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CHEMICAL SYSTEMS LABORATORY TECHNICAL REPORT

ARCSL-SP-81008

MULTIPLE ELECTROMAGNETIC SCATTERING FROM A CLUSTER OF SPHERES

VOLUME I

THEORY

by

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September 1981

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A method to calculate the electromagnetic scattering properties of a cluster of spheres of arbitrary radii and possibly complex refractive indexes is proposed. The suggested approach takes into account multiple scattering effects and does not require any approximation except for the truncation of the multipolar expansions describing the scattered field.

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PREFACE

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CONTENTS

MULTIPLE ELECTROMAGNETIC SCATTERING FROM A CLUSTER OF SPHERES. I. THEORY

1. INTRODUCTION

Scattering of light by molecules is commonly dealt with through the Rayleigh-Debye theory.[1,2] This approach is known to be applicable to molecules whose effective index of refraction is close to unity and to imply a number of approximations which may have rather severe effects.[3-6] In this paper, we face the problem of scattering of electromagnetic waves by adapting to the method devised and successfully used by Johnson[7] to calculate the electronic states of large molecules.[8] Accordingly, we model a molecule as a cluster of spherical scatterers, possibly of different radii and complex refractive indexes. A plane electromagnetic wave, incident to the cluster undergoes multiple scatterings which we account for by expanding the scattered wave as a multicentered series of multipoles. The expansion coefficients turn out to be the solutions of the system of linear equations, obtained by expanding the incident field in a series of multipoles within the spheres and imposing the boundary conditions across the surface. Due to the presence of the incident plane wave, the above system is nonhomogeneous so that the expansion coefficients are uniquely determined.

The approach outlined above is of general applicability as it is not based on any assumption - neither on the radii and refractive indexes of the single spherical scatterers, nor on the geometry and size of the cluster. The only approximation required is the truncation of the multipolar expansions in order to get a system of finite size. The number of terms to be retained in the multipolar expansions in order to get a fairly convergent scattered field, as well as a number of related topics, will be discussed in section 5.

2 MULTIPOLAR EXPANSIONS OF THE FIELDS

The cluster whose scattering properties we want to study is composed of N nonmagnetic spheres whose centers lie at $\underline{R}_\alpha$ and whose radii and (possibly complex) refractive indexes are b_α and n_α, respectively. We refer the cluster to a fixed system of axes and choose the direction of incidence of the incoming plane wave through the direction cosines of its wavevector.

A straightforward calculation along the lines sketched by Jackson[9] allows writing the field of a circularly polarized plane wave of wave vector $\underset{\sim}{k}$ as

$$\underset{\sim}{E}_{\eta}^{(i)} = \sum_{LM} W_{\eta LM}(\hat{\underset{\sim}{k}})[j_L(kr)\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}) + \eta\frac{1}{k}\nabla\times j_L(kr)\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}})] \tag{1a}$$

$$i\underset{\sim}{B}_{\eta}^{(i)} = \eta\underset{\sim}{E}_{\eta}^{(i)} \tag{1b}$$

with $\eta = \pm 1$ according to the polarization and

$$W_{\eta LM}(\hat{\underset{\sim}{k}}) = 4\pi i^L(\underset{\sim}{e}_1 + i\eta\underset{\sim}{e}_2) \cdot \underset{\sim}{X}^{*}_{LM}(\hat{\underset{\sim}{k}}) \tag{2}$$

where $\underset{\sim}{e}_1$ and $\underset{\sim}{e}_2$ are unit vectors orthogonal to $\underset{\sim}{k}$ and to each other. The vector spherical harmonics, $\underset{\sim}{X}_{LM}$, are defined according to Jackson.[9]

The field scattered by the cluster is expanded in a multicentered series of multipoles including only outgoing spherical waves at infinity

$$\underset{\sim}{E}_{\eta}^{(s)} = \sum_{\alpha}\sum_{LM} [A^{\alpha}_{\eta LM}h_L(kr_{\alpha})\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_{\alpha}) + B^{\alpha}_{\eta LM}\frac{1}{k}\nabla\times h_L(kr_{\alpha})\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_{\alpha})] \tag{3a}$$

$$i\underset{\sim}{B}_{\eta}^{(s)} = \sum_{\alpha}\sum_{LM} [B^{\alpha}_{\eta LM}h_L(kr_{\alpha})\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_{\alpha}) + A^{\alpha}_{\eta LM}\frac{1}{k}\nabla\times h_L(kr_{\alpha})\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_{\alpha})] \tag{3b}$$

with $\underset{\sim}{r}_{\alpha} = \underset{\sim}{r} - \underset{\sim}{R}_{\alpha}$. The superscript (1) on the spherical Hankel functions of the first kind will be omitted throughout for simplicity.

As regards the field within the spheres, we remark that our theory can even be applied to a cluster of nonhomogeneous spheres, provided the n_{α}'s are spherically symmetric, i.e. $n_{\alpha} = n_{\alpha}(r_{\alpha})$. Therefore, within the α-th sphere, we write[10]

$$\underset{\sim}{E}_{\eta}^{(t)\alpha} = \sum_{LM} [C^{\alpha}_{\eta LM}R^{\alpha}_L(r_{\alpha})\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_{\alpha}) + \frac{1}{n_{\alpha}^2}D^{\alpha}_{\eta LM}\frac{1}{k}\nabla\times S^{\alpha}_L(r_{\alpha})\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_{\alpha})] \tag{4a}$$

$$i\underset{\sim}{B}_{\eta}^{(t)\alpha} = \sum_{LM} [D^{\alpha}_{\eta LM}S^{\alpha}_L(r_{\alpha})\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_{\alpha}) + C^{\alpha}_{\eta LM}\frac{1}{k}\nabla\times R^{\alpha}_L(r_{\alpha})\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_{\alpha})] \tag{4b}$$

where R^{α}_L and S^{α}_L are the solutions, regular at $r_{\alpha} = 0$, of the equations

$$\left[\frac{d^2}{dr_{\alpha}{}^2} - \frac{L(L+1)}{r_{\alpha}{}^2} + k^2n_{\alpha}^2\right](r_{\alpha}R^{\alpha}_L) = 0 \tag{5a}$$

and

$$\left[\frac{d^2}{dr_\alpha{}^2} - \frac{2}{r_\alpha n_\alpha}\frac{dn_\alpha}{dr_\alpha}\frac{d}{dr_\alpha} - \frac{L(L+1)}{r_\alpha{}^2} + k^2 n_\alpha^2\right](r_\alpha S_L^\alpha) = 0 \tag{5b}$$

respectively. Of course, for uniform n_α's, $R_L^\alpha \equiv S_L^\alpha \equiv j_L(K_\alpha r_\alpha)$, with $K_\alpha = kn_\alpha$.

3. EQUATIONS FOR THE COEFFICIENTS

The expansion coefficients $A^\alpha_{\eta LM}$, $B^\alpha_{\eta LM}$, $C^\alpha_{\eta LM}$, and $D^\alpha_{\eta LM}$ in equations (3) and (4) are uniquely determined by the boundary conditions for $\underset{\sim}{E}$ and $\underset{\sim}{B}$ at the surface of each of the spheres. Therefore, we need to rewrite equations (1) and (3) in terms of multipoles centered at a single site, say $\underset{\sim}{R}_\alpha$. This can be done by means of the appropriate addition theorems[11,12] which, near the surface of the α-th sphere, i.e. for $r_\alpha \le R_{\alpha\beta} = \left|\underset{\sim}{R}_\beta - \underset{\sim}{R}_\alpha\right|$, yield

$$\begin{aligned}\underset{\sim}{E}_\eta^{(s)} = \sum_{LM}\Big\{&A^\alpha_{\eta LM} h_L(kr_\alpha)\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_\alpha) + B^\alpha_{\eta LM}\frac{1}{k}\nabla\times h_L(kr_\alpha)\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_\alpha)\\
&+ \sum_\beta \sum_{L'M'}\big[A^\beta_{\eta LM}\big(H^{\alpha\beta}_{L'M'LM}\, j_{L'}(kr_\alpha)\underset{\sim}{X}_{L'M'}(\hat{\underset{\sim}{r}}_\alpha) + K^{\alpha\beta}_{L'M'LM}\frac{1}{k}\nabla\times j_{L'}(kr_\alpha)\underset{\sim}{X}_{L'M'}(\hat{\underset{\sim}{r}}_\alpha)\big]\\
&+ B^\beta_{\eta LM}\big(K^{\alpha\beta}_{L'M'LM}\, j_{L'}(kr_\alpha)\underset{\sim}{X}_{L'M'}(\hat{\underset{\sim}{r}}_\alpha) + H^{\alpha\beta}_{L'M'LM}\frac{1}{k}\nabla\times j_{L'}(kr_\alpha)\underset{\sim}{X}_{L'M'}(\hat{\underset{\sim}{r}}_\alpha)\big)\big]\Big\}\end{aligned} \tag{6}$$

and

$$\begin{aligned}\underset{\sim}{E}_\eta^{(i)} = \sum_{LM} W_{\eta LM}(\underset{\sim}{k})\Big\{&\sum_{L'M'}\big[J^\alpha_{L'M'\,LM} j_{L'}(kr_\alpha)\underset{\sim}{X}_{L'M'}(\hat{\underset{\sim}{r}}_\alpha) + L^\alpha_{L'M'\,LM}\frac{1}{k}\nabla\times j_{L'}(kr_\alpha)\underset{\sim}{X}_{L'M'}(\hat{\underset{\sim}{r}}_\alpha)\big]\\
&+ \eta\sum_{L'M'}\big[L^\alpha_{L'M'\,LM} j_{L'}(kr_\alpha)\underset{\sim}{X}_{L'M'}(\hat{\underset{\sim}{r}}_\alpha) + J^\alpha_{L'M'\,LM}\frac{1}{k}\nabla\times j_{L'}(kr_\alpha)\underset{\sim}{X}_{L'M'}(\hat{\underset{\sim}{r}}_\alpha)\big]\Big\}\end{aligned} \tag{7}$$

and analogous expressions for $i\underset{\sim}{B}_\eta^{(s)}$ and $i\underset{\sim}{B}_\eta^{(i)}$. In equation (3-1) we define

$$\begin{aligned}H^{\alpha\beta}_{L'M'LM} = (1-\delta_{\alpha\beta})\sum_\mu &C(1,L',L';-\mu,M'+\mu)4\pi\sum_\lambda i^{L'-L-\lambda} I_\lambda(L',M'+\mu;L,M+\mu)\\
&\times h_\lambda(kR_{\alpha\beta})Y^*_{\lambda M'-M}(\hat{\underset{\sim}{R}}_{\alpha\beta})C(1,L,L;-\mu,M+\mu)\end{aligned} \tag{8a}$$

and

$$K^{\alpha\beta}_{L'M'LM} = -i\sqrt{\frac{2L'+1}{L'}}\,(1-\delta_{\alpha\beta})\sum_{\mu} C(1,L',L'+1;-\mu,M'+\mu)4\pi\sum_{\lambda} i^{L'-L-\lambda+1}$$

$$\times\ I_{\lambda}(L'+1,M'+\mu;L,M+\mu)h_{\lambda}(kR_{\alpha\beta})Y^{*}_{\lambda M'-M}(\hat{R}_{\alpha\beta})C(1,L,L;-\mu,M+\mu) \tag{8b}$$

while in equation (7), $J^{\alpha}_{L'M'LM}$ and $L^{\alpha}_{L'M'LM}$ are identical to $H^{\alpha\beta}_{L'M'LM}$ and $K^{\alpha\beta}_{L'M'LM}$, respectively, but for the substitution of j_{λ} to h_{λ} and $R_{\beta} = 0$. The Clebsch-Gordan coefficients are defined according to Rose[13] and the quantities

$$I_{\lambda}(L'M';LM) = \int Y^{*}_{L'M'}\,Y_{LM}Y_{\lambda M'-M}d\Omega$$

are the well-known Gaunt integrals.[14]

Now we take the dot product of equations (4), (6), and (7) in turn with $\hat{r}_{\alpha}Y^{*}_{\ell m}(\hat{r}_{\alpha})$, $X^{*}_{\ell m}(\hat{r}_{\alpha})$, and $\hat{r}_{\alpha}\times X_{\ell m}(\hat{r}_{\alpha})$ and get the radial and tangential components of the field at the surface of the α-th sphere. Imposition of the boundary conditions and integration over the angles then yield, for each α, ℓ, m, six equations, among which $C^{\alpha}_{\eta LM}$ and $D^{\alpha}_{\eta LM}$, the coefficients of the internal field, can easily be eliminated. This possibility allows getting, for each α, ℓ, m, two equations involving only the A's and B's as unknowns

$$\sum_{\beta}\sum_{LM}\left\{(\delta_{\alpha\beta}\delta_{L\ell}\delta_{Mm}[R^{\beta}_{L}]^{-1} + H^{\alpha\beta}_{\ell mLM})A^{\beta}_{\eta LM} + K^{\alpha\beta}_{\ell mLM}B^{\beta}_{\eta LM}\right\}$$

$$= -\sum_{LM} W_{\eta LM}(\hat{k})P^{\alpha}_{\eta,\ell mLM} \tag{9a}$$

$$\sum_{\beta}\sum_{LM}\left\{(\delta_{\alpha\beta}\delta_{L\ell}\delta_{Mm}[S^{\beta}_{L}]^{-1} + H^{\alpha\beta}_{\ell mLM})B^{\beta}_{\eta LM} + K^{\alpha\beta}_{\ell mLM}A^{\beta}_{\eta LM}\right\}$$

$$= -\sum_{LM} W_{\eta LM}(\hat{k})Q^{\alpha}_{\eta,\ell mLM} \tag{9b}$$

where we define

$$\mathcal{R}_{\ell}^{\alpha} = \left[\frac{j_{\ell}(kr_{\alpha})\frac{d}{dr_{\alpha}}(r_{\alpha}R_{\ell}^{\alpha}) - R_{\ell}^{\alpha}\frac{d}{dr_{\alpha}}(r_{\alpha}j_{\ell}(kr_{\alpha}))}{h_{\ell}(kr_{\alpha})\frac{d}{dr_{\alpha}}(r_{\alpha}R_{\ell}^{\alpha}) - R_{\ell}^{\alpha}\frac{d}{dr_{\alpha}}(r_{\alpha}j_{\ell}(kr_{\alpha}))}\right]_{r_{\alpha}=b_{\alpha}} \tag{10a}$$

$$\mathcal{S}_{\ell}^{\alpha} = \left[\frac{j_{\ell}(kr_{\alpha})\frac{d}{dr_{\alpha}}(r_{\alpha}S_{\ell}^{\alpha}) - n_{\alpha}^{2}S_{\ell}^{\alpha}\frac{d}{dr_{\alpha}}(r_{\alpha}j_{\ell}(kr_{\alpha}))}{h_{\ell}(kr_{\alpha})\frac{d}{dr_{\alpha}}(r_{\alpha}S_{\ell}^{\alpha}) - n_{\alpha}^{2}S_{\ell}^{\alpha}\frac{d}{dr_{\alpha}}(r_{\alpha}h_{\ell}(kr_{\alpha}))}\right]_{r_{\alpha}=b_{\alpha}} \tag{10b}$$

$$P_{\eta,\ell mLM}^{\alpha} = \mathcal{J}_{\ell mLM}^{\alpha} + \eta\mathcal{L}_{\ell mLM}^{\alpha} \tag{11a}$$

$$Q_{\eta,\ell mLM}^{\alpha} = \mathcal{L}_{\ell mLM}^{\alpha} + \eta\mathcal{J}_{\ell mLM}^{\alpha} \tag{11b}$$

The system composed of equations (9a) and (9b) for all values of α, ℓ, m, completely solves our scattering problem.

4. THE CROSS SECTIONS

Once the coefficients of the scattered wave, $A_{\eta LM}^{\alpha}$ and $B_{\eta LM}^{\alpha}$, are known, all of the scattering properties of the cluster can be easily calculated. For this purpose, it is convenient to express the scattered field *in terms of multipoles centered at a single point, say* $\underset{\sim}{R}_{o}$, *through* the addition theorem already used in the preceding section. If $\underset{\sim}{r}_{o} = \underset{\sim}{r} - \underset{\sim}{R}_{o}$, then we have

$$\underset{\sim}{E}_{\eta}^{(s)} = \sum_{\alpha}\sum_{LM}\Bigg\{A_{\eta LM}^{\alpha}\sum_{L'M'}\left[\mathcal{J}_{L'M'\,LM}^{0\alpha}h_{L'}(kr_{0})\underset{\sim}{X}_{L'M'}(\hat{\underset{\sim}{r}}_{0}) + \mathcal{L}_{L'M'\,LM}^{0\alpha}\frac{1}{k}\nabla\times h_{L'}(kr_{0})\underset{\sim}{X}_{L'M'}(\hat{\underset{\sim}{r}}_{0})\right]$$

$$+ B_{\eta LM}^{\alpha}\sum_{L'M'}\left[\mathcal{L}_{L'M'LM}^{0\alpha}h_{L'}(kr_{0})\underset{\sim}{X}_{L'M'}(\hat{\underset{\sim}{r}}_{0}) + \mathcal{J}_{L'M'LM}^{0\alpha}\frac{1}{k}\nabla\times h_{L'}(kr_{0})\underset{\sim}{X}_{L'M'}(\hat{\underset{\sim}{r}}_{0})\right]\Bigg\}$$

$$= \sum_{L'M'}\left\{\tilde{A}_{\eta L'M'}\,h_{L'}(kr_{0})\underset{\sim}{X}_{L'M'}(\hat{\underset{\sim}{r}}_{0}) + \tilde{B}_{\eta L'M'}\frac{1}{k}\nabla\times h_{L'}(kr_{0})\underset{\sim}{X}_{L'M'}(\hat{\underset{\sim}{r}}_{0})\right\} \tag{12}$$

and an analogous expression for $i\underline{B}_{\eta}^{(s)}$, with

$$\tilde{A}_{\eta L'M'} = \sum_{\alpha}\sum_{LM}\left[A^{\alpha}_{\eta LM}\mathcal{J}^{0\alpha}_{L'M'LM} + B^{\alpha}_{\eta LM}\mathcal{L}^{0\alpha}_{L'M'LM}\right] \quad (13a)$$

$$\tilde{B}_{\eta L'M'} = \sum_{\alpha}\sum_{LM}\left[A^{\alpha}_{\eta LM}\mathcal{L}^{0\alpha}_{L'M'LM} + B^{\alpha}_{\eta LM}\mathcal{J}^{0\alpha}_{L'M'LM}\right] \quad (13b)$$

$\mathcal{J}^{0\alpha}_{L'M'LM}$ and $\mathcal{L}^{0\alpha}_{L'M'LM}$ are given by equations (8a) and (8b) with J_{λ} substituted for h_{λ}. Equation (12) is valid, provided that $r_o \geq R_{o\alpha} = \left|\underline{R}_{\alpha} - \underline{R}_o\right|$, i.e., in the region outside a sphere centered at $\underline{R}_o$ and including the whole cluster. Therefore, choosing $\underline{R}_o = o$ and thus letting the center of the expansion (12) coincide with the center of symmetry of the cluster, the volume of the aforementioned sphere is minimized. Anyway, the coefficients $\tilde{A}_{\eta L'M'}$ and $\tilde{B}_{\eta L'M'}$, unlike those of the field scattered by a single sphere, depend on the direction of the incident wavevector, $\underline{k}$. As a consequence, all of the quantities of interest depend both on $\underline{k}$ and on the scattered wavevector $\underline{k}_s = k\hat{r}$, except, of course, the scattering, absorption, and total cross sections, which depend only on $\underline{k}$. A straightforward calculation shows, in fact, that

$$\sigma_{\eta}^{(s)} = \frac{2\pi^2}{k^2}\sum_{L'M'}\left\{|\tilde{A}_{\eta L'M'}|^2 + |\tilde{B}_{\eta L'M'}|^2\right\} \quad (14a)$$

$$\sigma_{\eta}^{(abs)} = \frac{2\pi^2}{k^2}\sum_{L'M'}\left\{2|W_{\eta L'M'}|^2 - |\tilde{A}_{\eta L'M'} + W_{\eta L'M'}|^2 - |\tilde{B}_{\eta L'M'} + W_{\eta L'M'}|^2\right\} \quad (14b)$$

$$\sigma_{\eta}^{(tot)} = \frac{4\pi^2}{k^2}\sum_{L'M'} \mathrm{Re}\left\{W^{*}_{\eta L'M'}\,(\tilde{A}_{\eta L'M'} + \tilde{B}_{\eta L'M'})\right\} \quad (14c)$$

Finally, we notice that the cross sections depend on the polarization of the incident wave, η, as explicitly indicated in equation (14a) and (14b).

5. DISCUSSION

In order to assist in discussing both the physical content and the rate of convergence of the theory developed in the preceding sections, let us rewrite the system of equations (9a) and (9b) in matrix form:

$$\begin{vmatrix} \underset{\sim}{R}^{-1} + \underset{\sim}{H} & \underset{\sim}{K} \\ \underset{\sim}{K} & \underset{\sim}{S}^{-1} + \underset{\sim}{H} \end{vmatrix} \begin{vmatrix} \underset{\sim}{A} \\ \underset{\sim}{B} \end{vmatrix} = \begin{vmatrix} \underset{\sim}{P} \\ \underset{\sim}{Q} \end{vmatrix} \tag{15}$$

According to Waterman,[15] equation (15) defines the matrix on the left-hand side as the inverse of the electromagnetic T-matrix for the whole cluster. The above matrix is non-diagonal, for the cluster lacks the full spherical symmetry; whereas, the matrices within it have an interesting physical meaning of their own. The matrices $\underset{\sim}{R}$ and $\underset{\sim}{S}$ are, in fact, the direct sum of the diagonal matrices $\underset{\sim}{R}$ and $\underset{\sim}{S}$ which in turn form the electromagnetic T-matrix for the α-th sphere in the absence of any other scatterer. The presence of more than one scatterer in the cluster is accounted for not only by the matrices $\underset{\sim}{R}^{\beta}$ and $\underset{\sim}{S}^{\beta}$, with $\beta \neq \alpha$, but also by the matrices $\underset{\sim}{H}$ and $\underset{\sim}{K}$ which couple all the scatterers to each other. The elements $H^{\alpha\beta}_{\ell m LM}$ and $K^{\alpha\beta}_{\ell m LM}$ are shown in the appendix to be the matrix elements, in the site and angular momentum representation, of the free space dyadic Green's function. As the above quantities appear as the coefficients of the addition theorem we used in section 4, this latter, besides being a useful mathematical tool, describes the propagation to the site α of the spherical vector waves scattered by the site β. Therefore, our previous statement that multiple scatterings are accounted for by expanding the wave scattered by the whole cluster in a multicentered series of multipoles remains fully justified.

The theory discussed thus far is based on general grounds and requires no approximation, apart from the truncation of the multipolar expansions. In this connection, a fairly good rate of convergence is expected even when the cluster is not small in comparison to the incident wavelength provided $kb_\alpha << 1$ for any α. Indeed R^{α}_{ℓ} and S^{α}_{ℓ} are known to decrease rapidly with increasing ℓ so that, for small kb_α, R^{α}_{1} and S^{α}_{1} are quite sufficient to describe the scattered wave even when n_α is not close to unity.[16,17] Thus the rate of convergence of our approach depends upon the behavior of $H^{\alpha\beta}_{\ell m LM}$ and $K^{\alpha\beta}_{\ell m LM}$. According to their definitions, the order of magnitude of equations (8a) and (8b) is determined by the Gaunt integrals, I_λ, and by the spherical Hankel functions, $h_\lambda(kR_{\alpha\beta})$. The I_λ integrals do not vanish when $|\ell - L| \leq \lambda \leq \ell + L$, but decrease very rapidly with increasing λ.[18] Thus, although the imaginary part of h_λ, $n_\lambda(kR_{\alpha\beta})$, tends to increase when $\lambda > kR_{\alpha\beta}$, the eventual effect is to decrease the magnitude both of $H^{\alpha\beta}_{\ell m LM}$ and of $K^{\alpha\beta}_{\ell m LM}$ with increasing ℓ, L and $R_{\alpha\beta}$. This behavior was to be expected, for when the intersphere distance increases, the present theory should reduce to that of the scattering from N spherical scatterers without any multiple scattering effect. As a consequence, it is reasonable to expect that the present approach converges well by truncating the multipolar expansions at $L_M = 3$. Since the order of the system (15) is $d_M = 2N(L_M + 1)^2 - 2N$, we should have $d_3 = 30N$, a rather high number even for small clusters. However, if our clusters possess symmetry properties, as is the case for actual molecules, we can use group theory to get the system (15) in factorized form. The application of group theory to the present approach to multiple electromagnetic scattering will be the subject of another paper.

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APPENDIX

MATRIX ELEMENTS OF THE DYADIC GREEN'S FUNCTION

The free space propagator for spherical vector waves (dyadic Green's function) is the solution of the inhomogeneous Helmholtz equation.

$$(\nabla^2 + k^2)\underset{\sim}{G}(\underset{\sim}{r},\underset{\sim}{r}') = -4\pi\underset{\sim}{1}\delta(\underset{\sim}{r}-\underset{\sim}{r}') \tag{A-1}$$

in spherical coordinates and can be written with respect to the molecular sites as

$$\underset{\sim}{G}(\underset{\sim}{r},\underset{\sim}{r}') = \frac{e^{ik|\underset{\sim}{r}_\alpha-\underset{\sim}{r}'_\beta-\underset{\sim}{R}_{\alpha\beta}|}}{|\underset{\sim}{r}_\alpha \underset{\sim}{r}'_\beta-\underset{\sim}{R}_{\alpha\beta}|}\underset{\sim}{1} \tag{A-2}$$

If we expand the unit dyadic, $\underset{\sim}{1}$, with respect to a spherical basis*

$$\underset{\sim}{1} = \sum_\mu(-)^\mu \underset{\sim}{\xi}_\mu \underset{\sim}{\xi}_{-\mu} = \sum_\mu \underset{\sim}{\xi}^*_{-\mu}\underset{\sim}{\xi}_{-\mu} \tag{A-3}$$

and assume $\left|\underset{\sim}{r}_\alpha - \underset{\sim}{R}_{\alpha\beta}\right| \geq \underset{\sim}{r}'_\beta$, the Neuman expansion of $\underset{\sim}{G}$ is**

$$\underset{\sim}{G}(\underset{\sim}{r}_\alpha,\underset{\sim}{r}'_\beta) = 4\pi ik\sum_\mu\sum_{LM} h_L(k|\underset{\sim}{r}_\alpha-\underset{\sim}{R}_{\alpha\beta}|)Y_{LM}(\underset{\sim}{r}_\alpha-\underset{\sim}{R}_{\alpha\beta})j_L(kr'_\beta)Y^*_{LM}(\hat{\underset{\sim}{r}}'_\beta)\underset{\sim}{\xi}^*_{-\mu}\underset{\sim}{\xi}_{-\mu}$$

Now the addition theorem for scalar Helmholtz harmonics can be applied to $h_L(k\left|\underset{\sim}{r}_\alpha - \underset{\sim}{R}_{\alpha\beta}\right|)Y_{LM}(\underset{\sim}{r}_\alpha - \underset{\sim}{R}_{\alpha\beta})$ to get***

$$\underset{\sim}{G}(\underset{\sim}{r}_\alpha,\underset{\sim}{r}'_\beta) = \sum_\mu\sum_{LM}\sum_{L'M'} j_L(kr'_\beta)Y^*_{LM}(\hat{\underset{\sim}{r}}'_\beta)\underset{\sim}{\xi}^*_{-\mu}G_{L'M'LM}(\underset{\sim}{R}_{\alpha\beta})$$

$$\times\, j_{L'}(kr_\alpha)Y_{L'M'}(\hat{\underset{\sim}{r}}_\alpha)\underset{\sim}{\xi}_{-\mu} \tag{A-4}$$

where we assumed $r_\alpha < R_{\alpha\beta}$ and thus define

$$G_{L'M'\,LM}(\underset{\sim}{R}_{\alpha\beta}) = 4\pi ik\sum_\lambda i^{L'-L-\lambda}I_\lambda(L'M';LM)h_\lambda(kR_{\alpha\beta})Y^*_{\lambda M'-M}(\hat{\underset{\sim}{R}}_{\alpha\beta})$$

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*Rose, E.M. Multiple Fields. John Wiley & Sons, Inc., New York, New York. 1955.

**Goertzel G. and Tralli, N. Some Mathematical Methods of Physics. McGraw-Hill, New York. 1960.

***Nozawa, R. J. Math. Phys. 7 1841 (1966).

Now we recall that the spherical harmonics and the irreducible spherical tensors are related through the equation*

$$\xi_{-\mu} Y_{LM}(\hat{r}) = \sum_{J} C(1,L,J;-\mu,M) T^{M}_{JL}(\hat{r})$$

so that equation (A-4) can be rewritten as

$$G(r_\alpha, r'_\beta) = \sum_{\mu} \sum_{JJ'} \sum_{LM} \sum_{L'M'} j_L(kr'_\beta) T^{M-\mu^*}_{JL}(\hat{r}'_\beta) C(1,L,J;-\mu,M)$$

$$\times\, G_{L'M',\,LM}(R_{\alpha\beta}) C(1,L',J';-\mu,M') j_{L'}(kr_\alpha) T^{M'-\mu}_{J'L'}(\hat{r}_\alpha)$$

which, through the position $M - \mu = m$, $M' - \mu = m'$ takes the final form

$$G(r_\alpha, r'_\beta) = \sum_{Jm} \sum_{J'm'} \sum_{LL'} j_L(kr'_\beta) T^{m^*}_{JL}(r'_\beta) \mathcal{G}^{m'm}_{J'L',\,JL}(R_{\alpha\beta}) j_{L'}(kr_\alpha) T^{m'}_{J'L'}(\hat{r}_\alpha) \quad \text{(A-5)}$$

Equation (A-5) shows that the quantities

$$\mathcal{G}^{m'm}_{J'L',\,JL}(R_{\alpha\beta}) = \sum_{\mu} C(1,L,J;-\mu,m+\mu) G_{L'm'+\mu,Lm+\mu}(R_{\alpha\beta}) C(1,L',J';-\mu,m'+\mu) \quad \text{(A-6)}$$

are just the matrix elements of G with respect to the irreducible spherical tensors. Moreover, direct comparison of equation (A-6) with equations (8a) and (8b) shows that

$$H^{\alpha\beta}_{L'M'\,LM} = -\frac{i}{k} \mathcal{G}^{M'M}_{L'L',\,LL}(R_{\alpha\beta}) \quad \text{(A-7a)}$$

$$K^{\alpha\beta}_{L'M'LM} = -\frac{1}{k}\sqrt{\frac{2L'+1}{L'}}\, \mathcal{G}^{M'M}_{L'L'+1,LL}(R_{\alpha\beta}) = \frac{1}{k}\sqrt{\frac{2L'+1}{L'+1}}\, \mathcal{G}^{M'M}_{L'L'-1,LL}(R_{\alpha\beta}) \quad \text{(A-7b)}$$

Now, since

$$j_L(kr) X_{LM}(\hat{r}) = M_{LM}(r) = -j_L(kr) T^{M}_{LL}(\hat{r})$$

$$\frac{1}{k}\nabla \times j_L(kr) X_{LM}(\hat{r}) = N_{LM}(r) = i\left[\sqrt{\frac{L}{2L+1}}\, j_{L+1} T^{M}_{LL+1} - \sqrt{\frac{L+1}{2L+1}}\, j_{L-1} T^{M}_{LL-1}\right]$$

an easy but lengthy calculation, with the help of the formulas of Borghese et al.,* shows that H and K are the matrix elements of G with respect to M

*Rose, E.M. op cit.
**Borghese, F., Denti, P., Toscano, G., and Sindoni, O. I. J. Math. Phys. 21, 2754 (1980).

and $\underset{\sim}{N}$. The other matrix elements of $\underset{\sim}{G}$ do not appear in the present work because we deal with solenoidal fields which require $\underset{\sim}{M}$ and $\underset{\sim}{N}$ only for their description. Finally, we notice that the above procedure also allows the definition of $J^{\alpha\beta}_{L'M' LM}$ and $L^{\alpha\beta}_{L'M' LM}$ as the matrix elements of $\underset{\sim}{G}$ with respect to $\underset{\sim}{M}$ and $\underset{\sim}{N}$. It is, in fact, sufficient to assume $r_\alpha > \underset{\sim}{R}_{\alpha\beta}$ and consequently substitute in G, $j_\lambda(kR_{\alpha\beta})$ to $h_\lambda(kR_{\alpha\beta})$ and in $\underset{\sim}{M}$ and $\underset{\sim}{N}$, $h_{L'}(kr_\alpha)$ to $j_{L'}(kr_\alpha)$.

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