Wavelength-dependent refractive and absorptive terms for propagation in small-spaced correlated distributions

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Earlier results for coherent propagation of light in correlated random distributions of dielectric particles of radius a (with minimum separation b ≥ 2a small compared with wavelength λ = 2π/k) are generalized to obtain the refractive and absorptive terms to order (ka)². The present results include the earlier multiple scattering by electric dipoles as well as scattering and multipole coupling by magnetic dipoles and electric quadrupoles. The correlation aspects are determined by the statistical-mechanics radial distribution function f(R) for impenetrable particles of diameter b. The new terms for slab scatterers and spheres involve the integral of |R| (first moment) or of f ln R for cylinders. The new packing factor is evaluated exactly for slabs as a simple algebraic function of the volume fraction ρ, and it is shown that the bulk index of refraction reduces to that of one particle in the limit ρ → 1. A similar result is achieved for spheres in terms of the Percus–Yevick approximation and the unrealizable limit ρ = 1.

INTRODUCTION

Earlier papers1–4 developed simple forms for the coherent bulk index of refraction (n = χ) for correlated random distributions of dielectric particles with minimum separation (b) of centers small compared with wavelength (λ = k/2π). Writing the index as n = n_r + in_i + in_e, we applied general scattering theory1–4 to the range of small kb to obtain results for the refractive (n_r) and absorptive (n_a) terms that were explicitly independent of λ and to obtain corresponding results for the scattering (n_s) loss term to lowest order in λ. The explicit approximations2 for n_s and n_a for spheres, cylinders, and slabs (m = 3, 2, 1, respectively) depend only on the particles' radius or half-width (a), their complex index of refraction (n' = χ + iχ'), and their average number (ρ) per unit volume; they exhibited the statistical aspect of the problem only in the volume fraction w = ρv, with v = v(a) as the volume of one particle. The corresponding scattering terms n_e were additionally dependent on (ka)² and on the low-frequency limit of the structure factor W/W, with W = 2v(b/2) = ω(b/2a)² as the volume fraction of impenetrable statistical particles with diameter b ≥ 2a, i.e., in general, each dielectric particle was visualized as having a transparent coating of thickness (b/2) – a. The present paper applies the general theory5–8 to derive the leading λ-dependent terms of n_s and n_a; these depend explicitly on (ka)² and a/b for all cases and on appropriate correlation integrals N(w).

The correlation aspects of the distribution that we consider are determined by the statistical-mechanics radial distribution function6 f(R) for impenetrable particles and are exhibited explicitly as simple integrals over all R of the total correlation function F = f – 1. The integrals for spherical and slab particles are of the form FRdR (moments of F), but cylindrical particles also involve FF ln R dR. These can all be evaluated numerically from existing statistical-mechanics results or approximations6–10 for f. We obtained explicit closed-form approximations before11–17 for the integrals that arise in the W set and also used the required N integral for spheres in a related development16 for large kb; for slabs, we obtain both the W and N integrals from our earlier Laplace transformation11 of the exact Zernike–Prins result13 for f. For cylinders, we may use the virial expansion for f to consider some of the properties of N.

In the following, for brevity, we use, for example, form (4: 113) to indicate Eq. (113) of Ref. 4, as well as essentially the same notation as before.1–4 We generalize the earlier multiple-scattering electric-dipole approximations for n² = χ given collectively in form (1:44) by including scattering (and multipole coupling) by magnetic dipoles and electric quadrupoles (for spheres and cylinders). For slabs and b = 2a (minimum separation of slab centers equal to slab thickness), the explicit approximation for n² = χ reduces to ε - χ if w → 1, as required from physical considerations: The particles occupy all space. The limit ε → 1 is not realizable for identical spheres, and we might take w ≤ w_d = 0.63, with w_d as the densest random packing introduced earlier11,17 to define the amorphous solid. However, our explicit approximation for ε for spheres also reduces to ε - χ as w reduces to 1; we regard this as consistent with the approximations involved in scaled-particle5 and Percus–Yevick5–7 statistical-mechanics theory and with the closure approximation used in the multiple-scattering theory.3,4 For cylinders, we take w ≤ w_d = 0.84, as before.1 Were an analogous closed form available for the N-integral for this case, we would expect the corresponding approximation for ε to show the same behavior for the unrealizable limit w → 1.

The present application of the general theory3,4 to larger kb than before1–4 plus the recent applications18 to large kb provide simple forms that explicitly display the functional dependence on all key parameters for many practical applications. Thus, in these ranges of kb, elaborate machine computations are no longer required, and the results help to delineate the fundamental physical processes.

PRELIMINARY CONSIDERATIONS

For a slab-region distribution and a normally incident wave φ = e^iωt (representing either the electric or the magnetic component) we write
\[\phi = \hat{e} e^{i k \hat{z}}, \quad \hat{e} \cdot \hat{z} = 0, \quad k = 2\pi / \lambda = 2\pi n_\gamma / \lambda,\]

with \(n_\gamma\) as the index of the embedding medium. The corresponding bulk coherent propagation constant

\[K = k n_\gamma / n_\gamma = k n_\gamma, \quad \eta^2 = \epsilon\]

is to be expressed in terms of \(\rho\) and \(F\) for pair-correlated particles specified by their isolated scattering amplitudes \(g(f, \hat{z})\). The normalization for \(g\) is such that for lossless particles

\[-\text{Re} \hat{e} \cdot g(\hat{z}) = -\text{Re} g = |\mathcal{H}| |g(f, \hat{z})|^2, (3)\]

with \(\mathcal{H}\) as the mean over all directions of observation \(f\). The corresponding known\(^{19}\) scattering coefficients \(a_n\) (which may represent two sets) are normalized by the form

\[g = \sum a_n, \quad a_n = a_n(\epsilon', x), \quad \epsilon' = \epsilon_p / \epsilon_n, \quad x = \kappa a\]

In addition to the dependence on \(\epsilon'\) (the relative dielectric parameter) and \(x\) (the normalized radius or half-width), the coefficients depend on the dimensionality of the problem and on the choice of field component for \(m = 2\) (i.e., on whether the electric polarization is lateral or transverse to the cylinder's axis). We obtain results for the bulk relative dielectric parameter \(\epsilon'\) in the form

\[\epsilon = \epsilon_1 + \epsilon_2 + i\epsilon_3 = \epsilon_1 + \chi^2 \epsilon_2 + \chi n \epsilon_3,\]

where the set \(\epsilon\) is independent of \(x = ka\). The forms for \(\epsilon_1\) and \(\epsilon_2\), corresponding to multiple scattering by electric dipoles, were discussed before\(^{22}\) in detail. Now we obtain \(\epsilon_3\).

From Rayleigh’s results for spherical dipoles,\(^{20}\) the first approximation for sparse uncorrelated distributions corresponds to

\[\eta^2 - 1 = -cG, \quad \eta^2 = \epsilon,\]

with \(G\) as a multiple-scattering amplitude. This form with \(G = g\) was obtained originally by Reiche\(^{23}\) and by Foldy\(^{24}\) for spherical cases, and Lax\(^{25}\) derived the form in terms of a more general amplitude than \(g\). The function \(G\) that we require is discussed in detail in Refs. 3 and 4. In particular for spheres, cylinders, and slabs, respectively, the systems of algebraic equations (4:113), (3:92), and (3:179) determine \(\epsilon = \eta^2\) functionally in terms of \(a_n\) and \(F\) for arbitrary \(\epsilon'\) and \(ka\). Before we kept only the electric-dipole coefficient \(a_1\) in these systems and considered only the leading terms of their imaginary and real parts, of order \(x^2\) and \(x^2 m\), respectively, for lossless scatterers. Now we include terms to order \(x^{n+2}\) for the refractive and absorptive effects.

Thus, for spheres with \(x\) small, the electric dipole approximates\(^{19}\)

\[a_1 = i x^2 (\epsilon' - 1) - x^2 (\epsilon' - 1)^2 / 4 - x^4 (\epsilon' - 1)^2 / 16,\]

where the next terms are of order \(ix^6\) and \(x^8\). The corresponding magnetic dipole \(\eta_1 M\) and electric quadrupole are, respectively,

\[a_{1M} = i x^2 (\epsilon' - 1) / 30, \quad a_2 = i x^4 (\epsilon' - 1) / (6(2\epsilon' + 3)),\]

where the next terms are proportional to \(ix^7\) and \(x^9\), and \(x^{10}\). The resulting amplitudes for Rayleigh’s approximation (6) is\(^{19}\)

\[g = a_1 + a_{1M} + a_2. (We use \(a_n, a_{1M}\) for the earlier b_n, c_1.)\]

For cylinders\(^{19}\) and polarization transverse to the axis, we have \(g = a_0 + a_1 + a_2\) with dominant dipole

\[a_1 = i x^2 (\epsilon' - 1) - x^2 (\epsilon' - 1)^2 / 8 \quad \text{and} \quad a_0 = -x^2 (\epsilon' - 1) / (16(\epsilon' + 1)),\]

where the next terms are of order \(ix^6\) and \(x^8\). For cylinders and polarization along the axis, we use \(g = a_0 + a_1\), with

\[a_0 = i x^2 (\epsilon' - 1) / 4 \quad \text{and} \quad a_1 = -x^2 (\epsilon' - 1)^2 / 16,\]

where the next terms are of order \(ix^6\) and \(x^8\). In addition, we keep

\[a_1 = i x^2 (\epsilon' - 1) / 16,\]

and ignore \(ix^6, ix^8, \) and \(x^8\) terms.

For normal incidence on slabs, \(g = a_0 + a_1\), as discussed for (3:179). The dominant term is

\[a_0 = i x (\epsilon' - 1) - x^2 (\epsilon' - 1) / 3 - x^4 (\epsilon' - 1)^2,\]

where the next terms are of order \(ix^5\) and \(x^6\). We also retain

\[a_1 = i x^4 (\epsilon' - 1) / 3,\]

but not \(ix^5, ix^7, \) and \(x^6\) terms.

**DISTRIBUTION OF SLABS**

From form (3:177) we have

\[G = A_0 + A_1, \quad c = i 2\rho / k = i u / x, \quad c = \rho 2\pi = \rho e(a).\]

where

\[A_0 = a_0(1 + A_0 / H_0 + A_1 / H_1 / \eta), \quad A_1 = \eta_1 a_1(1 + A_0 / H_1 / \eta + A_1 / H_{11} / \eta).\]
where the next terms are \(O(x^2)\). We obtain form (20) to \(O(x^2)\), from which

\[
\epsilon = 1 + u - x^2u^2/2 - 2u + 4M/8 + i\pi x^2u^2W/4.
\]

\[
\delta = \epsilon' - 1.
\]  

(26)

Here

\[
W = 1 + 2\pi p \int F_{rdR} + 2\pi pF_{1} = 1 - 8WF_{1},
\]

\[
W = \pi(p/2)^2 + \rho(c/2) = u(b/2)^2.
\]  

(27)

and

\[
M = L - \pi N/2
\]

\[
= \ln(b/a) + W\ln(2/c'kb) - 2\pi p \int F\ln(R/b)RdR
\]

\[
= \ln(b/a) + W\ln(b) + 8WF_{1},
\]

\[
F_{1} = \int_{0}^{\infty} F(\ln u)u du.
\]  

(28)

with \(F(R) = F(R/b) = F[u] + L_{0} = \ln(2/c'kb)\).

We may evaluate \(F_{1}\) and \(F_{1}\) numerically by using tabulated values of \(f\) or the original integral-equation approximations in the computing routine. To first order in \(W\), we use the virial expansion: \(F = -1\) for \(u < 1\).

\[
F = \frac{8W}{\pi} \left[ \cos^{-1} \left( \frac{u}{2} \right) \right]^{-1} \left( \frac{1 - (u/2)^{2}}{2} \right)^{1/2}, 1 \leq u \leq 2,
\]  

(29)

and \(F = 0\) for \(u > 2\). A closed-form approximation of \(W^\prime\) (and consequently of \(F_{1}\)) was derived earlier by differentiating the scaled-particle equation of state. Thus we obtained

\[
W = (1 - W)^3/(1 + W),
\]  

(30)

from which \(W^\prime = 1 - 4W + 7W^2 + \ldots\). The rigorous virial expansion to \(O(W^3)\) is

\[
W = 1 - 4W + \sqrt{12}W^2/\pi \approx 1 - 4W + 6.6159W^2.
\]  

(30')

For the unrealizable value \(W = 1\), the closed-form \(W^\prime\) vanishes; a comparable approximation of \(M\) would reduce to \(-3\) for \(b = 2a\) and \(W = 1\) in order for \(\epsilon\) to equal \(\epsilon'\). The corresponding moments are then \(F_{1} = 1/8\) and \(F_{2} = -(\ln 2)/2\).

For polarization transverse to the axes we use \(G = A_{0} + A_{1} + A_{2}\) with \(A_{0}\) satisfying the system (3.89). From the solution (3.90) and the corresponding form of \(n_{2}^{0}\), in terms of \(a_{n}\) and \(H_{n}\) of form (3.91), to the accuracy required for present purposes, we work with

\[
-(\epsilon - 1)/c = a_{n} + \epsilon a_{1} + \epsilon a_{2}(1 + \epsilon a_{2}/2)^{1/2};
\]

\[
A_{1} = a_{1}/(1 - a_{1}H_{1}),
\]

\[
2H_{1} = c + (W - 1) + i\pi - i(\epsilon W - 1)/2\pi.
\]  

where \(\epsilon a_{1}\) is a multiple-scattering coefficient that includes all electric-dipole-dipole coupling. The coefficient \(a_{0}\) (essentially the magnetic dipole) is uncoupled, and the multiplier of the electric quadrupole \(a_{2}\) includes all orders of electric-dipole coupling to the required accuracy.

Using approximations (10) and (11), we obtain initially
\[ \epsilon_1 = 1 + \frac{2w \epsilon' - 1}{1 + w' - w (\epsilon' - 1)} = 1 + \frac{w \delta}{D}, \]

\[ D = 1 + \frac{(1 - w) \delta}{2}, \quad \delta = \epsilon' - 1. \quad (32) \]

with which

\[ \epsilon = \frac{\epsilon_1 - \epsilon_2 \delta}{\epsilon_1 + \epsilon_2 \delta} \left[ 3 + \epsilon' - \delta M (\epsilon' - 1) \right] \]

\[ \epsilon_2 = \frac{\epsilon_1 (\epsilon_1 - 1)}{2 \epsilon' + 1} \]

\[ \epsilon = \epsilon_1 - \frac{\epsilon_1 (\epsilon_1 - 1)}{2 \epsilon' + 1} + \frac{ix \delta^2 w W}{8}. \quad (33) \]

For \( b = 2a \) and the unrealistic value \( w = 1 \), we have \( D = 1 \) and \( \epsilon_1 = \epsilon' \) and the result for \( W = 0 \) and \( M = -\frac{1}{2} \) is again \( \epsilon = \epsilon' \).

For comparison with (26), say, \( \epsilon_1 \), we have to \( 0(\delta^2) \) for the present \( \epsilon \).

\[ \epsilon' \approx 1 + w \delta - w(1 - w) \delta^2/2 + x^2 \delta^2/(1 + 2w + 4M + W)/16 + ix \delta^2 w W /8. \quad (34) \]

The corresponding bipolarization is

\[ \epsilon_i - \epsilon = w(1 - w) \delta^2/2 + x^2 \delta^2/(1 + 2w + 4M - W)/16 + ix \delta^2 w W /8. \quad (35) \]

and we may obtain higher-order terms in \( \delta \) from Eq. (33).

**DISTRIBUTION OF SPHERES**

For spheres we use form (7) with

\[ G = A_1 + A_M + A_2, \quad c = i 4 \pi p/k^3 = 3 \pi e / x^3, \]

\[ w = \rho 4 \pi a^3 / 3 = \rho e (a), \quad (36) \]

with the \( A \)'s satisfying the system (4:113) in terms of the isolated coefficients \( a_n \) and the correlation integrals \( h_n \) of form (4:80) or (3:148). Introducing the low-frequency forms (3:149), we solve the system and obtain

\[ -(q^2 - 1)/c = q^2 A_1 + q^2 A_M [1 + A_4 (e'/c + 1/2)] + q^2 A_2 (1 + A_4 (3/5)) \]

\[ A_1 = a / (1 - a / A_4), \]

\[ 3 A_4 = 2e + 2A_1 + A_2 = 2e + 2(1 - W) + i N(2 + \eta^2)/x. \quad (37) \]

\[ W = 1 + 4 \pi p \int_0^\infty FR^2 dR = 1 + 4 \pi p F_2 = 1 - 24 W F_2, \]

\[ W = \rho 4 \pi (b^2 / 2)^{3/3} = \rho e (b^2 / 2) = w (b / 2a)^3, \quad (38) \]

and

\[ N = -4 \pi \rho a \int_0^\infty FR dR = -4 \pi \rho a F_1 = 24 (a / b) W F_1. \quad (39) \]

Here \( q^2 A_1 / q_1 \) includes all electric-dipole–dipole coupling, and the multipliers of the magnetic dipole \( A_M \) and the electric quadrupole \( q_2 \) incorporate multiple coupling with all orders of electric dipoles to the required accuracy.

Using approximations (12) and (14), we obtain initially

\[ \epsilon_1 = 1 + \frac{3w (\epsilon' - 1)}{2 + \epsilon' - w(\epsilon' - 1)} = 1 + \frac{w \delta}{D}, \]

\[ D = 1 - \frac{(1 - w) \delta}{3}, \quad \delta = \epsilon' - 1. \quad (40) \]

with which

\[ \epsilon = \epsilon_1 - x^2 \delta \left[ \frac{1}{2 \epsilon' + 1} \frac{2 - \epsilon'}{5} + \frac{N}{9} \frac{2 - \epsilon'}{5} \right] \]

\[ - \frac{\epsilon_1}{10} \left[ 1 + \frac{(2 \epsilon_1 + 3)^2}{5(2 \epsilon_1 + 3)} \right] + \frac{i x^2 \delta^2 w W}{9}. \quad (41) \]

The magnetic-dipole contribution \( x^2 \delta e_1 = a_1 M e_1 \) shows that the effects of the function in brackets in Eq. (37) have canceled.

By differentiating the scaled-particle approximation for the equation of state, we showed that

\[ W = (1 - W)^4/(1 + 2W)^2, \quad (42) \]

which also follows from the Percus–Yevick approximation. The first moment \( F_1 \) obtained from the Wertheim–Thiele solution of the Percus–Yevick integral equation gives the closed form

\[ N = 2a / b + 2W [1 - W / 5 + W^2 / 10]. \quad (43) \]

Although the physically realizable range corresponds to \( W \leq W_d \approx 0.63 \), we see that, for \( b = 2a \) and the unrealistic value \( w = 1 \), it follows that \( W = 0 \) and \( N = 9/5 \), then \( \epsilon = \epsilon' \) as was discussed for slabs and cylinders.

For comparison with forms (21), (26), and (34), we have to \( 0(\delta^2) \) for the present case of the sphere

\[ \epsilon' \approx 1 + w \delta - w(1 - w) \delta^2/2 + x^2 w \delta^2/[6 + 3w - 5N]11/(15)^2 + i x^2 \delta^2 w W /9. \quad (44) \]

For \( b = 2a \), the function in brackets reduces to

\[ [ \frac{1}{x} ] = 3(2 - 5w + 4w^2 - w^3)/(1 + 2a). \quad (44') \]

**BULK INDEX OF REFRACTION**

We write forms (21), (26), (33), and (41) collectively as

\[ \epsilon = \epsilon_1 + w \delta^2 (P(x^2) + i\epsilon \delta^2 S \infty), \quad \delta = \epsilon' - 1. \quad (45) \]

where \( P \) and \( S \), proportioned to \( x^2 \) and \( x^m \), are obtained by \( \epsilon \)–\( \delta \) correlation. The \( k \)-independent term

\[ \epsilon_1 = 1 + w \delta/(1 + \delta D), \quad D = (1 - w) Q, \]

\[ Q_1 = Q_2 = 0, \quad Q_2 = 1/2, \quad Q_3 = 1/3 \quad (46) \]

represents special cases of the result for ellipsoids. The corresponding \( S \) for ellipsoids is also known, and form (45) holds for all small dielectric particles (discounting a resonant multipole).

The corresponding bulk index of refraction may be written

\[ \eta = [\epsilon_1 + w \delta^2 (P + i S)]^{1/2} = \eta_1 + w \delta^2 (P + i S) / 2 \eta_1, \quad (47) \]

with \( \eta_1 = \sqrt{\epsilon_1} \). More generally, we write \( \epsilon = \epsilon_1 + i \epsilon_2 \) and \( \eta = \eta_1 + i \eta_2 \),

\[ \eta_2 = \left| \frac{\epsilon_1 + \epsilon_2}{2} \right|^{1/2}, \quad | \epsilon_2 | = | \epsilon_1 - \epsilon_2 |^{1/2}. \quad (48) \]
In terms of $\Delta = \epsilon - 1$, we obtain form (23), so that part of the earlier development is appropriate.

In particular, if we retain $\epsilon_1$ to $0(\delta^3)$ and $P$ to $0(\delta^6)$, then

$$(t - 1)/\epsilon = \Delta/\epsilon = \delta(1 - \delta D + \delta^2 D^2) + \delta^2(P + iS)$$  \hspace{1cm} (49)$$

is correct to $0(\delta^3)$, $0(x^2\delta^3)$, and $0(x\delta^6)$. For complex $\delta = \delta_1 + i\delta_2$, we construct $\Delta = \delta_1 + i\delta_2$, essentially as for form (22:25) with the earlier $iS$ replaced by $iS + P$ but retain only the leading term of the earlier $\delta^1$ contribution. Thus

$$\Delta_r/\epsilon = \delta_1(1 - \delta_1 D + \delta_1^2 D^2) + \delta_1^2(1 - 3\delta_1 D)$$
$$- 2\delta_1\delta_2 (S + (\delta_2^2 - \delta_2^3)P) \hspace{1cm} (50)$$

and

$$\Delta_i/\epsilon = \delta_1(1 - 2\delta_1 D + (3\delta_1^2 - \delta_2^3)D^2)$$
$$- (\delta_2^2 - \delta_2^3)S + 2\delta_1\delta_2 P \hspace{1cm} (51)$$

The terms in $P$ account for the $0(\delta^3)$ corrections indicated for forms (22:22) ff. If $\delta_1 = 0$, then $\Delta_i$ is independent of $S$, and $\Delta_r$ of $P$.

Similarly in terms of $v = \eta - 1$ with $\eta = \eta_1/\eta_2$, we write $\delta_1 = \eta_2 - 1 = v(2 + v)$ and express $\eta - 1$ to $0(v^3)$, $0(x^2\nu^2)$, and $0(\nu^6)$ as

$$t\eta - 1/\epsilon = v + v^2(4 + 4S)/2 + v^3R/2, \hspace{1cm} (52)$$
$$A = 1 - (w + 4D), \hspace{1cm} B = -(1 - w(u + 4D) + 8D^2).$$

For complex $v = v_r + i v_i$, we construct $\eta = \eta_r + i\eta_i$ essentially as for form (22:28). Thus the refractive contrast $\eta - 1$ is given by

$$t\eta_r - 1/\epsilon = v_r(1 + v_r A + v_r^2 R)(2) - v_i^2 A + 3v_r R)/2$$
$$- 4v_r v_i S' + 2(v_r^2 - v_i^2)P, \hspace{1cm} (53)$$

and the net attenuation is determined by

$$t\eta_i/\epsilon = v_r(1 + v_r A + (3v_r^2 - v_i^2)R)/2$$
$$+ 2(v_r^2 - v_i^2)S + 4v_r v_i P. \hspace{1cm} (54)$$

If the scatterers are lossless ($v = 0$), then $\eta_i$ does not depend on $S$, and $\eta_i$ is independent of $P$. The present forms (50)-(54) hold for all small-scaled particles to aligned ellipsoidal particles and co-centered nonspherical ellipsoid exclusion regions are given in Ref. 2.

For measurements in which the parameters $\epsilon_0$ and $\eta_0$ of the embedding medium are varied, we normalize $\epsilon_1$ and $\eta_0$ with respect to $\epsilon_0$ and $\eta_0$ instead of $\epsilon_0$ and $\eta_0$. See Ref. 2 for details.

The values of $D$, $P$, $A$, $B$, and $R$ for forms (49)-(54) are given by the following for the special cases at hand. For slabs,

$$D_1 = 0, \hspace{1cm} A_1 = 1 - w, \hspace{1cm} B_1 = -w(1 - u), \hspace{1cm} (55)$$

$$P_1 = -4x^2(2 - W + 3N)/3, \hspace{1cm} S_1 = xW, \hspace{1cm} (56)$$

with $W$ and $N$ as forms (19) and (23). For cylinders and lateral polarization, $D_{2y}$, $A_{2y}$, $B_{2y}$ equals $D_1$, $A_1$, $B_1$, and

$$P_{2y} = x^2(1 + 2u + 4M)/8, \hspace{1cm} S_{2y} = x^2\pi W/4. \hspace{1cm} (57)$$

For cylinders and transverse polarization,

$$D_{2x} = (1 - u)/2, \hspace{1cm} A_{2x} = -(1 - u), \hspace{1cm} B_{2x} = -u(1 - u) = B_{2y}, \hspace{1cm} (58)$$

$$P_{2x} = x^2(1 + 2u + 4M + W)/16 = P_{2y}/2 + x^2W/16, \hspace{1cm} (59)$$

$$S_{2x} = x^2\pi W/8 = S_{2y}/2, \hspace{1cm} (60)$$

with $M$ and $W$ as in forms (25)-(30). For spheres,

$$D_3 = (1 - u)/3, \hspace{1cm} A_3 = -(1 - u)/3, \hspace{1cm} B_3 = -(1 - u)(4 + 5u)/9, \hspace{1cm} (60)$$

$$P_3 = x^2(6 + 3\nu - 5N)11/(15)^2, \hspace{1cm} S_3 = x^2\pi W/9, \hspace{1cm} (61)$$

with $N$ and $W$ as in forms (38), (39), (42), and (43).

For cylinders, the values of $\epsilon'$ and $\eta'$ for the lateral and transverse cases may differ, and the corresponding birefringence ($\eta_1 - \eta_0$) will then display intrinsic as well as form effects. See Ref. 2 for details. The relations $A_{2l} = -A_{2t}$ and $B_{2l} = B_{2t}$ simplify considerations. In particular, if there is a common $v$, then

$$\eta_1 - \eta_0 = v v^2[A_1 - P_1 - x^2W/16 + iS_1]$$
$$= v v^2[1 - w + x(1 - 2u + M - W)/16$$
$$+ v x^2W/4] = v^2(R + il). \hspace{1cm} (62)$$

Then the birefringence corresponds to

$$\text{Ret}(\eta_1 - \eta_0) = (v_r - v_i)R - 2v_r v_iJ \hspace{1cm} (63)$$

and the dichroism to

$$\text{Im}(\eta_1 - \eta_0) = 2v_r v_i R + (v_r^2 - v_i^2)I. \hspace{1cm} (64)$$

For experiments in which $\eta_i$ is varied, we introduce the variable $x_\perp = \eta_\perp/\eta_0$, and the constant $\mu = \eta_\perp/\eta_0$ to construct $x = (\xi_\perp + i\mu)/(1 + \xi)$. For small $\xi$, we have

$$\text{Ret}(\eta_1 - \eta_0)/\eta_0 = \mu(x^2 - \mu^2)R + 2\xi_\mu I[1 - (1 - \xi)], \hspace{1cm} (65)$$

$$\text{Im}(\eta_1 - \eta_0)/\eta_0 = [-2\xi_\mu R + (\xi^2 - \mu^2)I](1 - \xi), \hspace{1cm} (66)$$

which generalize the result in form (1:67).

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