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Abstract. Two different formalisms for the homogenization of composite materials containing oriented ellipsoidal particles of isotropic dielectric materials are being named after Bruggeman. Numerical studies reveal clear differences between the two formalisms which may be exacerbated: (i) if the component particles become more aspherical, (ii) at mid-range values of the volume fractions, and (iii) if the homogenized component material is dissipative. Only the correct Bruggeman formalism uses the correct polarizability density dyadics of the component particles in the homogenized composite material (HCM) and is directly derivable from the frequency-domain Maxwell postulates specialized for the HCM. © 2012 Society of Photo-Optical Instrumentation Engineers (SPIE). [DOI: 10.1117/1.JNP.6.069501]

The Bruggeman formalism provides a well-established technique for estimating the effective constitutive parameters of homogenized composite materials (HCMs).[1–3] The scope of its applicability is not restricted to dilute composite materials and it is easy to implement numerically, both of which contribute to its enduring popularity.

The Bruggeman formalism was originally devised for isotropic dielectric HCMs, comprising two (or more) isotropic dielectric component materials distributed randomly as electrically small spherical particles.[4] Generalizations of the Bruggeman formalism which accommodate anisotropic and bianisotropic HCMs have been developed.[5] A rigorous basis for the Bruggeman formalism—for isotropic dielectric,[6] anisotropic dielectric,[7,8] and bianisotropic[9,10] HCMs—is provided by the strong-property-fluctuation theory, whose lowest-order formulation is the Bruggeman formalism. (In Ref. 7, the formula of Polder and van Santen, for the effective relative permittivity of an isotropic dielectric HCM, yields the same results as the formula of Bruggeman.[4])

Our focus in this letter is on HCMs arising from two isotropic dielectric component materials, labeled a and b. Their relative permittivities are ϵ^a and ϵ^b, while their volume fractions are f_a and $f_b \equiv 1 - f_a$. Both component materials are assumed to be randomly distributed as electrically small ellipsoidal particles. For simplicity, all component particles have the same shape and orientation. The surface of each ellipsoid, relative to its centroid, may be represented by the vector

$$\underline{r}_e(\theta, \phi) = \eta \underline{\underline{U}} \bullet \hat{\underline{r}}(\theta, \phi), \tag{1}$$

with $\hat{\underline{r}}$ being the radial unit vector from the ellipsoid's centroid, specified by the spherical polar coordinates θ and ϕ. The linear dimensions of each ellipsoid, as determined by the parameter η,

are assumed to be small relative to the electromagnetic wavelength(s). Let us choose our coordinate system to be such that the Cartesian axes are aligned with the principal axes of the ellipsoids. Then the ellipsoidal shape is captured by the dyadic

$$\underline{U} = U_x \hat{\underline{x}}\,\hat{\underline{x}} + U_y \hat{\underline{y}}\,\hat{\underline{y}} + U_z \hat{\underline{z}}\,\hat{\underline{z}}, \tag{2}$$

wherein the shape parameters $U_{x,y,z} > 0$ and $\{\hat{\underline{x}}, \hat{\underline{y}}, \hat{\underline{z}}\}$ are unit vectors aligned with the Cartesian axes.

The ellipsoidal shape of the component particles results in the corresponding HCM being an orthorhombic biaxial dielectric material. That is, the Bruggeman estimate of the HCM relative permittivity dyadic has the form

$$\underline{\epsilon}^{Br1} = \epsilon_x^{Br1} \hat{\underline{x}}\,\hat{\underline{x}} + \epsilon_y^{Br1} \hat{\underline{y}}\,\hat{\underline{y}} + \epsilon_z^{Br1} \hat{\underline{z}}\,\hat{\underline{z}}. \tag{3}$$

The relative permittivity parameters $\epsilon_{x,y,z}^{Br1}$ are given implicitly by the three coupled equations[11]

$$\frac{\epsilon^a - \epsilon_\ell^{Br1}}{1 + D_\ell(\epsilon^a - \epsilon_\ell^{Br1})} f_a + \frac{\epsilon^b - \epsilon_\ell^{Br1}}{1 + D_\ell(\epsilon^b - \epsilon_\ell^{Br1})} f_b = 0, \qquad (\ell \in \{x, y, z\}). \tag{4}$$

Herein D_ℓ are components of the depolarization dyadic

$$\underline{D} = D_x \hat{\underline{x}}\,\hat{\underline{x}} + D_y \hat{\underline{y}}\,\hat{\underline{y}} + D_z \hat{\underline{z}}\,\hat{\underline{z}}, \tag{5}$$

specified by the double integrals[5]

$$\left. \begin{array}{l} D_x = \dfrac{1}{4\pi} \displaystyle\int_0^{2\pi} d\phi \int_0^\pi d\theta \dfrac{\sin^3\theta \cos^2\phi}{U_x^2 \rho} \\[3mm] D_y = \dfrac{1}{4\pi} \displaystyle\int_0^{2\pi} d\phi \int_0^\pi d\theta \dfrac{\sin^3\theta \sin^2\phi}{U_y^2 \rho} \\[3mm] D_z = \dfrac{1}{4\pi} \displaystyle\int_0^{2\pi} d\phi \int_0^\pi d\theta \dfrac{\sin\theta \cos^2\theta}{U_z^2 \rho} \end{array} \right\}, \tag{6}$$

which involve the scalar parameter

$$\rho = \frac{\sin^2\theta \cos^2\phi}{U_x^2} \epsilon_x^{Br1} + \frac{\sin^2\theta \sin^2\phi}{U_y^2} \epsilon_y^{Br1} + \frac{\cos^2\theta}{U_z^2} \epsilon_z^{Br1}. \tag{7}$$

The coupled nature of the three equations in Eq. (4) means that numerical methods are generally needed to extract the relative permittivity parameters $\epsilon_{x,y,z}^{Br1}$ from them.

An alternative formalism for the homogenization of the same composite material as in the foregoing paragraph is also referred to as the Bruggeman formalism.[12–15] Let us write the estimate of the HCM's relative permittivity dyadic provided by this alternative formalism as

$$\underline{\epsilon}^{Br2} = \epsilon_x^{Br2} \hat{\underline{x}}\,\hat{\underline{x}} + \epsilon_y^{Br2} \hat{\underline{y}}\,\hat{\underline{y}} + \epsilon_z^{Br2} \hat{\underline{z}}\,\hat{\underline{z}}. \tag{8}$$

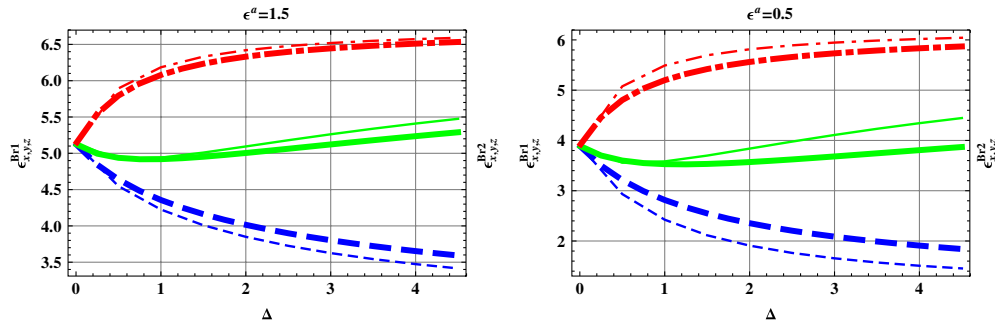The relative permittivity parameters $\epsilon_{x,y,z}^{Br2}$ are given by the three equations

$$\frac{\epsilon^a - \epsilon_\ell^{Br2}}{\epsilon_\ell^{Br2} + L_\ell(\epsilon^a - \epsilon_\ell^{Br2})} f_a + \frac{\epsilon^b - \epsilon_\ell^{Br2}}{\epsilon_\ell^{Br2} + L_\ell(\epsilon^b - \epsilon_\ell^{Br2})} f_b = 0, \qquad (\ell \in \{x, y, z\}), \tag{9}$$

wherein the depolarization factors[16]

$$L_\ell = \frac{U_x U_y U_z}{2} \int_0^\infty ds \frac{1}{(s + U_\ell^2)\sqrt{(s + U_x^2)(s + U_y^2)(s + U_z^2)}}, \qquad (\ell \in \{x, y, z\}) \tag{10}$$

are components of the depolarization dyadic

$$\underline{L} = L_x \hat{\underline{x}}\,\hat{\underline{x}} + L_y \hat{\underline{y}}\,\hat{\underline{y}} + L_z \hat{\underline{z}}\,\hat{\underline{z}}. \tag{11}$$

Fig. 1 The estimates $\epsilon_x^{Br1,2}$ (blue, dashed curves), $\epsilon_y^{Br1,2}$ (green, solid curves), and $\epsilon_z^{Br1,2}$ (red, broken dashed curves) plotted versus the asphericity parameter $\Delta \in (0, 4.5)$. The $\epsilon_{x,y,z}^{Br1}$ estimates are represented by thick curves and the $\epsilon_{x,y,z}^{Br2}$ estimates are represented by thin curves. The ellipsoidal shapes of the component material particles are described by shape parameters $U_x = 1$, $U_y = 1 + (\Delta/3)$, and $U_z = 1 + 2\Delta$. The relative permittivities of the component materials are $\epsilon^a \in \{0.5, 1.5\}$ and $\epsilon^b = 12$; and the volume fraction $f_a = 0.5$.

Each of the three equations in Eq. (9) is a quadratic equation in ϵ_ℓ^{Br2} whose solution may be explicitly expressed as

$$\epsilon_\ell^{Br2} = \frac{-\beta \pm \sqrt{\beta^2 - 4\alpha\gamma}}{2\alpha}, \qquad (\ell \in \{x, y, z\}), \tag{12}$$

with $\alpha = L_\ell - 1$, $\beta = \epsilon^a(f_a - L_\ell) + \epsilon^b(f_b - L_\ell)$, and $\gamma = L_\ell \epsilon^a \epsilon^b$. The sign of the square root term in the solution seen in Eq. (12) may be determined by appealing to the anisotropic dielectric generalization of the Hashin–Shtrikman bounds,[17] for example.

Let us illustrate the differences between the estimates $\underline{\epsilon}^{Br1}$ and $\underline{\epsilon}^{Br2}$ by means of some representative numerical results. The two estimates are identical for the limiting case represented by $U_x = U_y = U_z = 1$ (i.e., for isotropic dielectric HCMs), but differences emerge as the asphericity of the component particles intensifies. Suppose that the shape parameters describing the component ellipsoids have the form $U_x = 1$, $U_y = 1 + (\Delta/3)$, and $U_z = 1 + 2\Delta$. Thus, the asphericity of the ellipsoids is governed by the scalar parameter Δ. We begin with the nondissipative scenario wherein $\epsilon^a \in \{0.5, 1.5\}$ and $\epsilon^b = 12$. Also, we fix $f_a = 0.5$. Plots of the relative permittivity parameters $\epsilon_{x,y,z}^{Br1,Br2}$ versus the asphericity parameter Δ are presented in Fig. 1. The difference between ϵ_x^{Br1} and ϵ_x^{Br2} grows steadily as Δ increases, reaches a maximum for $1 < \Delta < 2$, and then slowly shrinks as Δ increases beyond 2. The difference between ϵ_z^{Br1} and ϵ_z^{Br2} follows a similar pattern. However, in the case of ϵ_y^{Br1} and ϵ_y^{Br2}, the difference increases uniformly as Δ increases. The differences between $\epsilon_{x,y,z}^{Br1}$ and $\epsilon_{x,y,z}^{Br2}$ are generally greater for $\epsilon^a = 0.5$ than for $\epsilon^a = 1.5$. In the former case the maximum difference is approximately 15%, whereas in the latter case it is approximately 5%.

We turn now to the effect of volume fraction. The calculations of Fig. 1 are repeated for Fig. 2 except that here the relative permittivity parameters $\epsilon_{x,y,z}^{Br1,Br2}$ are plotted versus the volume fraction f_a, while the asphericity parameter is fixed at $\Delta = 4.5$. The differences between the estimates of the two formalisms are clearly greatest at mid-range values of f_a, and they are generally greater for $\epsilon^a = 0.5$ than for $\epsilon^a = 1.5$.

Lastly, the effects of dissipation are considered. We repeated the calculations of Fig. 1 but with $\Delta = 4.5$ and $\epsilon^a \in \{0.5 + i\delta, 1.5 + i\delta\}$. Here $\delta > 0$ governs the degree of dissipation exhibited by component material a. The real and imaginary parts of the relative permittivity parameters $\epsilon_{x,y,z}^{Br1,Br2}$ are plotted versus the dissipation parameter δ in Fig. 3. The differences between the real parts of the estimates $\epsilon_{x,y,z}^{Br1}$ and $\epsilon_{x,y,z}^{Br2}$ are largest when component material a is nondissipative and they decrease uniformly as δ increases. In contrast, the differences between the imaginary parts of the estimates $\epsilon_{x,y,z}^{Br1}$ and $\epsilon_{x,y,z}^{Br2}$ increase as δ increases. These differences in the imaginary parts generally reach a maximum for mid-range values of δ and thereafter decrease as δ increases. For both the real and imaginary parts of the estimates $\epsilon_{x,y,z}^{Br1}$ and $\epsilon_{x,y,z}^{Br2}$, generally larger differences arise for $\epsilon^a = 0.5 + i\delta$ than for $\epsilon^a = 1.5 + i\delta$.
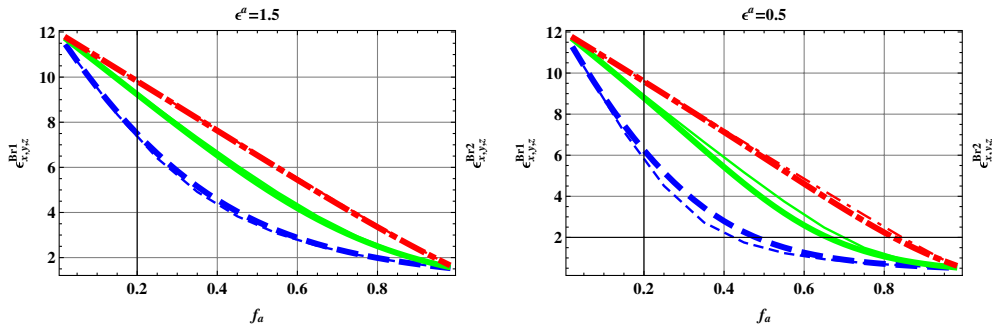
Fig. 2 As Fig. 1 except that $\Delta = 4.5$ and the estimates $\epsilon_{x,y,z}^{Br1,2}$ are plotted versus the volume fraction $f_a \in (0,1)$.
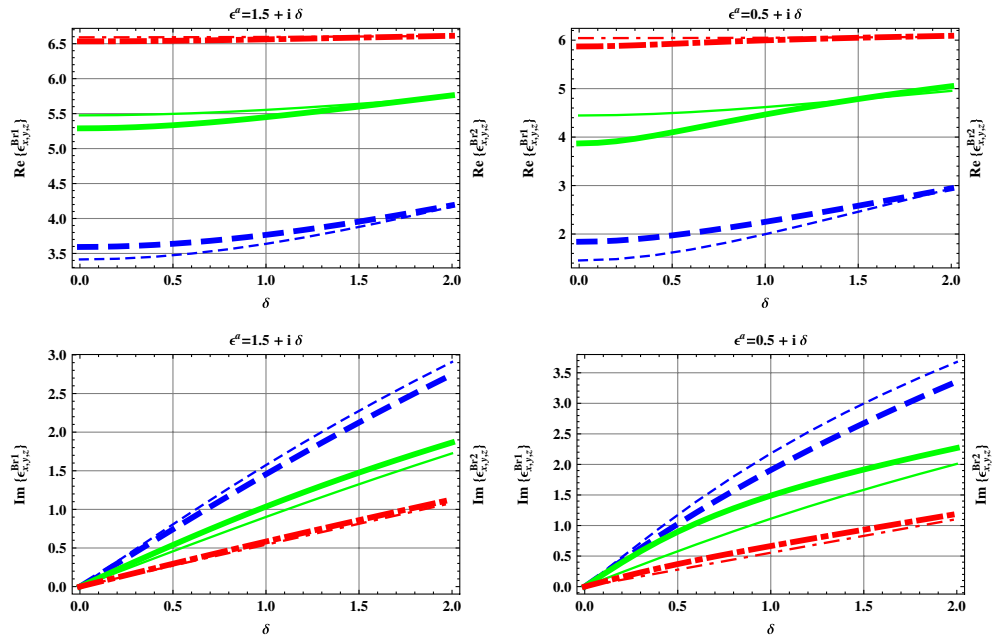
Fig. 3 As Fig. 1 except that $\Delta = 4.5$, $\epsilon^a \in \{0.5 + i\delta, 1.5 + i\delta\}$, and the real and imaginary parts of the estimates $\epsilon_{x,y,z}^{Br1,2}$ are plotted versus the dissipation parameter $\delta \in (0,2)$.

Thus, there are significant differences between the estimates $\underline{\epsilon}^{Br1}$ and $\underline{\epsilon}^{Br2}$ when ellipsoidal component particles are considered. These differences may be exacerbated: (i) if the component particles become more aspherical, (ii) at mid-range values of the volume fractions of the component materials, and (iii) if the HCM is dissipative. The differences between the two estimates may be further exacerbated if one of the component materials has a positive-valued relative permittivity which is less than unity (or a relative permittivity whose real part is positive-valued and less than unity). Relative permittivities in this range are associated with novel materials possessing engineered nanostructures; these artificial materials have been the subject of intense research lately.[18–20] (The parameter regime wherein one of the component materials has a positive-valued relative permittivity while the other has a negative-valued relative permittivity (or likewise for the real parts of the relative permittivities in the case of dissipative HCMs) is avoided here because the Bruggeman formalism can deliver estimates in this regime which are not physically plausible.[21])

The differences between the two formalisms stem from the differences between the depolarization dyadics $\underline{D}$ and $\underline{L}$. The Bruggeman formalism conceptually employs an average-polarizability-density approach:[22] Suppose the composite material has been homogenized into an HCM. Into this HCM, let additional aligned ellipsoidal particles of the two component materials be dispersed in such a way as to maintain the overall volume fractions of a and b. This

dispersal cannot change the effective properties of the HCM—because the volume-fraction-average of the polarization density dyadics of two particles, one of each component material, embedded in the HCM with relative permittivity $\underline{\epsilon}^{Br1}$ is exactly zero. In computing the polarizability density dyadic of each particle, it must therefore be assumed that the particle is surrounded by the HCM. This fact legitimizes the use of $\underline{D}$, which contains the anisotropic HCM's effective constitutive properties via the scalar ρ of Eq. (7). Indeed, Eq. (4) is directly derivable from the frequency-domain Maxwell postulates specialized for the actual HCM.[6–10]

On the other hand, use of $\underline{L}$ to compute the polarizability density dyadic of a particle implies that it is surrounded by an isotropic HCM, which is clearly incorrect. The alternative formalism that delivers $\underline{\epsilon}^{Br2}$ is a heuristic extrapolation of the Bruggeman formalism for isotropic dielectric HCMs,[12,13] and it lacks the rigorous basis that underpins the estimate of $\underline{\epsilon}^{Br1}$. The dispersal of aligned ellipsoidal particles of the two component materials without disturbing the overall volume fractions into the HCM with relative permittivity $\underline{\epsilon}^{Br2}$ would change the effective properties of the HCM—because the volume-fraction-average of the polarization density dyadics of two particles, one of each component material, embedded in the HCM with relative permittivity $\underline{\epsilon}^{Br2}$ would not be zero.

We have thus delineated the differences between the two formalisms and identified one of them as the correct Bruggeman formalism. We hope that this exposition will prevent confusion between the two formalisms from perpetuating.

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