/\xi_n(kr)$.

It is important to recognize the definition of $[\tilde{T}_{l+1,l}]^{\mathbf{A}}_n$ as the part of the transfer matrix that can be expressed entirely in terms of logarithmic derivatives of Bessel functions. These and additional $\tilde{Q}$, $\tilde{P}$, $\tilde{V}$, $\tilde{U}$,... matrices will be used in Section 4 when we discuss relations among normalized coefficients. The coefficients $[Q_{01}]^{\mathbf{A}}_n$ in Eq. (12) are given (with $l = 0$) by

$$\begin{aligned}
[Q_{l,l+1}]^{\mathbf{M}}_n &= \frac{1}{\xi_n(k_lR_{l+1})\psi_n(k_{l+1}R_{l+1})} \\
&\quad \times \frac{i}{\dfrac{\mu_{l+1}}{\mu_l}\Psi_n(k_lR_{l+1}) - \rho_{l+1,l}\Phi_n(k_{l+1}R_{l+1})} \\
&= \frac{1}{\xi_n(k_lR_{l+1})\psi_n(k_{l+1}R_{l+1})}[\tilde{Q}_{l,l+1}]^{\mathbf{M}}_n, \\
[Q_{l,l+1}]^{\mathbf{N}}_n &= \frac{1}{\xi_n(k_lR_{l+1})\psi_n(k_{l+1}R_{l+1})} \\
&\quad \times \frac{i}{\rho_{l+1,l}\Psi_n(k_lR_{l+1}) - \dfrac{\mu_{l+1}}{\mu_l}\Phi_n(k_{l+1}R_{l+1})} \\
&\equiv \frac{1}{\xi_n(k_lR_{l+1})\psi_n(k_{l+1}R_{l+1})}[\tilde{Q}_{l,l+1}]^{\mathbf{N}}_n.
\end{aligned} \tag{14}$$

The notation can be compressed still further by defining matrices T_{10} and Q_{01} of the general form,

$$O = \begin{bmatrix} O^{\mathbf{MM}} & O^{\mathbf{MN}} \\ O^{\mathbf{NM}} & O^{\mathbf{NN}} \end{bmatrix}, \tag{15}$$

where the $O^{\mathbf{AB}}$ are matrices in the partial-wave space with elements written $[O]^{\mathbf{AB}}_{nm,\nu\mu}$. From Eq. (12) we see that $T_{l+1,l}$ and $Q_{l,l+1}$ are diagonal; i.e., their elements are of the form

$$[O]^{\mathbf{AB}}_{nm,\nu\mu} = \delta_{\mathbf{A},\mathbf{B}}\delta_{n,\nu}\delta_{m,\mu}[O]^{\mathbf{A}}_{n}. \tag{16}$$

In the matrix notation, Eq. (12) takes the compact and transparent form

$$f_0 = T_{10}e_0 + Q_{01}\beta^{[1,2]}_{k_1}f_1. \tag{17}$$

Eliminating the internal field coefficients e_2 from Eqs. (9) and (11) yields a standard Mie scattering equation,

$$f_1 = T_{21}\beta^{[2,1]}_{k_1}e_1, \tag{18}$$

where the addition theorem matrix $\beta^{[2,1]}_{k_1}$ is present on account of the fact that e_1 and f_1 express partial-wave developments centered on $\mathbf{x}_1$ and $\mathbf{x}_2$, respectively.

Eliminating f_1 from Eqs. (8) and (10) provides the relation

$$e_1 = V_{10}e_0 + U_{10}f_0, \tag{19}$$

where V_{10} and U_{10} are diagonal, and their components given by

$$\begin{aligned}
[V_{l+1,l}]^{\mathbf{M}}_n &= i\psi_n(k_lR_{l+1})\xi_n(k_{l+1}R_{l+1}) \\
&\quad\times\left(\frac{\mu_{l+1}}{\mu_l}\Phi_n(k_lR_{l+1}) - \rho_{l+1,l}\Psi_n(k_{l+1}R_{l+1})\right) \\
&\equiv \psi_n(k_lR_{l+1})\xi_n(k_{l+1}R_{l+1})[\tilde{V}_{l+1,l}]^{\mathbf{M}}_n, \\
[V_{l+1,l}]^{\mathbf{N}}_n &= i\psi_n(k_lR_{l+1})\xi_n(k_{l+1}R_{l+1}) \\
&\quad\times\left(\rho_{l+1,l}\Phi_n(k_lR_{l+1}) - \frac{\mu_{l+1}}{\mu_l}\Psi_n(k_{l+1}R_{l+1})\right) \\
&\equiv \psi_n(k_lR_{l+1})\xi_n(k_{l+1}R_{l+1})[\tilde{V}_{l+1,l}]^{\mathbf{N}}_n, \qquad (20) \\
[U_{l+1,l}]^{\mathbf{M}}_n &= i\xi_n(k_lR_{l+1})\xi_n(k_{l+1}R_{l+1}) \\
&\quad\times\left(\frac{\mu_{l+1}}{\mu_l}\Psi_n(k_lR_{l+1}) - \rho_{l+1,l}\Psi_n(k_{l+1}R_{l+1})\right) \\
&\equiv \xi_n(k_lR_{l+1})\xi_n(k_{l+1}R_{l+1})[\tilde{U}_{l+1,l}]^{\mathbf{M}}_n, \\
[U_{l+1,l}]^{\mathbf{N}}_n &= i\xi_n(k_lR_{l+1})\xi_n(k_{l+1}R_{l+1}) \\
&\quad\times\left(\rho_{l+1,l}\Psi_n(k_lR_{l+1}) - \frac{\mu_{l+1}}{\mu_l}\Psi_n(k_{l+1}R_{l+1})\right) \\
&\equiv \xi_n(k_lR_{l+1})\xi_n(k_{l+1}R_{l+1})[\tilde{U}_{l+1,l}]^{\mathbf{N}}_n. \qquad (21)
\end{aligned}$$

Eliminating the fields e_1 and f_1 from equations (17)–(19), we obtain an expression for the generalized transfer matrix, $\mathcal{T}_{10}$, of a two-layer spherical scatterer,

$$f_0 = \left[\frac{T_{10} + Q_{01}\beta^{[1,2]}_{k_1}T_{21}\beta^{[2,1]}_{k_1}V_{10}}{I - Q_{01}\beta^{[1,2]}_{k_1}T_{21}\beta^{[2,1]}_{k_1}U_{10}}\right]e_0 \equiv \mathcal{T}_{10}e_0, \tag{22}$$

where I is the identity matrix.

C. Spheres with Multiple Coatings

Let us now generalize to the case of a sphere with multiple coatings. The media of the innermost homogeneous sphere is labeled L. The index decreases by one with each successive coating until we arrive at the outermost coating, labeled 1 (see Fig. 2). A recursive relation for the transfer matrix is obtained after we notice that in the formula for $\mathcal{T}_{10}$, Eq. (22), the only information required of the interior sphere is its transfer matrix, T_{21}. Therefore, if the interior sphere is itself a coated sphere, we need only to replace T_{21} with $\mathcal{T}_{21}$. The overall transfer matrix of a multiply coated sphere can thus be obtained recursively by

$$\mathcal{T}_{l+1,l} = \left[\frac{T_{l+1,l} + Q_{l,l+1}\beta^{[l+1,l+2]}_{k_l}\mathcal{T}_{l+2,l+1}\beta^{[l+2,l+1]}_{k_l}V_{l+1,l}}{I - Q_{l,l+1}\beta^{[l+1,l+2]}_{k_l}\mathcal{T}_{l+2,l+1}\beta^{[l+2,l+1]}_{k_l}U_{l+1,l}}\right] \tag{23}$$

and is initialized by a Mie transfer matrix of the innermost sphere, $\mathcal{T}_{L,L-1} = T_{L,L-1}$.

If one is interested in detailed field information within the scatterer, the scattering coefficients f_{l+1} can be calculated by using the $\mathcal{T}_{l+1,l}$, via a recursion relation derived from Eqs. (22), and (17),

$$f_{l+1} = \beta^{[l+2,l+1]}_{k_{l+1}}P_{l,l+1}(I - T_{l+1,l}\mathcal{T}^{-1}_{l+1,l})f_l, \tag{24}$$

where $\underline{P}$ is defined as the inverse of $\underline{Q}$:

$$\begin{aligned}
[P_{l,l+1}]^{\mathbf{M}}_n &\equiv [[Q_{l,l+1}]^{\mathbf{M}}_n]^{-1} \equiv \xi_n(k_lR_{l+1})\psi_n(k_{l+1}R_{l+1}) \\
&\quad\times[\tilde{P}_{l,l+1}]^{\mathbf{M}}_n, \\
[P_{l,l+1}]^{\mathbf{N}}_n &\equiv [[Q_{l,l+1}]^{\mathbf{N}}_n]^{-1} \equiv \xi_n(k_lR_{l+1})\psi_n(k_{l+1}R_{l+1}) \\
&\quad\times[\tilde{P}_{l,l+1}]^{\mathbf{N}}_n. \qquad (25)
\end{aligned}$$

The recurrence in Eq. (24) is initialized by the f_0, calculated by means of the multiple-scattering theory presented in Section 5 below.

The excitation coefficients are obtained from the relations

$$\begin{aligned}
e_l &= \beta^{[l,l+1]}_{k_l}\mathcal{T}^{-1}_{l+1,l}f_l, \qquad l = 1,\ldots,\ L-1, \\
[e_L]^{\mathbf{A}}_{nm} &= [\Lambda_{L,L-1}]^{\mathbf{A}}_n[f_{L-1}]^{\mathbf{A}}_{nm}, \qquad (26)
\end{aligned}$$

where the $[\Lambda_{L,L-1}]^{\mathbf{A}}_n$ elements are

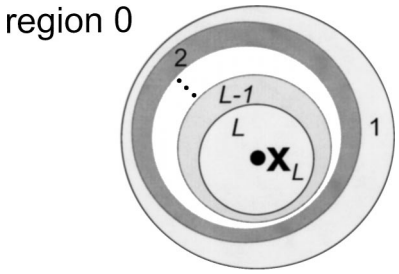

Fig. 2. Multiply coated spherical scatterer.

$$[\Lambda_{L,L-1}]_n^{\mathbf{M}} = \frac{1}{\psi_n(k_{L-1}R_L)\psi_n(k_LR_L)} \times \frac{i\rho_{L,L-1}}{\frac{\mu_{L-1}}{\mu_L}\rho_{L,L-1}\Phi_n(k_LR_L) - \Phi_n(k_{L-1}R_L)} \equiv \frac{1}{\psi_n(k_{L-1}R_L)\psi_n(k_LR_L)}[\tilde{\Lambda}_{L,L-1}]_n^{\mathbf{M}},$$

For concentric spheres in the normalized notation, we find

$$[\bar{f}_0]_{nm}^{\mathbf{A}} = [\tilde{\mathcal{T}}_{10}]_n^{\mathbf{A}}[\bar{e}_0]_{nm}^{\mathbf{A}}, \qquad [\mathcal{T}_{l+1,l}]_n^{\mathbf{A}} \equiv \frac{\psi_n(k_lR_{l+1})}{\xi_n(k_lR_{l+1})}[\tilde{\mathcal{T}}_{l+1,l}]_n^{\mathbf{A}}, \tag{30}$$

where the elements $[\tilde{\mathcal{T}}_{10}]_n^{\mathbf{A}}$ are obtained by means of bounded recurrence relations:

$$[\tilde{\mathcal{T}}_{l+1,l}]_n^{\mathbf{A}} = \left[\frac{[\tilde{\mathrm{T}}_{l+1,l}]_n^{\mathbf{A}} + \frac{\xi_n(k_{l+1}R_{l+1})}{\xi_n(k_{l+1}R_{l+2})}\frac{\psi_n(k_{l+1}R_{l+2})}{\psi_n(k_{l+1}R_{l+1})}[\tilde{Q}_{l,l+1}]_n^{\mathbf{A}}[\tilde{\mathcal{T}}_{l+2,l+1}]_n^{\mathbf{A}}[\tilde{V}_{l+1,l}]_n^{\mathbf{A}}}{1 - \frac{\xi_n(k_{l+1}R_{l+1})}{\xi_n(k_{l+1}R_{l+2})}\frac{\psi_n(k_{l+1}R_{l+2})}{\psi_n(k_{l+1}R_{l+1})}[\tilde{Q}_{l,l+1}]_n^{\mathbf{A}}[\tilde{\mathcal{T}}_{l+2,l+1}]_n^{\mathbf{A}}[\tilde{U}_{l+1,l}]_n^{\mathbf{A}}}\right],$$
$$[\tilde{\mathcal{T}}_{L,L-1}]_n^{\mathbf{A}} = [\tilde{\mathcal{T}}_{L,L-1}]_n^{\mathbf{A}}, \qquad \mathbf{A} = \mathbf{M},\ \mathbf{N}. \tag{31}$$

$$[\Lambda_{L,L-1}]_n^{\mathbf{N}} = \frac{1}{\psi_n(k_{L-1}R_L)\psi_n(k_LR_L)} \times \frac{i\rho_{L,L-1}}{\Phi_n(k_LR_L) - \frac{\mu_{L-1}}{\mu_L}\rho_{L,L-1}\Phi_n(k_{L-1}R_L)} \equiv \frac{1}{\psi_n(k_{L-1}R_L)\psi_n(k_LR_L)}[\tilde{\Lambda}_{L,L-1}]_n^{\mathbf{N}}. \tag{27}$$

4. CONCENTRIC SPHERES

Concentric spheres are an important special case of Eq. (23) and present useful numerical simplifications. If all the spherical interfaces have the same center, the transfer matrices become diagonal, and each individual component $[\mathcal{T}_{l+1,l}]_n^{\mathbf{A}}$ of the transfer matrices can then be obtained recursively:

$$[\mathcal{T}_{l+1,l}]_n^{\mathbf{A}} = \left[\frac{[T_{l+1,l}]_n^{\mathbf{A}} + [Q_{l,l+1}]_n^{\mathbf{A}}[\mathcal{T}_{l+2,l+1}]_n^{\mathbf{A}}[V_{l+1,l}]_n^{\mathbf{A}}}{1 - [Q_{l,l+1}]_n^{\mathbf{A}}[\mathcal{T}_{l+2,l+1}]_n^{\mathbf{A}}[U_{l+1,l}]_n^{\mathbf{A}}}\right], \quad \mathbf{A} = \mathbf{M},\ \mathbf{N}. \tag{28}$$

A. Normalized Coefficients

The numerical properties of Eq. (28) can be greatly improved by adopting a normalization of the coefficients along the lines proposed by Sitarski,[12,14,19] after which only logarithmic derivatives of Bessel functions and ratios of Bessel functions appear in the recurrence formulas of the transfer matrices.

We define normalized coefficients $\bar{f}_l$, $\bar{e}_l$ as

$$[\bar{f}_l]_{nm}^{\mathbf{A}} \equiv \xi_n(k_lR_{l+1})[f_l]_{nm}^{\mathbf{A}} \qquad (l = 0,\ldots,\ L-1) \quad \mathbf{A} = \mathbf{M},\ \mathbf{N},$$
$$[\bar{e}_0]_{nm}^{\mathbf{A}} \equiv \psi_n(k_0R_1)[e_0]_{nm}^{\mathbf{A}}, \qquad [\bar{e}_l]_{nm}^{\mathbf{A}} \equiv \psi_n(k_lR_l)[e_l]_{nm}^{\mathbf{A}} \qquad (l = 1,\ldots,\ L)\mathbf{A} = \mathbf{M},\ \mathbf{N}. \tag{29}$$

We recall that the bounded $\tilde{T}$, $\tilde{Q}$, $\tilde{V}$, and $\tilde{U}$ matrices (defined in Section 3) are all expressed entirely in terms of logarithmic derivatives of Bessel functions that can be reliably computed by using the recurrence relations developed in the literature.[14] The ratios $\psi_n(z)/\xi_n(z)$ can be calculated reliably for complex arguments by using recurrence relations, or, alternatively, one can calculate $[\psi_n(z_1)]/[\psi_n(z_2)]$ and $[\xi_n(z_1)]/[\xi_n(z_2)]$. If one wishes to use Eq. (31) to model materials with a radial dependent refractive index by calculating a large number of very thin layers,[15] it is preferable to avoid round-off errors by approximating the ratios of Bessel functions $R_{l+1} = R_{l+2} + \delta$, $\delta \ll R_{l+2}$ to obtain a nonrecursive differential form of this equation.

The recurrence relation for the normalized $\bar{f}_l$ coefficients is

$$[\bar{f}_{l+1}]_{nm}^{\mathbf{A}} = \psi_n(k_{l+1}R_{l+1})\xi_n(k_{l+1}R_{l+2})[\tilde{P}_{l,l+1}]_n^{\mathbf{A}} \times \left(1 - [\tilde{T}_{l+1,l}]_n^{\mathbf{A}}[\tilde{\mathcal{T}}_{l+1,l}^{-1}]_n^{\mathbf{A}}\right)[\bar{f}_l]_{nm}^{\mathbf{A}}, \tag{32}$$

and the normalized excitation coefficients are given by

$$[\bar{e}_0]_{nm}^{\mathbf{A}} = [\tilde{\mathcal{T}}_{10}^{-1}]_{nm}^{\mathbf{A}}[\bar{f}_0]_{nm}^{\mathbf{A}},$$
$$[\bar{e}_l]_{nm}^{\mathbf{A}} = \frac{\psi_n(k_lR_l)}{\psi_n(k_lR_{l+1})}[\tilde{\mathcal{T}}_{l+1,l}^{-1}]_n^{\mathbf{A}}[\bar{f}_l]_{nm}^{\mathbf{A}}, \qquad (l = 1,\ldots,\ L-1),$$
$$[\bar{e}_L]_{nm}^{\mathbf{A}} = \frac{1}{\psi_n(k_{L-1}R_L)\xi_n(k_{L-1}R_L)}[\tilde{\Lambda}_{L,L-1}]_n^{\mathbf{A}}[\bar{f}_{L-1}]_{nm}^{\mathbf{A}}. \tag{33}$$

5. MULTIPLE-SCATTERING FORMULATION

In this section we briefly describe a reliable recursive algorithm[1] for the calculation of the body-centered T-matrix.[1,5] Unlike previous recursive algorithms,[8] this method is not plagued by severe numerical errors.[20] Furthermore, when employing this technique, one is no longer obliged to consider that additional scatterers are added outside a circumscribing sphere surrounding all the other scatterers,[8] and spheres can be added to the system in an arbitrary order.

As we saw in Section 3, the individual transfer matrices $\mathcal{T}_{10}^{(j)}$ give the scattering information of the entire coated scatterer,

$$f_0^{(j)} = \mathcal{T}_{10}^{(j)} e_0^{(j)} \equiv \tau_1^{(j)} e_0^{(j)}, \tag{34}$$

and the $\mathcal{T}_{10}^{(j)}$ therefore contain all the information necessary for a multiple-scattering calculation. The index 10 on $\mathcal{T}$ is unnecessary and repetitive in this context, and therefore in this section we replace the transfer matrix $\mathcal{T}_{10}^{(j)}$ by the notation $\tau_1^{(j)}$. The index 1 in $\tau_1^{(j)}$ denotes that the transfer matrix describes a single scatterer, and such matrix shall be henceforth called the one-body transfer matrix.

The solution of an electromagnetic wave interacting with an N-particle system is then formulated in terms of N-body transfer matrices $\tau_N^{(j,k)}$, containing all multiple-scattering information and satisfying[1,5]

$$f_0^{(j)} = \sum_{k=1}^{N} \tau_N^{(j,k)} \beta^{(k,0)} a, \tag{35}$$

where we recall that a are the coefficients of the field incident on the entire system.

Our recursive solution for the N-body matrices, $\tau_N^{(j,k)}$, is to add a new scatterer, N, to a known solution of $\tau_{N-1}^{(j,k)}$ matrices describing a system of $N-1$ scatterers,[1,2]

$$\tau_N^{(N,N)} = \tau_1^{(N)} \left[1 - \sum_{j,k=1}^{N-1} \alpha^{(N,k)} \tau_{N-1}^{(k,j)} \alpha^{(j,N)} \tau_1^{(N)} \right]^{-1}, \tag{36}$$

where the $\alpha_l^{[i,k]}$ are the irregular translation-addition matrices (see Appendix B) between the centers of scatterers i and k, with the notation $\alpha^{[i,k]} \equiv \alpha[k_0(\mathbf{x}_1^{(i)} - \mathbf{x}_1^{(k)})]$.

The $\tau_N^{(i,N)}$, $\tau_N^{(N,i)}$ matrices are then obtained by matrix multiplications:

$$\tau_N^{(N,k)} = \tau_N^{(N,N)} \sum_{j=1}^{N-1} \alpha^{(N,j)} \tau_{N-1}^{(j,k)}, \qquad k \neq N,$$

$$\tau_N^{(j,N)} = \sum_{k=1}^{N-1} \tau_{N-1}^{(j,k)} \alpha^{(k,N)} \tau_N^{(N,N)}, \qquad j \neq N. \tag{37}$$

The readjustment of the $\tau_{N-1}^{(j,k)}$ matrices to become $\tau_N^{(j,k)}$ matrices is given by

$$\tau_N^{(j,k)} = \tau_{N-1}^{(j,k)} + \sum_{i=1}^{N-1} \tau_{N-1}^{(j,i)} \alpha^{(i,N)} \tau_N^{(N,k)}, \qquad j \neq N, \ k \neq N. \tag{38}$$

The $\tau_N^{(j,k)}$ matrices are thus obtained by recurrence, starting with an arbitrary $\tau_1^{(j)}$ scatterer of the system and successively adding all the other scatterers of the system.

In the case of an absorption-free media, the total-absorption cross section of the system σ_a may be obtained from energy conservation, $\sigma_a^{\mathrm{cl}} = \sigma_c^{\mathrm{cl}} - \sigma_s^{\mathrm{cl}}$, where σ_e^{cl} and σ_s^{cl} are the cluster extinction and scattering cross sections respectively. For the case of incident plane waves, σ_e^{cl} and σ_s^{cl} can be expressed[3] as

$$\sigma_e^{\mathrm{cl}} = -\frac{1}{k^2} \sum_{j,l}^{N} \mathrm{Re}\{\exp[i\mathbf{k}_0 \cdot (\mathbf{x}_l - \mathbf{x}_j)] p^\dagger \tau_N^{(j,l)} p\},$$

$$\sigma_s^{\mathrm{cl}} = \frac{1}{k^2} \mathrm{Re}\left\{ \sum_{j,k,l,i}^{N} \exp[i\mathbf{k}_0 \cdot (\mathbf{x}_i - \mathbf{x}_l)] \times p^\dagger [T_N^{(j,l)}]^\dagger \beta^{(j,k)} \tau_N^{(k,i)} p \right\}, \tag{39}$$

where the p are coefficients of a homogeneous incident wave (see Appendix A). Similarly, the orientation-averaged absorption for plane waves can be obtained from $\langle \sigma_a^{\mathrm{cl}} \rangle_o = \langle \sigma_e^{\mathrm{cl}} \rangle_o - \langle \sigma_s^{\mathrm{cl}} \rangle_o$, and the formulas[1,5]

$$\langle \sigma_e^{\mathrm{cl}} \rangle_o = -\frac{2\pi}{k^2} \sum_{j,l}^{N} \mathrm{Re}\{\mathrm{Tr}\{\tau_N^{(j,l)} \beta^{(l,j)}\}\},$$

$$\langle \sigma_s^{\mathrm{cl}} \rangle_o = \frac{2\pi}{k^2} \mathrm{Re}\left\{ \sum_{j,k,i,l}^{N} \mathrm{Tr}\{[\tau_N^{(j,l)}]^\dagger \beta^{(j,k)} \tau_N^{(k,i)} \beta^{(i,l)}\} \right\}. \tag{40}$$

6. ABSORPTION CROSS SECTIONS FOR COATED SPHERES

The absorption flux, $P_l^{(j)}$, through the outermost interface bounding region l is calculated by integrating the scalar product of the Poynting vector with the inward normal to the lth interface, $-\hat{\mathbf{r}}_l^{(j)}$,

$$P_l^{(j)} = -\oint_A \hat{\mathbf{r}}_l^{(j)} \cdot \mathbf{S}_l^{(j)} \mathrm{d}A = -\frac{(R_l^{(j)})^2}{2} \times \int \mathrm{d}\Omega \hat{\mathbf{r}}_l^{(j)} \cdot \mathrm{Re}\{\mathbf{E}_l^{(j)} \times \mathbf{H}_l^{(j)*}\}, \tag{41}$$

where $R_l^{(j)}$ is the radius of the interface.

Invoking Eqs. (5b), (7), (24), and the relation

$$e_l^{(j)} = (V_{l,l-1}^{(j)} [\mathcal{T}_{l,l-1}^{(j)}]^{-1} + U_{l,l-1}^{(j)}) f_{l-1}^{(j)}, \tag{42}$$

we find that the $\sigma_{a,l}^{(j)}$ can be conveniently expressed as

$$\sigma_{a,l}^{(j)} = \frac{1}{|k_l^{(j)}|^2} \mathrm{Re}\left\{ \sqrt{\frac{\mu_0}{\varepsilon_0}} f_{l-1}^{(j)\dagger} \Gamma_l^{(j)} f_{l-1}^{(j)} \right\}, \tag{43}$$

with

$$\Gamma_l^{(j)} = [P_{l-1,l}^{(j)}(I - T_{l,l-1}^{(j)}[\mathcal{T}_{l,l-1}^{(j)}]^{-1})]^\dagger C_l^{(j)}[P_{l-1,l}^{(j)}(I - T_{l,l-1}^{(j)}[\mathcal{T}_{l,l-1}^{(j)}]^{-1})] + [V_{l,l-1}^{(j)}[\mathcal{T}_{l,l-1}^{(j)}]^{-1} + U_{l,l-1}^{(j)}]^\dagger D_l^{(j)}[V_{l,l-1}^{(j)}[\mathcal{T}_{l,l-1}^{(j)}]^{-1} + U_{l,l-1}^{(j)}] + [V_{l,l-1}^{(j)}[\mathcal{T}_{l,l-1}^{(j)}]^{-1} + U_{l,l-1}^{(j)}]^\dagger F_l^{(j)}[P_{l-1,l}^{(j)}(I - T_{l,l-1}^{(j)}[\mathcal{T}_{l,l-1}^{(j)}]^{-1})] + [P_{l-1,l}^{(j)}(I - T_{l,l-1}^{(j)} \times [\mathcal{T}_{l,l-1}^{(j)}]^{-1})]^\dagger G_l^{(j)}[V_{l,l-1}^{(j)}[\mathcal{T}_{l,l-1}^{(j)}]^{-1} + U_{l,l-1}^{(j)}]. \quad (44)$$

The $C_l^{(j)}$, $D_l^{(j)}$, $F_l^{(j)}$ and $G_l^{(j)}$ are diagonal matrices of the form of Eq. (16) with

$$[C_l^{(j)}]_n^{\mathbf{A}} \equiv i[\tilde{\eta}_l^{(j)}]^{\mathbf{A}}\xi_n^*(k_l^{(j)}R_l^{(j)})\xi_n'(k_l^{(j)}R_l^{(j)}),$$
$$[D_l^{(j)}]_n^{\mathbf{A}} \equiv i[\tilde{\eta}_l^{(j)}]^{\mathbf{A}}\psi_n^*(k_l^{(j)}R_l^{(j)})\psi_n'(k_l^{(j)}R_l^{(j)}),$$
$$[F_l^{(j)}]_n^{\mathbf{A}} \equiv i[\tilde{\eta}_l^{(j)}]^{\mathbf{A}}\psi_n^*(k_l^{(j)}R_l^{(j)})\xi_n'(k_l^{(j)}R_l^{(j)}),$$
$$[G_l^{(j)}]_n^{\mathbf{A}} \equiv i[\tilde{\eta}_l^{(j)}]^{\mathbf{A}}\psi_n'(k_l^{(j)}R_l^{(j)})\xi_n^*(k_l^{(j)}R_l^{(j)}),$$
$$\mathbf{A} = \mathbf{M}, \ \mathbf{N}, \quad (45)$$

where the $[\tilde{\eta}_l^{(j)}]^{\mathbf{A}}$ factors are defined by

$$[\tilde{\eta}_l^{(j)}]^{\mathbf{M}} \equiv (\varepsilon_l^{(j)}/\mu_l^{(j)})^{1/2}, [\tilde{\eta}_l^{(j)}]^{\mathbf{N}} \equiv (\varepsilon_l^{(j),*}/\mu_l^{(j),*})^{1/2}.$$

The absorption cross section of an entire coated sphere j is given by $\sigma_{\mathrm{a},1}^{(j)}$. The orientation average of $\sigma_{\mathrm{a},1}^{(j)}$, denoted $\langle \sigma_{\mathrm{a},1}^{(j)} \rangle_o$, is readily obtained from Eqs. (43) and (35), on which one applies an analytic average over all possible polarizations and directions of the incident field vector $\hat{\mathbf{k}}_i$. Following the prescription in Refs. 1 and 5 for orientation averages yields

$$\langle \sigma_{\mathrm{a},1}^{(j)} \rangle_o = \frac{2\pi}{|k_1^{(j)}|^2} \sum_{i,l=1}^{N} \times \mathrm{Re}\left\{ \sqrt{\frac{\mu_0}{\varepsilon_0}} \times \mathrm{Tr}\{[\tau_N^{(j,l)}]^\dagger \Gamma_1^{(j)} \tau_N^{(j,i)} \beta^{(i,l)}\} \right\}. \quad (46)$$

For concentric spheres, one obtains a rather compact and numerically reliable expression in terms of normalized matrix elements:

$$\sigma_{\mathrm{a},l}^{(j)} = \frac{1}{|k_l^{(j)}|^2} \sum_{\mathbf{A}=\mathbf{M},\mathbf{N}} \sum_{n,m} |\psi_n^{(j)}(k_l^{(j)}R_l^{(j)}) \times \xi_n^{(j)}(k_l^{(j)}R_l^{(j)})|^2 |[\bar{T}_{l-1}^{(j)}]_{nm}^{\mathbf{A}}|^2 \times \mathrm{Re}\left\{ \sqrt{\frac{\mu_0}{\varepsilon_0}} (i\Phi_n(k_l^{(j)}R_l^{(j)})[\tilde{\eta}_l^{(j)}]^{\mathbf{A}} |[\tilde{A}_l^{(j)}]_n^{\mathbf{A}}|^2 + i\Psi_n(k_l^{(j)}R_l^{(j)})[\tilde{\eta}_l^{(j)}]^{\mathbf{A}} |[\tilde{B}_l^{(j)}]_n^{\mathbf{A}}|^2 + i\Psi_n(k_l^{(j)}R_l^{(j)}) \times [\tilde{\eta}_l^{(j)}]^{\mathbf{A}} [\tilde{A}_l^{(j)}]_n^{\mathbf{A},*} [\tilde{B}_l^{(j)}]_n^{\mathbf{A}} + i\Phi_n(k_l^{(j)}R_l^{(j)}) \times [\tilde{\eta}_l^{(j)}]^{\mathbf{A}} [\tilde{A}_l^{(j)}]_n^{\mathbf{A}} [\tilde{B}_l^{(j)}]_n^{\mathbf{A},*}) \right\}, \quad (47)$$

with the definitions

$$[\tilde{A}_l^{(j)}]_n^{\mathbf{A}} \equiv [\tilde{V}_{l,l-1}^{(j)}]_n^{\mathbf{A}} [[\tilde{\mathcal{T}}_{l,l-1}^{(j)}]_n^{\mathbf{A}}]^{-1} + [\tilde{U}_{l,l-1}^{(j)}]_n^{\mathbf{A}},$$
$$[\tilde{B}_l^{(j)}]_n^{\mathbf{A}} \equiv [\tilde{P}_{l-1,l}^{(j)}]_n^{\mathbf{A}} (1 - [\tilde{T}_{l,l-1}^{(j)}]_n^{\mathbf{A}} [[\tilde{T}_{l,l-1}^{(j)}]_n^{\mathbf{A}}]^{-1}). \quad (48)$$

7. REDUCTION OF THE ABSORPTION LENGTH IN COATED-SPHERE MEDIA

In this section we show how inhomogeneous media composed of coated spheres may exhibit enhanced absorption properties. Furthermore, we demonstrate that simulations may be able to determine the optimal design of such media. The model problem that we consider is the enhancement of absorption per unit volume of a material of index $n_1 = 1.5 + 0.01i$ imbedded in a transparent media of index $n_0 = 1.5$. Without going into considerations of an interface with an external medium, we shall simply take that enhanced absorption corresponds to a reduced effective absorption length $l_{\mathrm{a}}^{\mathrm{eff}}$ within an inhomogeneous medium.

A. Low-Density Systems

For the purpose of comparison, let us first consider material 1 to be dispersed as "small" grains (i.e., much smaller than the in-medium wavelength) within a material 0. The weak dielectric contrast renders scattering negligible in comparison with absorption, which implies an approximately zero scattering cross section, $\sigma_{\mathrm{s}}^{\mathrm{disp}} \simeq 0$, for the dispersed grains of material 1. The absorption (extinction) cross sections of the dispersed grains are then given by

$$\sigma_{\mathrm{e}}^{\mathrm{disp}} \simeq \sigma_{\mathrm{a}}^{\mathrm{disp}} \simeq 2n_1'' \frac{\omega}{c} V_1 = \frac{4\pi n_1''}{\lambda_v} V_1, \quad (49)$$

where λ_v is the vacuum wavelength of the radiation and V_1 the volume of the grains of material 1. The absorption length within the inhomogeneous composite is then

$$l_{\mathrm{a}} \simeq \frac{1}{N_1 \sigma_{\mathrm{a}}^{\mathrm{disp}}} = \frac{1}{2f_1 k_v n_1''}, \quad (50)$$

where N_1 and f_1 are respectively the number density and the volume density of absorbing material 1. To eliminate the wavelength dependence of l_{a}, which is simply a scale factor, it seems preferable to speak in terms of a dimensionless absorption length, $k_v l_{\mathrm{a}} = 2\pi l_{\mathrm{a}}/\lambda_v$:

$$k_v l_{\mathrm{a}} \simeq \frac{1}{2f_1 n_1''} = \frac{50}{f_1}, \quad (51)$$

where, we have used our chosen value of $n_1'' = 0.01$.

We now propose to decrease the effective absorption length by placing material 1 as concentric coatings around spherical grains of a high-index scatterer such as TiO^2 ($n_2 \simeq 2.5$). The coated grains are then randomly dispersed in medium n_0. At low concentrations, the scattering mean free path within the medium is $l_{\mathrm{s}} \simeq 1/(N_{\mathrm{s}}\sigma_{\mathrm{s}})$, where σ_{s} and N_{s} are respectively the scattering cross section and the number density of the coated scatterers. The absorption mean free path is likewise $l_{\mathrm{a}} \simeq 1/(N_{\mathrm{s}}\sigma_{\mathrm{a}})$. The average number of scatterings that the light undergoes before being absorbed, denoted ν, is therefore $\nu = l_{\mathrm{a}}/l_{\mathrm{s}} = \sigma_{\mathrm{s}}/\sigma_{\mathrm{a}}$.

The asymmetry parameter g of the individual scatterings gives the projection of the scattered component of light along the direction of incidence after a single collision.[16] With the usual transport assumption that after m collisions, the projection of the scattered light along the initial direction of incidence is g^m, correlated random-walk calculations yield that the average distance

that the light travels before being absorbed, henceforth referred to as the effective absorption length l_a^{eff} is given by

$$k_v l_a^{\text{eff}} = \frac{\chi_1\{\nu + 2[g(\nu - 1) - \nu g^2 + g^{\nu+1}]/(1-g)^2\}^{1/2}}{n_0 f_s Q_s}, \tag{52}$$

where f_s is the volume density occupied by the coated spheres and the scattering efficiency has its usual definition, $Q_s \equiv \sigma_s/(\pi R_1^2)$. If the scatterings completely randomize the light propagation, then $g = 0$, and one obtains the classical diffusion result that the average distance traveled is proportional to $\sqrt{\nu}$, and $k_v l_a^{\text{eff}} = k_v l_s \sqrt{\nu}$. The limit of entirely forward scattering, i.e., $g \to 1$, yields $k_v l_a^{\text{eff}} \to k_v/N_s\sigma_a$, as one would expect.

With the above model parameters, calculations indicate that $k_v l_a^{\text{eff}}$ is minimized for optimal values of [see Fig. 3(a)] $R_1 - R_2 \simeq 0.18R_2$ and $\chi_1 = k_0 R_1 \simeq 2.2$. With these values, one obtains from Eq. (52) that $k_v l_a^{\text{eff}} \simeq 5.1/f_1$, i.e., a factor of 10 decrease in the absorption length per unit volume compared with that of homogeneous absorbing grains. The asymmetry parameter played an important role in these calculations since resonant scatterings tend to scatter light in the forward direction. This is clear from Fig. 3(b), where we plot the g of the scatterings as a function of the size parameter.

B. High-Density Multiple-Scattering Media

The predictions of Eq. (52) apply only in the low scatterer-density limit. At high densities coherent-scattering effects will come into play. Coherent-scattering effects for a finite number of scatterers can be treated by the methods of Section 5. In the design of inhomogeneous media however, we seek to calculate information to be put into a transport calculation where an averaging process is being carried at some point (e.g., configuration averaging). A study of the best approximation to be employed is beyond the scope of the present paper, and here we treat only one possible approximation technique in order to estimate coherent-scattering effects within the context of Eq. (52) above.

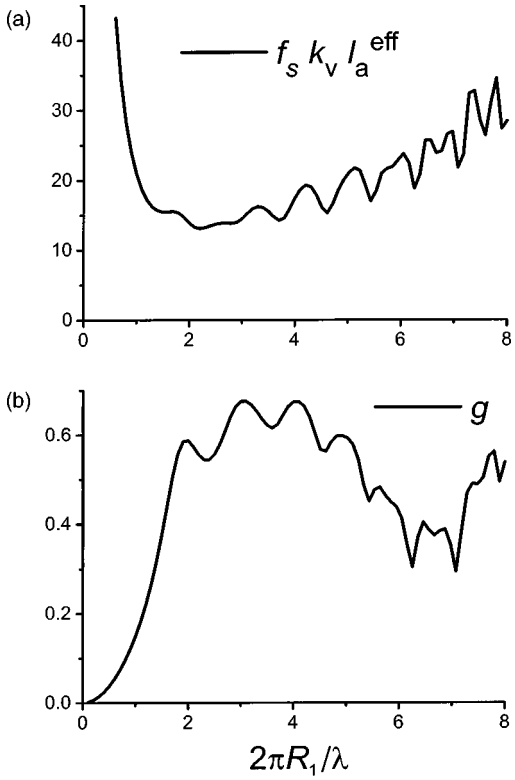

Fig. 3. (a) Dimensionless effective absorption length plotted as a function of the size parameter, $\chi_1 = 2\pi R_1/\lambda_0$ for $R_1 - R_2 = 0.18R_2$. (b) Asymmetry parameter of the scatterings plotted as a function of χ_1.

Since coherent-scattering effects are largely limited to nearest-neighbor interactions, we shall model them by taking a coated sphere with the optimized size and coating parameters of the independent-scattering case and surround it by 12 identical neighbors in a close-packing configuration. We seek to calculate averaged modifications to the independent scatters of Eq. (52). The most outstanding drawback of Eq. (52) is that it does not take into account self-polarization effects (i.e., modifications to the light scattered by the particle as a result of this light returning to the particle after having undergone other scattering events). A diagrammatic analysis of scattering[21] shows that self-polarization effects are those diagrams whose first and last scattering events are on the same scatterer. Adapting the diagrammatic analysis to the exact scattering framework shows that these diagrams correspond to the $\tau_N^{(j,j)}$ matrices.

We therefore calculate the $\tau_N^{(j,j)}$ matrix of the central sphere and use it to calculate modified scattering cross sections $\tilde{\sigma}_s$, $\tilde{\sigma}_a$, which now include the self-polarization effects that are due to the nearest neighbors. We can furthermore partially take configuration averaging into account by performing an orientation average of the modified cross sections $\langle\tilde{\sigma}_s\rangle_o$, $\langle\tilde{\sigma}_a\rangle_o$. Results for different densities are obtained by varying the interparticle distances. Our most severe approximation is then to insert the modified coherent scattering cross sections into the independent transport calculation of Eq. (52). Although this procedure avoids overcounting problems and can be justified for scattering events separated by a few wavelengths, it is only an approximation to the exact scattering calculations of neighboring spheres. More detailed transport considerations will be the subject of future studies, and here we simply show the predictions of our adopted approximation.

Using the above approximation techniques, we plot in Fig. 4 the effective absorption length from the multiple-scattering calculations as a function of the inverse volume fraction of coated spheres, the scale being modified such that close packing corresponds to $f_s = 1$. The results indicate that even at high filling fractions, the absorption path length is a near-linear function of $1/f_s$, presumably owing in part to the fact that the rather thick coatings create a natural separation of the strong scattering cores. Inspection of Fig. 4 leads one to predict that coherent-scattering effects can actually shorten the effective absorption length in the high-density limit. In the limit of close packing ($f_1 \approx 25\%$), the calculations indicate that l_a^{eff} could be more than five times shorter than that of a pure n_1 index material.

In order for other workers to compare their calculations

with ours and to show that coherent-scattering effects can influence absorption, we present orientation-fixed (Table 1) and orientation-averaged (Table 2) absorption cross section efficiencies, $Q_{\mathrm{abs}}^{(j)} \equiv \sigma_{\mathrm{abs}}^{(j)}/(\pi R_1^2)$, using the same optimal size parameters and coating thicknesses as above in a close-packing configuration where 12 spheres are in contact with the central sphere. In the orientation-fixed case, the electric field of the incident plane wave is taken to be plane polarized along the $\hat{\mathbf{y}}$ axis. The Cartesian coordinates of the centers of the spheres are specified in units where $R_1 = 1$. Results from use of different partial-wave cutoffs are shown. We also verified that the sum of the individual absorption cross sections is in agreement with far-field calculations of the total absorption cross sections [see Eqs. (39) and (40)].

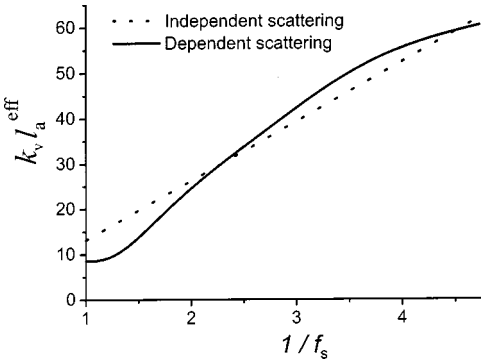

Fig. 4. Twelve nearest-neighbor coherent-scattering calculations of l_a^{eff} plotted as a function of the density $1/f_s$. The independent-scattering prediction is plotted as a dashed line for comparison.

We remark that partial-wave-space cutoffs of $n_{\max} = 3, 4, 5, 6$ yield respectively ~95%, 99%, 99.7%, 99.95% of the total $Q_{\mathrm{abs}}^{(j)}$. For comparison, we give in Table 2, the Q_{abs} of an isolated scatterer. Two things are worth noting. First, coherence effects do indeed increase the absorption of the individual scatterers, as the calculations in Fig. 4 suggest. Second, the coherence (evanescant-wave) effects in closely packed aggregates oblige us to invoke higher orders in the partial-wave developments than are necessary to describe isolated scatterers. Notably, the $n_{\max} = 3$ cutoff for isolated scatterers already gives more than 99% of the total Q_{abs}, whereas in the aggregate it yields only ~95% of the $Q_{\mathrm{abs}}^{(1)}$ of the central sphere. Although this effect remains relatively small (but nonnegligible) for most dielectric scatterers, it can become quite important for scatterers containing metallic layers.

8. CONCLUSION

We have presented recursive techniques and formulas permitting detailed, essentially exact calculations of absorption in finite systems composed of multiply coated spheres. We have also rapidly demonstrated the possible applications of such calculations in determining the optimum design parameters of dense macroscopic media composed of coated spheres. Such simulations can potentially economize both time and effort in the elaboration of coated-sphere heterogeneous materials. Besides the applications to dielectric coatings considered here, our tech-

Table 1. Individual Absorption Efficiencies $Q_{\mathrm{abs}}^{(j)}$ for a System of 13 Touching Coated Dielectric Spheres with $\chi_1 = 2.2$, $n_0 = 1.5$, $n_1 = 1.5 + 0.01i$, $n_2 = 2.5$, $R_1 - R_2 = 0.18R_2$, and an Electric Field Polarized along the $\hat{\mathbf{y}}$ Axis

Orientation-Fixed Absorption Efficiencies										
				$100 \times Q_{\mathrm{abs}}^{(j)}$						
Particle Positions				$n_{\max}$						
j	x_j	y_j	z_j	1	2	3	4	5	6	≥7
1	0	0	0	0.953	2.034	2.389	2.478	2.500	2.506	2.508
2,3,4,5	±0.8502	±0.5265	0	1.087	1.841	2.133	2.185	2.194	2.195	2.196
6,7	±0.5265	0	0.8502	0.645	2.249	2.630	2.667	2.671	2.672	2.673
8,9	±0.5265	0	−0.8502	1.284	2.261	2.484	2.510	2.522	2.524	2.525
10,11	0	±0.8502	0.5265	0.417	1.857	2.178	2.258	2.271	2.273	2.274
12,13	0	±0.8502	−0.5265	0.925	1.879	2.091	2.135	2.151	2.153	2.153

Table 2. Orientation-Averaged Absorption Efficiencies and Isolated Sphere Efficiencies for the Same Aggregate and Parameters as in Table 1

Orientation-Averaged Absorption Efficiencies							
	$100 \times \langle Q_{\mathrm{abs}}^{(j)} \rangle_o$						
Particles	$n_{\max}$						
j	1	2	3	4	5	6	≥7
Central sphere, $j = 1$	1.083	2.117	2.542	2.657	2.689	2.695	2.697
External spheres $j = 2,\ldots, 13$	0.898	2.091	2.359	2.412	2.423	2.425	2.425
Isolated sphere	0.853	2.019	2.223	2.243	2.244	2.244	2.244

niques can also be applied to systems of spheres with metallic coatings, thus opening the possibility of using these techniques in the numerous applications involving surface plasmon resonances. With the elaboration of new fabrication techniques of coated scatterers, we hope to be able to compare our predictions with experimental values.

APPENDIX A: SPHERICAL WAVES AND VECTOR SPHERICAL HARMONICS

The three normalized vector spherical harmonics used by us can be explicitly written in terms of the associated Legendre functions:

$$\begin{aligned}
\mathbf{Y}_{nm}(\hat{\mathbf{r}}) &= \gamma_{nm}\sqrt{n(n+1)}P_n^m(\cos\theta)\exp(im\phi)\hat{\mathbf{r}} \\
&= Y_{nm}(\theta,\phi)\hat{\mathbf{r}}, \\
\mathbf{X}_{nm}(\hat{\mathbf{r}}) &= \gamma_{nm}\left[-\frac{im}{\sin\theta}P_n^m(\cos\theta)\exp(im\phi)\hat{\theta}\right. \\
&\quad \left. + \frac{\mathrm{d}}{\mathrm{d}\theta}P_n^m(\cos\theta)\exp(im\phi)\hat{\phi}\right], \\
\mathbf{Z}_{nm}(\hat{\mathbf{r}}) &= \gamma_{nm}\left[\frac{\mathrm{d}}{\mathrm{d}\theta}P_n^m(\cos\theta)\exp(im\phi)\hat{\theta}\right. \\
&\quad \left. + \frac{im}{\sin\theta}P_n^m(\cos\theta)\exp(im\phi)\hat{\phi}\right], \\
\gamma_{nm} &= \left[\frac{(2n+1)(n-m)!}{4\pi n(n+1)(n+m)!}\right]^{1/2}. \qquad \text{(A1)}
\end{aligned}$$

The incident field coefficients for a homogeneous plane wave, $\mathbf{E}_\mathrm{i}(\mathbf{x}) = E\mathbf{e}_\mathrm{i}\exp(i\mathbf{k}_\mathrm{i}\cdot\mathbf{x})$, are given by

$$\begin{aligned}
[p]_{nm}^{\mathbf{M}} &= -i^n 4\pi\mathbf{X}_{nm}^*(\hat{\mathbf{k}}_\mathrm{i})\cdot\hat{\mathbf{e}}_\mathrm{i}, \\
[p]_{nm}^{\mathbf{N}} &= -i^{n+1}4\pi\mathbf{Z}_{nm}^*(\hat{\mathbf{k}}_\mathrm{i})\cdot\hat{\mathbf{e}}_\mathrm{i}. \qquad \text{(A2)}
\end{aligned}$$

APPENDIX B: VECTOR-WAVE ADDITION THEOREM

The addition theorem permits the transformation of partial waves centered on $\mathbf{x}$ (i.e., functions of $\mathbf{r}' \equiv \mathbf{r} - \mathbf{x}$) into partial waves centered at the origin.[22,23] Using the condensed notation introduced in Section 2, we may write the translation-addition theorem as[8]

$$\begin{aligned}
\Xi^t(k\mathbf{r}) &= \Xi^t(k\mathbf{r}')\beta(k\mathbf{r}_0), & r' > r_0, \\
\Xi^t(k\mathbf{r}) &= \mathcal{R}g\{\Xi^t(k\mathbf{r}')\}\alpha(k\mathbf{r}_0), & r' < r_0, \\
\mathcal{R}g\{\Xi^t(k\mathbf{r})\} &= \mathcal{R}g\{\Xi^t(k\mathbf{r}')\}\beta(k\mathbf{r}_0), & \forall|\mathbf{r}_j|, \qquad \text{(B1)}
\end{aligned}$$

where $\alpha(k\mathbf{r}_0)$ and $\beta(k\mathbf{r}_0)$ are respectively the irregular and the regular normalized translation-addition matrices. They have the form

$$\alpha(k\mathbf{r}_0) = \begin{bmatrix} \bar{A}(k\mathbf{r}_0) & \bar{B}(k\mathbf{r}_0) \\ \bar{B}(k\mathbf{r}_0) & \bar{A}(k\mathbf{r}_0) \end{bmatrix}, \qquad \beta(k\mathbf{r}_0) \equiv \mathcal{R}g\{\alpha(k\mathbf{r}_0)\}, \qquad \text{(B2)}$$

where the $\bar{A}_{\nu,\mu;n,m}$ and $\bar{B}_{\nu,\mu;n,m}$ matrix elements are those for normalized vector partial-wave functions.[1,9]

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