Depolarization regions of nonzero volume for anisotropic, cubically nonlinear, homogenized nanocomposites

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Abstract

An implementation of the strong–permittivity–fluctuation theory (SPFT) is presented in order to estimate the constitutive parameters of a homogenized composite material (HCM) which is both cubically nonlinear and anisotropic. Unlike conventional approaches to homogenization, the particles which comprise the component material phases are herein assumed to be small but not vanishingly small. The influence of particle size on the estimates of the HCM constitutive parameters is illustrated by means of a representative numerical example. It is observed that, by taking the nonzero particle size into consideration, attenuation is predicted and nonlinearity enhancement is somewhat diminished. In these respects, the effect of particle size is similar to that of correlation length within the bilocally–approximated SPFT.

Keywords: Strong–permittivity–fluctuation theory, nonlinearity enhancement, ellipsoidal particles, depolarization dyadic

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1 INTRODUCTION

Formalisms based on the strong–permittivity–fluctuation theory (SPFT) have been developed to estimate the constitutive parameters of homogenized composite materials (HCMs), within the realms of both linear \([1, 2, 3, 4, 5, 6]\) and weakly nonlinear \([7, 8, 9, 10]\) materials. The SPFT approach to homogenization has the advantage over more conventional approaches, such as those named after Maxwell Garnett and Bruggeman, that the distributional statistics of the component material phases is generally better taken into account \([11]\). For example, the SPFT is most commonly implemented at the level of the bilocal approximation, wherein the distributional statistics are described in terms of a two–point covariance function and its associated correlation length. Thereby, the

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bilocally–approximated SPFT predicts attenuation in the HCM, even when the component phase materials are nondissipative.

In the SPFT approach to homogenization, and likewise the Maxwell Garnett and Bruggeman approaches, the electromagnetic responses of the particles which make up the component material phases are represented by electrically–small depolarization regions [11]. Often, these depolarization regions are taken to be vanishingly small [12, 13]. The spatial extent of the component phase particles is thereby neglected. However, the importance of the nonzero spatial extent of the component phase particles has been underlined by several studies pertaining to linear isotropic HCMs [14, 15, 16, 17, 18, 19]. Furthermore, versions of the SPFT which accommodate depolarization regions of nonzero volume were recently developed for linear anisotropic [20] and bianisotropic [21] HCMs. While these studies take into consideration the nonzero spatial extent of the component phase particles, it is essential that the particle sizes are much smaller than the electromagnetic wavelengths, in order for the assembly of component material phases to be regarded as an effectively homogeneous material [11]. Accordingly, at an optical wavelength of 600 nm, for example, component phase particles with linear dimensions less than approximately 60 nm are envisaged.

In the present study, we extend the consideration of depolarization regions of nonzero volume into the weakly nonlinear regime for anisotropic HCMs. Our starting point is the SPFT for cubically nonlinear HCMs which incorporates vanishingly small depolarization regions [10]. In §2 the component material phases and their statistical distributions are described. The linear and weakly nonlinear contributions to the depolarization dyadics, arising from depolarization regions of nonzero volume, are presented in §3. The corresponding bilocally–approximated SPFT estimate of the HCM permittivity dyadic is given in §4; and a representative numerical example is used to illustrate these results in §5. Finally, a few closing remarks are provided in §6.

In the notation adopted, single underlining denotes a 3 vector whereas double underlining denotes a 3×3 dyadic. The inverse, determinant, trace and adjoint of a dyadic $M$ are represented as $M^{-1}$, $\det(M)$, $\tr(M)$ and $M^{\text{adj}}$, respectively. The permittivity and permeability of free space are written as $\varepsilon_0$ and $\mu_0$.

## 2 HOMOGENIZATION PRELIMINARIES

The homogenization of two component material phases, namely phase $a$ and phase $b$, is considered. Each component phase is an isotropic dielectric material; and, in general, each is cubically nonlinear. Thus, the permittivities of the component material phases are expressed as

$$\varepsilon\ell = \varepsilon\ell_0 + \chi\ell |E\ell|^2,$$

(\ell = a, b),

with $\varepsilon\ell_0$ being the linear permittivity, $\chi\ell$ the nonlinear susceptibility, and $E\ell$ is the electric field developed inside a region of phase $\ell$ by illumination of the composite material. The assumption of weak nonlinearity ensures that $|\varepsilon\ell_0| \gg |\chi\ell||E\ell|^2$. Notice that nonlinear permittivities of the form (1) describe electrostrictive materials which can induce stimulated Brillouin scattering [22].

The component material phases $a$ and $b$ are made up of ellipsoidal particles. The particles all have

\footnote{In the context of bianisotropic HCMs, the initials SPFT stand for strong–property–fluctuation theory.}
the same shape and orientation, as specified by the shape dyadic

\[ U = \frac{1}{\sqrt{U_x U_y U_z}} \text{diag}(U_x, U_y, U_z), \quad (U_x, U_y, U_z > 0), \quad (2) \]

which parameterizes the particle surface as

\[ r^e(\theta, \phi) = \eta \hat{U} \cdot \hat{r}(\theta, \phi), \quad (3) \]

where \( \hat{r}(\theta, \phi) \) is the radial unit vector specified by the spherical polar coordinates \( \theta \) and \( \phi \). The size parameter \( \eta \) provides a measure of the linear dimensions of the ellipsoidal particles. It is assumed that \( \eta \) is much smaller than the electromagnetic wavelengths, but not vanishingly small.

The component phase particles are randomly distributed throughout a region of volume \( V \), which is partitioned into the disjoint regions of volume \( V_a \) and \( V_b \) containing phase \( a \) and \( b \), respectively. Thus, the component phase distributions are characterized in terms of statistical moments of the characteristic functions

\[ \Phi_{\ell}(r) = \begin{cases} 1, & r \in V_{\ell}, \\ 0, & r \not\in V_{\ell}, \end{cases} \quad (\ell = a, b). \quad (4) \]

The first statistical moment of \( \Phi_{\ell} \) delivers the volume fraction of phase \( \ell \), i.e., \( \langle \Phi_{\ell}(r) \rangle = f_{\ell} \). Plainly, \( f_a + f_b = 1 \). The two-point covariance function which constitutes the second statistical moment of \( \Phi_{\ell} \) is taken as the physically-motivated form [23]

\[ \langle \Phi_{\ell}(r) \Phi_{\ell}(r') \rangle = \begin{cases} \langle \Phi_{\ell}(r) \rangle \langle \Phi_{\ell}(r') \rangle, & |U^{-1} \cdot (r - r')| > L, \\ \langle \Phi_{\ell}(r) \rangle, & |U^{-1} \cdot (r - r')| \leq L, \end{cases} \quad (5) \]

where \( L > 0 \) is the correlation length. Within the SPFT, the estimates of HCM constitutive parameters are largely insensitive to the specific form of the covariance function, as has been shown by comparative studies [8, 24].

### 3 DEPOLARIZATION DYADIC

Let us focus our attention on a single component phase particle of volume \( V^e \), characterized by the shape dyadic \( U \) and size parameter \( \eta \). Suppose that this particle is embedded in a comparison medium. In consonance with the ellipsoidal geometry of the component phase particles and the weakly nonlinear permittivities of the component material phases, the comparison medium is a weakly nonlinear, anisotropic, dielectric medium characterized by the permittivity dyadic

\[ \varepsilon_{cm} = \varepsilon_{cm0} + \chi_{cm} |\bar{E}_{HCM}|^2 = \text{diag}(\varepsilon^e_{cm0}, \varepsilon^y_{cm0}, \varepsilon^z_{cm0}) + \text{diag}(\chi^x_{cm}, \chi^y_{cm}, \chi^z_{cm}) |\bar{E}_{HCM}|^2, \quad (6) \]

where \( \bar{E}_{HCM} \) denotes the spatially-averaged electric field in the HCM. The eigenvectors of \( \varepsilon_{cm} \) are aligned with those of \( U \).
The depolarization dyadic [12]

\[ D(\eta) = \int_{V_e} G_{cm}(r) \, d^3 \mathbf{r} \]  

(7)

provides the electromagnetic response of the ellipsoidal particle embedded in the comparison medium. Here, the dyadic Green function of the comparison medium, namely \( G_{cm}(\mathbf{r}) \), satisfies the nonhomogeneous vector Helmholtz equation

\[ \nabla \times \nabla \times I - \omega^2 \mu_0 \varepsilon_{cm} \cdot G_{cm}(\mathbf{r} - \mathbf{r}') = i\omega \mu_0 \delta(\mathbf{r} - \mathbf{r}') I. \]  

(8)

An explicit representation of \( G_{cm}(\mathbf{r}) \) is not generally available [25], but its Fourier transform,

\[ \hat{G}_{cm}(\mathbf{q}) = \int_{\mathbb{R}^3} G_{cm}(\mathbf{r}) \exp(-i\mathbf{q} \cdot \mathbf{r}) \, d^3 \mathbf{r}, \]  

(9)

may be deduced from (8) as

\[ \hat{G}_{cm}(\mathbf{q}) = -i\omega \mu_0 \left( \mathbf{q} \times \frac{\mathbf{q}}{q^2} + \omega^2 \mu_0 \varepsilon_{cm} \right)^{-1}. \]  

(10)

By combining (7), (9) and (10),

\[ D = \frac{\eta}{2\pi^2} \int_{\mathbb{R}^3} \frac{1}{q^2} \left[ \sin(q\eta) \frac{q}{q\eta} - \cos(q\eta) \right] \left[ \lim_{q \to \infty} \hat{G}_{cm}(U^{-1} \cdot \mathbf{q}) \right] \, d^3 \mathbf{q} \]  

(11)

is obtained, after some simplification [12, 26].

3.1 Depolarization contributions from regions of nonzero volume

As in [20], we express the depolarization dyadic as the sum

\[ D = D^{\eta=0} + D^{\eta>0}, \]  

(12)

where the dyadic

\[ D^{\eta=0} = \frac{\eta}{2\pi^2} \int_{\mathbb{R}^3} \frac{1}{q^2} \left[ \sin(q\eta) \frac{q}{q\eta} - \cos(q\eta) \right] \left[ \lim_{q \to \infty} \hat{G}_{cm}(U^{-1} \cdot \mathbf{q}) \right] \, d^3 \mathbf{q} \]  

(13)

represents the depolarization contribution arising from the region of vanishingly small volume \( \lim_{\eta \to 0} V_e \), whereas the dyadic

\[ D^{\eta>0} = \frac{\eta}{2\pi^2} \int_{\mathbb{R}^3} \frac{1}{q^2} \left[ \sin(q\eta) \frac{q}{q\eta} - \cos(q\eta) \right] \left\{ \hat{G}_{cm}(U^{-1} \cdot \mathbf{q}) - \left[ \lim_{q \to \infty} \hat{G}_{cm}(U^{-1} \cdot \mathbf{q}) \right] \right\} \, d^3 \mathbf{q} \]  

(14)

provides the depolarization contribution arising from the region of nonzero volume \( \left( V_e - \lim_{\eta \to 0} V_e \right) \).
Depolarization dyadics associated with vanishingly small regions have been studied extensively [13, 26]. The volume integral (13) reduces to the \( \eta \)-independent surface integral [12]

\[
D^{\eta=0} = \frac{1}{4\pi i \omega} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \left[ \frac{1}{\text{tr} \left( \begin{array}{c} \epsilon_{cm} \cdot \mathbf{A} \end{array} \right)} \mathbf{A} \right] \sin \theta \, d\theta \, d\phi,
\]

(15)

with

\[
\mathbf{A} = \text{diag} \left( \frac{\sin^2 \theta \cos^2 \phi}{U_x^2}, \frac{\sin^2 \theta \sin^2 \phi}{U_y^2}, \frac{\cos^2 \theta}{U_z^2} \right).
\]

(16)

An elliptic function representation for \( D^{\eta=0} \) is available [27] (which simplifies to a hyperbolic function representation in the case of a spheroidal depolarization region [12]), but for our present purposes the integral representation (15) is more convenient.

Depolarization dyadics associated with small regions of nonzero volume have lately come under scrutiny for anisotropic [20] and bianisotropic [21] HCMs. As described elsewhere [10, 20], by the calculus of residues (14) reduces to

\[
D^{\eta>0} = \frac{1}{4\pi i \omega} W(\eta),
\]

(17)

where the dyadic function

\[
W(\eta) = \eta^3 \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \sin \theta \left\{ \left[ \frac{3(\kappa_+ - \kappa_-)}{2\eta} + i \left( \kappa_+^2 - \kappa_-^2 \right) \right] A_0 + i\omega^2 \mu_0 \left( \kappa_+ - \kappa_- \right) \beta \right\} d\theta d\phi
\]

(18)

is introduced. Herein, the dyadics

\[
\alpha = \left[ 2\epsilon_{cm} - \text{tr} \left( \begin{array}{c} \epsilon_{cm} \end{array} \right) \mathbf{I} \right] \cdot \mathbf{A} - \text{tr} \left( \begin{array}{c} \epsilon_{cm} \cdot \mathbf{A} \end{array} \right) \mathbf{I} - \frac{\text{tr} \left( \begin{array}{c} \epsilon_{adj} \cdot \mathbf{A} \end{array} \right) - \left[ \text{tr} \left( \begin{array}{c} \epsilon_{adj} \end{array} \right) \text{tr} \left( \begin{array}{c} \mathbf{A} \end{array} \right) \right] \mathbf{A}}{\text{tr} \left( \begin{array}{c} \epsilon_{cm} \cdot \mathbf{A} \end{array} \right)}\mathbf{A},
\]

(19)

\[
\beta = \frac{\text{det} \left( \begin{array}{c} \epsilon_{cm} \end{array} \right)}{\text{tr} \left( \begin{array}{c} \epsilon_{cm} \cdot \mathbf{A} \end{array} \right)} \mathbf{A}
\]

(20)

and the scalars

\[
\Delta = \sqrt{t_B^2 - 4t_At_C},
\]

(21)

\[
\kappa_\pm = \mu_0 \omega^2 \frac{t_A \pm \Delta}{2t_C},
\]

(22)

with

\[
t_A = \text{det} \left( \begin{array}{c} \epsilon_{cm} \end{array} \right),
\]

\[
t_B = \text{tr} \left( \begin{array}{c} \epsilon_{adj} \cdot \mathbf{A} \end{array} \right) - \left[ \text{tr} \left( \begin{array}{c} \epsilon_{adj} \end{array} \right) \text{tr} \left( \begin{array}{c} \mathbf{A} \end{array} \right) \right] \right),
\]

(23)

\[
t_C = \text{tr} \left( \begin{array}{c} \epsilon_{cm} \cdot \mathbf{A} \end{array} \right) \text{tr} \left( \begin{array}{c} \mathbf{A} \end{array} \right)
\]

Often the approximation \( D \approx D^{\eta=0} \) is implemented in homogenization studies [11]. However, studies of isotropic [14, 15, 16, 17, 18, 19], anisotropic [20] and bianisotropic [21] HCMs have emphasized the importance of the nonzero spatial extent of depolarization regions.
3.2 Linear and weakly nonlinear depolarization contributions

We exploit the fact that the comparison medium permittivity (6) is the sum of a linear part and a weakly nonlinear part to similarly express

\[ D = D_0 + D_1 |E_{HCM}|^2 = D^{\eta=0} + D^{\eta>0} + (D_1^{\eta=0} + D_1^{\eta>0}) |E_{HCM}|^2, \]  

where

\[ D^{\eta\geq0} = D_0^{\eta\geq0} + D_1^{\eta\geq0} |E_{HCM}|^2. \]  

The linear and weakly nonlinear contributions to \( D^{\eta=0} \) have been derived earlier [10]; these are

\[ D^{\eta=0}_0 = \frac{1}{4\pi i\omega} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \frac{1}{\text{tr}(\epsilon_{cm0} \cdot A)} A |A| \sin \theta \, d\theta \, d\phi, \]

\[ D^{\eta=0}_1 = -\frac{1}{4\pi i\omega} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \left\{ \frac{\text{tr}(\chi_{cm} \cdot A)}{\text{tr}(\epsilon_{cm0} \cdot A)} A \right\} \sin \theta \, d\theta \, d\phi. \]

The linear and weakly nonlinear contributions to \( D^{\eta>0} \) — and, equivalently, \( W(\eta) \) — follow from corresponding contributions for an expression analogous to (18) which crops up in the bilocally–approximated SPFT [5, 10]. Thus, we have

\[ W(\eta) = W_0(\eta) + W_1(\eta) |E_{HCM}|^2 \]  

with

\[ W_0(\eta) = \eta^3 \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \frac{\sin \theta}{3\Delta_0} \left[ \tau_\alpha(\eta) \alpha_0 + \tau_\beta(\eta) \beta_0 \right] \, d\theta \, d\phi \]

and

\[ W_1(\eta) = \eta^3 \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \frac{\sin \theta}{3\Delta_0} \left\{ \tau_\alpha(\eta) \left( \frac{\alpha_1 - \Delta_1}{\Delta_0} \alpha_0 \right) + \tau_\beta(\eta) \left( \beta_1 - \frac{\Delta_1}{\Delta_0} \beta_0 \right) \right\} \, d\theta \, d\phi, \]

where

\[ \tau_\alpha(\eta) = \frac{3(\kappa_0 - \kappa_{0-})}{2\eta} + i \left( \kappa_{0+}^2 - \kappa_{0-}^2 \right) \]

\[ \tau_\beta = i\omega^2 \mu_0 \left( \kappa_{0+}^2 - \kappa_{0-}^2 \right). \]
The dyadics $\underline{\underline{\alpha}}_0$ and $\underline{\beta}_0$, and scalars $\kappa_0\pm$ and $\Delta_0$, herein represent the linear parts of their counterpart dyadics $\underline{\underline{\alpha}}$ and $\underline{\beta}$, and scalars $\kappa_\pm$ and $\Delta$, as per [10]

$$\underline{\underline{\alpha}}_0 = \left[ 2 \underline{\varepsilon}_{cm0} - \text{tr} \left( \underline{\varepsilon}_{cm0} \cdot \underline{A} \right) \right] \cdot \underline{A} - \text{tr} \left( \underline{\varepsilon}_{cm0} \cdot \underline{A} \right) \underline{I} - \frac{\text{tr} \left( \underline{\varepsilon}_{cm0} \cdot \underline{A} \right) - \left[ \text{tr} \left( \underline{\varepsilon}_{cm0} \cdot \underline{A} \right) \right] \underline{A}}{\text{tr} \left( \underline{\varepsilon}_{cm0} \cdot \underline{A} \right)} \underline{A},$$

(32)

$$\underline{\beta}_0 = \underline{\varepsilon}_{cm0} - \frac{\text{det} \left( \underline{\varepsilon}_{cm0} \right)}{\text{tr} \left( \underline{\varepsilon}_{cm0} \cdot \underline{A} \right)} \underline{A},$$

(33)

$$\kappa_0\pm = \mu_0 \omega^2 \frac{t_{B0} \pm \Delta_0}{2 t_{C0}},$$

(34)

$$\Delta_0 = \sqrt{t_{B0}^2 - 4 t_{A0} t_{C0}},$$

(35)

with

$$t_{A0} = \text{det} \left( \underline{\varepsilon}_{cm0} \right),$$

$$t_{B0} = \text{tr} \left( \underline{\varepsilon}_{cm0} \cdot \underline{A} \right) - \left[ \text{tr} \left( \underline{\varepsilon}_{cm0} \cdot \underline{A} \right) \right],$$

$$t_{C0} = \text{tr} \left( \underline{\varepsilon}_{cm0} \cdot \underline{A} \right) \text{tr} \left( \underline{A} \right) \right),$$

(36)

Moreover, the weakly nonlinear contributions to $\underline{\underline{\alpha}}$, $\underline{\beta}$, $\kappa_\pm$ and $\Delta$ are provided as [10]

$$\underline{\underline{\alpha}}_1 = \left[ 2 \underline{\chi}_{cm} - \frac{t_{B1} t_{C0} - t_{B0} t_{C1}}{t_{C0} \text{tr} \left( \underline{\varepsilon}_{cm0} \cdot \underline{A} \right)} \right] \cdot \underline{A} - \text{tr} \left( \underline{\chi}_{cm} \cdot \underline{A} \right) \underline{I},$$

(37)

$$\underline{\beta}_1 = \underline{\chi} - \frac{t_{B1} t_{C0} - t_{B0} t_{C1}}{t_{C0} \text{tr} \left( \underline{\varepsilon}_{cm0} \cdot \underline{A} \right)} \underline{A},$$

(38)

$$\kappa_{1\pm} = \frac{\omega^2 \left( -t_{B1} \pm \Delta_1 \right) - 2 t_{C1} \kappa_{0\pm}}{2 t_{C0}},$$

(39)

$$\Delta_1 = \frac{t_{B0} t_{B1} - 2 \left( t_{A1} t_{C0} + t_{A0} t_{C1} \right)}{\Delta_0},$$

(40)

with

$$t_{A1} = \chi_{cm}^x e_{cm0}^x + e_{cm0}^y \chi_{cm}^y e_{cm0}^z + e_{cm0}^z \chi_{cm}^z e_{cm0}^x + e_{cm0}^x \chi_{cm}^x e_{cm0}^y,$$

$$t_{B1} = \text{tr} \left( \chi \cdot \underline{A} \right) - \left[ \text{tr} \left( \underline{\chi} \right) \text{tr} \left( \underline{A} \right) \right],$$

$$t_{C1} = \text{tr} \left( \underline{\chi} \cdot \underline{A} \right) \text{tr} \left( \underline{A} \right) \right),$$

(41)

and

$$\underline{\chi} = \text{diag} \left( e_{cm0}^x \chi_{cm0}^x + e_{cm0}^y \chi_{cm0}^y + e_{cm0}^z \chi_{cm0}^z, \chi_{cm}^x e_{cm0}^x + e_{cm0}^y \chi_{cm}^y + e_{cm0}^z \chi_{cm}^z, \chi_{cm}^x e_{cm0}^y + e_{cm0}^z \chi_{cm}^z \right).$$

(42)

4 SPFT ESTIMATE OF HCM PERMITTIVITY

Now that the linear and nonlinear contributions to the depolarization dyadic have been established for depolarization regions of nonzero volume, we can amalgamate these expressions with the SPFT
for weakly nonlinear anisotropic dielectric HCMs — which is presented elsewhere [10] — and thereby estimate the HCM permittivity.

As a precursor, an estimate of permittivity dyadic of the comparison medium must first be computed. The Bruggeman homogenization formalism (which is, in fact, equivalent to the lowest–order SPFT [5]) is used for this purpose. Thus, \( \varepsilon_{cm} \) is found by solving the nonlinear equations

\[
f_a X_{a j} + f_b X_{b j} = 0, \quad (j = 0, 1),
\]

where

\[
X_{\ell 0} = -i \omega \left( \varepsilon_{\ell 0} I - \varepsilon_{cm0} \right) \cdot \Gamma_{\ell 0}^{-1},
\]

\[
X_{\ell 1} = -i \omega \left[ \left( g_\ell \chi_{\ell} I - \chi_{cm} \right) \cdot \Gamma_{\ell 0}^{-1} + \left( \varepsilon_{\ell 0} I - \varepsilon_{cm0} \right) \cdot \Lambda_{\ell} \right],
\]

are the linear and nonlinear parts, respectively, of the corresponding polarizability dyadics. Herein,

\[
\Lambda_{\ell} = \frac{1}{\det \left( \Gamma_{\ell 0} \right)} \left[ \text{diag} \left( \Gamma_{\ell 1}^{x}, \Gamma_{\ell 0}^{x}, \Gamma_{\ell 1}^{y}, \Gamma_{\ell 0}^{y}, \Gamma_{\ell 1}^{z}, \Gamma_{\ell 0}^{z} \right) - \rho_{\ell} \Gamma_{\ell 0}^{-1} \right],
\]

with

\[
\rho_{\ell} = \Gamma_{\ell 0}^{x} \Gamma_{\ell 1}^{y} \Gamma_{\ell 0}^{z} + \Gamma_{\ell 0}^{x} \Gamma_{\ell 1}^{y} \Gamma_{\ell 0}^{z} + \Gamma_{\ell 1}^{x} \Gamma_{\ell 0}^{y} \Gamma_{\ell 1}^{z},
\]

are expressed in terms of components of the dyadics

\[
\Gamma_{\ell 0} = I + i \omega D_0 \cdot \left( \varepsilon_{\ell 0} I - \varepsilon_{cm0} \right) = \text{diag} \left( \Gamma_{\ell 0}^{x}, \Gamma_{\ell 0}^{y}, \Gamma_{\ell 0}^{z} \right),
\]

\[
\Gamma_{\ell 1} = i \omega \left[ D_0 \cdot \left( g_\ell \chi_{\ell} I - \chi_{cm} \right) + D_1 \cdot \left( \varepsilon_{\ell 0} I - \varepsilon_{cm0} \right) \right] = \text{diag} \left( \Gamma_{\ell 1}^{x}, \Gamma_{\ell 1}^{y}, \Gamma_{\ell 1}^{z} \right); \quad (\ell = a, b),
\]

and the local field factor is estimated by [28]

\[
g_\ell = \left| \frac{1}{3} \left[ \text{tr} \left( \Gamma_{\ell 0}^{-1} \right) \right] \right|^2.
\]

Estimates of the \( \varepsilon_{cm0} \) and \( \chi_{cm} \) may be straightforwardly extracted from (43) by recursive schemes; see [10] for details.

Finally, the bilocally–approximated SPFT estimate of the HCM permittivity dyadic, namely

\[
\varepsilon_{\Omega} = \varepsilon_{\Omega 0} + \chi_{\Omega} \mid E_{HCM} \mid^2 = \text{diag} \left( \varepsilon_{\Omega 0}^{x}, \varepsilon_{\Omega 0}^{y}, \varepsilon_{\Omega 0}^{z} \right) + \text{diag} \left( \chi_{\Omega}^{x}, \chi_{\Omega}^{y}, \chi_{\Omega}^{z} \right) \mid E_{HCM} \mid^2,
\]

is given as [5]

\[
\varepsilon_{\Omega 0} = \varepsilon_{cm0} - \frac{1}{i \omega} \frac{Q^{-1} \cdot \Sigma_0}{\Pi \cdot \Sigma_0},
\]

\[
\chi_{\Omega} = \chi_{cm} - \frac{1}{i \omega} \left( Q^{-1} \cdot \Sigma_1 + \Pi \cdot \Sigma_0 \right),
\]
Herein, the linear and nonlinear parts of the mass operator are represented, respectively, by the dyadics
\[
\Sigma_0 = \frac{f_a f_b}{4 \pi \omega} \left( \begin{array}{c}
X_{a0} - X_{b0} \end{array} \right) \cdot W_0(L) \cdot \left( \begin{array}{c}
X_{a0} - X_{b0} \end{array} \right)
\]
\[
\Sigma_1 = \frac{f_a f_b}{4 \pi \omega} \left[ 2 \left( \begin{array}{c}
X_{a0} - X_{b0} \end{array} \right) \cdot W_0(L) \cdot \left( \begin{array}{c}
X_{a1} - X_{b1} \end{array} \right)
\right] + \left( \begin{array}{c}
X_{a0} - X_{b0} \end{array} \right) \cdot W_1(L) \cdot \left( \begin{array}{c}
X_{a0} - X_{b0} \end{array} \right) \right];
\] (51)
and the dyadic
\[
\Pi = \frac{1}{\det (Q_{00})} \left[ \text{diag} \left( Q_0^0 Q_0^0 + Q_0^0 Q_1^x, Q_1^x Q_0^0 + Q_0^0 Q_1^x, Q_1^y Q_0^0 + Q_0^0 Q_1^y \right) \right] - \nu Q_0^{-1},
\] (52)
with
\[
\nu = Q_0^0 Q_0^0 Q_1^x + Q_0^0 Q_0^0 Q_0^x + Q_1^x Q_0^0 Q_0^x,
\] (53)
is expressed in terms of the components of
\[
Q_{00} = I + \Sigma_0 \cdot D_{00} = \text{diag} (Q_0^0, Q_0^0, Q_0^0)
\]
\[
Q_{11} = \Sigma_0 \cdot D_{11} + \Sigma_1 \cdot D_{00} = \text{diag} (Q_1^x, Q_1^y, Q_1^z)
\] (54)

5 NUMERICAL STUDIES

The SPFT estimates (50) of the HCM linear permittivity and nonlinear susceptibility are represented by mathematically complicated expressions. In order to discern the influence of the size parameter \( \eta \), parametric numerical studies are called for. To this end, we investigate the following representative example of a homogenization scenario. Let component phase \( a \) be a cubically nonlinear material with linear permittivity \( \varepsilon_{a0} = 2 \varepsilon_0 \) and nonlinear susceptibility \( \chi_a = 9.07571 \times 10^{-12} \varepsilon_0 \text{m}^2 \text{V}^{-2} \equiv 6.5 \times 10^{-4} \text{esu} \); and component phase \( b \) be a linear material with permittivity \( \varepsilon_b \equiv \varepsilon_{b0} = 12 \varepsilon_0 \). The eccentricities of the ellipsoidal component phase particles are specified by \( U_x = 1 \), \( U_y = 3 \) and \( U_z = 15 \). These choices of parameter values facilitate direct comparisons with a previous investigation in which the effects of the size parameter \( \eta \) were not included [10]. Results are presented for an angular frequency of \( \omega = \pi \times 10^{15} \text{rad s}^{-1} \) (equivalent to a free–space wavelength of 600 nm).

We begin with the relatively straightforward case where neither the size parameter nor the correlation length is taken into account; i.e. \( \eta = L = 0 \). In this case, the SPFT estimates of the constitutive parameters are equivalent to those of the conventional Bruggeman formalism for weakly nonlinear, anisotropic, dielectric HCMs [28]. In Fig. 1, the HCM linear and nonlinear constitutive parameters are plotted against volume fraction \( f_a \). The HCM linear permittivity parameters \( \varepsilon_{a0} \) uniformly decrease from \( \varepsilon_{a0} \) at \( f_a = 0 \) to \( \varepsilon_{a0} \) at \( f_a = 1 \). In contrast, the HCM nonlinear susceptibility parameter \( \chi_{1x}^x \), and to a lesser extent \( \chi_{1y}^x \), exceeds the nonlinear susceptibility of component phase \( a \) for a wide range of values of \( f_a \). This nonlinearity enhancement phenomenon, and its potential for technological exploitation, have been reported on previously for both isotropic [7, 9, 29, 30]
Figure 1: The HCM relative linear permittivity and nonlinear susceptibility parameters plotted against $f_a$, calculated for $\eta = L = 0$. Key: $\epsilon_{f_0\Omega}^x/\epsilon_0$ and $\chi_{\Omega a}^x$ dashed curves; $\epsilon_{f_0\Omega}^y/\epsilon_0$ and $\chi_{\Omega a}^y$ broken dashed curves; and $\epsilon_{f_0\Omega}^z/\epsilon_0$ and $\chi_{\Omega a}^z$ solid curves. Component phase parameter values: $\epsilon_{a0} = 2\epsilon_0$, $\chi_a = 9.07571 \times 10^{-12} \epsilon_0$ m$^2$V$^{-2}$, $\epsilon_b \equiv \epsilon_{b0} = 12\epsilon_0$, $U_x = 1$, $U_y = 3$ and $U_z = 15$.

Figure 2: Real and imaginary parts of the HCM linear permittivity and nonlinear susceptibility parameters plotted against $\eta$ (in nm), calculated for $L = 0$ and $f_a = 0.3$. Key: $\epsilon_{f_{0r}\Omega}^x$ and $\chi_{\Omega r}^x$ dashed curves; $\epsilon_{f_{0r}\Omega}^y$ and $\chi_{\Omega r}^y$ broken dashed curves; and $\epsilon_{f_{0r}\Omega}^z$ and $\chi_{\Omega r}^z$ solid curves. Component phase parameter values as in Fig. 1.

How does the size parameter $\eta$ influence the estimates of the HCM constitutive parameters? To answer this question, we fix the volume fraction at $f_a = 0.3$ and calculate the HCM constitutive parameters for $0 < \eta < 20$ nm with $L = 0$. The presentation of results is aided by the introduction and anisotropic [10, 28] HCMs. The anisotropy reflected by the constitutive parameters, and the nonlinearity enhancement, stems from the ellipsoidal geometry of the component phase particles.
of the relative constitutive parameters

\[
\begin{align*}
\epsilon^n_{\Omega_0 r} &= \frac{\epsilon^n_{\Omega_0} - (\epsilon^n_{\Omega_0}|_{\eta=L=0})}{\epsilon_0}, \\
\chi^n_{\Omega r} &= \frac{\chi^n_{\Omega} - (\chi^n_{\Omega}|_{\eta=L=0})}{\chi_0}
\end{align*}
\]

\[ (n = x, y, z), \tag{55} \]

which measure the difference between the SPFT estimates calculated for \( \eta, L \neq 0 \) and \( \eta = L = 0 \). The results are plotted in Fig. 2. It is notable that the HCM constitutive parameters have nonzero imaginary parts whereas the component material phases are specified by real–valued constitutive parameters. As previously described for linear HCMs [20], the presence of nonzero imaginary parts for the HCM constitutive parameters may be attributed to radiative scattering losses associated with the nonzero size of the component phase particles. Plainly, increasing the size parameter \( \eta \) has the effect of increasing the real and imaginary parts of the HCM linear permittivity, but decreasing the real and imaginary parts of the HCM nonlinear susceptibility. In fact, the influence of the size parameter is very similar to the influence of the correlation length, as has been noted for linear HCMs [10].

![Figure 3: Real and imaginary parts of the HCM linear permittivity and nonlinear susceptibility parameters \( \epsilon^{\Omega}_{\Omega_0 r} \) and \( \chi^{\Omega}_{\Omega r} \) plotted against \( L \) (in nm) and \( \eta/L \), calculated for \( f_a = 0.3 \). Component phase parameter values as in Fig. 1.](image-url)
Fig. 2 reveals that by taking into consideration the nonzero size of the component phase particles — but not the correlation length — the predicted nonlinearity enhancement is somewhat diminished. We now consider the estimates of the HCM constitutive parameters when both the size parameter and the correlation length are taken into account. In Fig. 3, the HCM relative constitutive parameters are plotted against both $L$ and $\eta/L$ with the volume fraction fixed at $f_a = 0.3$. Only the results for $\varepsilon_\Omega^{x}$ and $\chi_\Omega^{x}$ are presented; the corresponding plots for $\varepsilon_\Omega^{y,z}$ and $\chi_\Omega^{y,z}$ are similar. It may be observed in Fig. 3 that the effects of $\eta$ and $L$ are cumulative insofar as the increase in the real and imaginary parts of $\varepsilon_\Omega^{x}$ and $\chi_\Omega^{x}$, and the decrease in the real and imaginary parts of $\varepsilon_\Omega^{y,z}$ and $\chi_\Omega^{y,z}$, which occur as $\eta$ increases, become steadily more exaggerated as $L$ increases.

6 CONCLUDING REMARKS

The size of the component phase particles can have a significant bearing upon the estimated constitutive parameters of weakly nonlinear anisotropic HCMs, within the bilocally–approximated SPFT. Most obviously, by taking nonzero particle size into consideration, attenuation is predicted and the degree of nonlinearity enhancement is somewhat diminished. In respect of both of these effects, the influence of particle size is similar to the influence of correlation length. Furthermore, the effects of particle size and correlation length on both the linear and nonlinear HCM constitutive parameters are found to be cumulative.

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