Geometrically-Derived Anisotropy in Cubically Nonlinear Dielectric Composites

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Abstract

We consider an anisotropic homogenized composite medium (HCM) arising from isotropic particulate component phases based on ellipsoidal geometries. For cubically nonlinear component phases, the corresponding zeroth-order strong-permittivity-fluctuation theory (SPFT) (which is equivalent to the Bruggeman homogenization formalism) and second-order SPFT are established and used to estimate the constitutive properties of the HCM. The relationship between the component phase particulate geometry and the HCM constitutive properties is explored. Significant differences are highlighted between the estimates of the Bruggeman homogenization formalism and the second-order SPFT estimates. The prospects for nonlinearity enhancement are investigated.

1 Introduction

The constitutive properties of a homogenized composite medium (HCM) are determined by both the constitutive properties and the topological properties of its component phases [1]–[5]. In particular, component phases based on nonspherical particulate geometries may give rise to anisotropic HCMs, despite the component phases themselves being isotropic with respect to their electromagnetic properties. Such geometrically-derived anisotropy has been extensively characterized for linear dielectric HCMs [6]–[8] and more general bianisotropic HCMs [8]–[10]. For weakly nonlinear HCMs, the role of the component phase particulate geometry was emphasized recently in this journal by Goncharenko, Popelnukh and Venger [11], using an approach founded on the mean-field approximation. However, their analysis was restricted to the Maxwell Garnett homogenization formalism [3, 12]. A more comprehensive study is communicated here based on the strong-permittivity-fluctuation theory (SPFT) [13]. In contrast to the aforementioned Maxwell Garnett approach [11], the SPFT approach (i) incorporates higher-order statistics to describe the component phase dis-

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tributions; (ii) is not restricted to only dilute composites; and (iii) is not restricted to only weakly nonspherical particulate geometries.

The early development of the SPFT concerned wave propagation in continuous random mediums [14, 15], but more recently the theory has been applied to the estimation of HCM constitutive parameters [16, 17]. The SPFT represents a significant advance over conventional homogenization formalisms, such as the Maxwell Garnett approach and the Bruggeman approach [3, 6], through incorporating a comprehensive description of the distributional statistics of the HCM component phases. In estimating the constitutive parameters of an HCM, the SPFT employs a Feynman–diagrammatic technique to calculate iterative refinements to the constitutive parameters of a comparison medium; successive iterates incorporate successively higher-order spatial correlation functions. It transpires that the SPFT comparison medium is equivalent to the effective medium of the (symmetric) Bruggeman homogenization theory [20, 21]. In principle, correlation functions of arbitrarily high order can be accommodated in the SPFT. However, the theory is most widely-implemented at the level of the bilocal approximation (i.e., second-order approximation), wherein a two-point covariance function and its associated correlation length \( L \) characterize the component phase distributions. As indicated in figure 1, coherent interactions between pairs of scattering centres within a region of linear dimensions \( L \) are incorporated in the bilocal SPFT; scattering centres separated by distances much greater than \( L \) are assumed to act independently. Thereby, the SPFT provides an estimation of coherent scattering losses, unlike the Maxwell Garnett and Bruggeman homogenization formalisms. Notice that the bilocally-approximated SPFT gives rise to the Bruggeman homogenization formalism in the limit \( L \to 0 \) [21].

The SPFT has been widely applied to linear homogenization scenarios, where generalizations\(^2\) have been developed for anisotropic dielectric [18, 19], isotropic chiral [20] and bianisotropic [21, 22] HCMs. Investigations of the trilocally-approximated SPFT for isotropic HCMs have recently confirmed the convergence of the second-order theory [17, 23, 24]. In the weakly nonlinear regime, developments of the bilocally-approximated SPFT have been restricted to isotropic HCMs, based on spherical component phase geometry [16, 17, 24]. The present study advances the nonlinear SPFT through developing the theory for cubically nonlinear, anisotropic HCMs. Furthermore, it is assumed that the component phases are composed of electrically-small ellipsoidal particles. The relationship between the HCM constitutive parameters and the underlying particulate geometry of the component phases is investigated via a representative numerical example.

In our notational convention, dyadics are double underlined whereas vectors are in bold face. The inverse, adjoint, determinant and trace of a dyadic \( \underline{A} \) are denoted by \( \underline{A}^{-1} \), \( \underline{A}^{\text{adj}} \), \( \det \left[ \underline{A} \right] \) and \( \text{tr} \left[ \underline{A} \right] \), respectively. The identity dyadic is represented by \( \underline{I} \). The ensemble average of a quantity \( \psi \) is written as \( \langle \psi \rangle \). The permittivity and permeability of free space (i.e., vacuum) are given by \( \varepsilon_0 \) and \( \mu_0 \), respectively; \( k_0 = \omega \sqrt{\varepsilon_0 \mu_0} \) is the free-space wavenumber while \( \omega \) is the angular frequency.

\(^2\)The generalized SPFT is referred to as the strong-property-fluctuation theory.
2 Homogenization generalities

2.1 Component phases

Consider the homogenization of a two-phase composite with component phases labelled as \( a \) and \( b \). The component phases are taken to be isotropic dielectric mediums with permittivities

\[
\epsilon_\ell = \epsilon_{\ell 0} + \chi_\ell |E_\ell|^2, \quad (\ell = a, b),
\]

(1)

where \( \epsilon_{\ell 0} \) is the linear permittivity, \( \chi_\ell \) is the nonlinear susceptibility, and \( |E_\ell|^2 \) is the electric field developed inside a region of phase \( \ell \) by illumination of the composite medium. We assume weak nonlinearity; i.e., \( |\epsilon_{\ell 0}| \gg |\chi_\ell| |E_\ell|^2 \). Notice that such electrostrictive mediums as characterized by (1) can induce Brillouin scattering which is often a strong process [25]. The component phases \( a \) and \( b \) are taken to be randomly distributed as identically-oriented, conformal ellipsoids. The shape dyadic

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

(2)

parameterizes the conformal ellipsoidal surfaces as

\[
\mathbf{r}_e(\theta, \phi) = \eta U \mathbf{\hat{r}}(\theta, \phi),
\]

(3)

where \( \mathbf{\hat{r}}(\theta, \phi) \) is the radial unit vector specified by the spherical polar coordinates \( \theta \) and \( \phi \). Thus, a wide range of ellipsoidal particulate shapes, including highly elongated forms, can be accommodated. The linear ellipsoidal dimensions, as determined by \( \eta \), are assumed to be sufficiently small that the electromagnetic long-wavelength regime pertains.

In the SPFT, statistical moments of the characteristic functions

\[
\Phi_\ell(\mathbf{r}) = \begin{cases} 
1, & \mathbf{r} \in V_\ell, \\
0, & \mathbf{r} \notin V_\ell,
\end{cases} \quad (\ell = a, b),
\]

(4)

are utilized to take account of the component phase distributions. The volume fraction of phase \( \ell \), namely \( f_\ell \), is given by the first statistical moment of \( \Phi_\ell \); i.e., \( \langle \Phi_\ell(\mathbf{r}) \rangle = f_\ell \). Clearly, \( f_a + f_b = 1 \). The second statistical moment of \( \Phi_\ell \) provides a two-point covariance function; we adopt the physically-motivated form [26]

\[
\langle \Phi_\ell(\mathbf{r}) \Phi_\ell(\mathbf{r}') \rangle = f_\ell [1 + (f_\ell - 1) \mathcal{H}(\sigma - L) ],
\]

(5)

where \( \mathcal{H} \) is the Heaviside function (i.e., \( \mathcal{H}(x) = \int_{-\infty}^{x} \delta(y) dy \) where \( \delta \) is the Dirac delta function), \( \sigma = |U^{-1} \cdot \mathbf{R}| \) with \( \mathbf{R} = \mathbf{r} - \mathbf{r}' \), and \( L > 0 \) is the correlation length. The specific nature of the covariance function has been found to exert little influence on the SPFT estimates for linear [19] and weakly nonlinear [17] HCMs.

2.2 Homogenized composite medium

Let \( \mathbf{E}_{HCM} \) denote the spatially-averaged electric field in the HCM. In this communication we derive the estimate

\[
\xi_{ba} = \xi_{ba0} + \chi_{ba} |\mathbf{E}_{HCM}|^2
\]

(6)

\[
= \text{diag} (\epsilon_{ba0}^x, \epsilon_{ba0}^y, \epsilon_{ba0}^z) + \text{diag} (\chi_{ba}^x, \chi_{ba}^y, \chi_{ba}^z) |\mathbf{E}_{HCM}|^2
\]

(7)
of the HCM permittivity. The bilocally-approximated SPFT is utilized (hence the subscripts \( \text{ba} \) in (6), (7)). Note that the Bruggeman estimate of the HCM permittivity, namely

\[
\varepsilon_{\text{Br}} = \varepsilon_{\text{Br}0} + \chi_{\text{Br}} |\mathbf{E}_{\text{HCM}}|^2
\]

characterizes the comparison medium which is adopted in the bilocally-approximated SPFT [21]. As the Bruggeman homogenization formalism — in which the component phases \( a \) and \( b \) are treated symmetrically [6] — provides the comparison medium, the SPFT homogenization approach (like the Bruggeman formalism) is applicable for all volume fractions \( f_a \in (0, 1) \).

2.3 Depolarization and polarizability dyadics

The depolarization dyadic \( \mathbb{D} \) is a key element in both Bruggeman and SPFT homogenizations. It provides the electromagnetic response of a \( U \)-shaped exclusion volume, immersed in a homogeneous background, in the limit \( \eta \to 0 \). For the component phases described by (1) and (2), we find [27, 28]

\[
\mathbb{D} = \frac{1}{i \omega 4 \pi} \int_{0}^{2\pi} d\phi \int_{0}^{\pi} d\theta \sin \theta \left( \frac{1}{\text{tr}[\varepsilon_{\text{Br}} \cdot \mathbb{A}]} \right) \mathbb{A},
\]

wherein

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

The integrations of (10) reduce to elliptic function representations [29]. In the case of spheroidal particulate geometries, hyperbolic functions provide an evaluation of \( \mathbb{D} \) [27], while for the degenerate isotropic case \( U_x = U_y = U_z \) we have the well-known result \( \mathbb{D} = (1/3i \omega) \varepsilon_{\text{Br}}^{-1} \) [30]. We express \( \mathbb{D} \) as the sum of linear and weakly nonlinear parts

\[
\mathbb{D} = \mathbb{D}_0 + \mathbb{D}_1 |\mathbf{E}_{\text{HCM}}|^2,
\]

with

\[
\mathbb{D}_0 = \frac{1}{i \omega 4 \pi} \int_{0}^{2\pi} d\phi \int_{0}^{\pi} d\theta \sin \theta \left( \frac{1}{\text{tr}[\varepsilon_{\text{Br}0} \cdot \mathbb{A}]} \right) \mathbb{A}
\]

\[
\mathbb{D}_1 = -\frac{1}{i \omega 4 \pi} \int_{0}^{2\pi} d\phi \int_{0}^{\pi} d\theta \sin \theta \left[ \frac{\text{tr}[\chi_{\text{Br}0} \cdot \mathbb{A}]}{\text{tr}[\varepsilon_{\text{Br}0} \cdot \mathbb{A}]} \right]^2 \mathbb{A}.
\]

A convenient construction in homogenization formalisms is the polarizability dyadic \( \mathbb{X} \), defined as

\[
\mathbb{X}_\ell = -i \omega \left( \epsilon_I - \varepsilon_{\text{Br}} \right) \cdot \Gamma_{\ell}^{-1}, \quad (\ell = a, b),
\]

where

\[
\Gamma_{\ell} = \left[ \mathbb{I} + i \omega \mathbb{D}_0 \epsilon_I \left( \epsilon_I - \varepsilon_{\text{Br}} \right) \right].
\]

Let us proceed to calculate the linear and nonlinear contributions in the decomposition

\[
\mathbb{X}_\ell = \mathbb{X}_{\ell 0} + \mathbb{X}_{\ell 1} |\mathbf{E}_{\text{HCM}}|^2, \quad (\ell = a, b).
\]
Under the assumption of weak nonlinearity, we express (16) in the form
\[ \Gamma_\ell = \Gamma_\ell^0 + \Gamma_\ell^1 |E_{HCM}|^2, \] (18)
with linear term
\[ \Gamma_\ell^0 = \text{diag} (\Gamma_x^\ell_0, \Gamma_y^\ell_0, \Gamma_z^\ell_0) = I + i\omega D_0 \cdot \left( \epsilon_\ell^0 I - \epsilon_{Br0} \right), \] (19)
and nonlinear term
\[ \Gamma_\ell^1 = \text{diag} (\Gamma_x^\ell_1, \Gamma_y^\ell_1, \Gamma_z^\ell_1) = i\omega \left[ D_0 \cdot \left( g_\ell \chi I - \chi_{Br} \right) + D_1 \cdot \left( \epsilon_\ell^0 I - \epsilon_{Br0} \right) \right]. \] (20)
The local field factor
\[ g_\ell = \frac{d |E_\ell|^2}{d |E_{HCM}|^2}, \] (\ell = a, b),
has been incorporated in deriving (18)-(20), via the Maclaurin series expansion \( \epsilon_\ell = \epsilon_\ell^0 + g_\ell \chi I |E_{HCM}|^2. \)
An appropriate estimation of the local field factor is provided by [31]
\[ g_\ell = \left| \frac{1}{3} \left( \text{tr} \left[ \Gamma_\ell^{-1} \right] \right) \right|^2. \] (22)
Thus, the inverse of \( \Gamma_\ell \) is given as
\[ \Gamma_\ell^{-1} = \Gamma_\ell^{-1} + \Lambda_\ell |E_{HCM}|^2, \] (23)
wherein
\[ \Lambda_\ell = \frac{1}{\det \left[ \Gamma_\ell^1 \right]} \left[ \text{diag} \left( \Gamma_y^\ell_1 \Gamma_z^\ell_0 + \Gamma_y^\ell_0 \Gamma_z^\ell_1, \Gamma_z^\ell_1 \Gamma_x^\ell_0 + \Gamma_z^\ell_0 \Gamma_x^\ell_1, \Gamma_x^\ell_1 \Gamma_y^\ell_0 + \Gamma_x^\ell_0 \Gamma_y^\ell_1 \right) - \rho_\ell \Gamma_\ell^1 \right], \] (24)
and
\[ \rho_\ell = \Gamma_x^\ell_0 \Gamma_y^\ell_0 \Gamma_z^\ell_1 + \Gamma_x^\ell_1 \Gamma_y^\ell_0 \Gamma_z^\ell_0 + \Gamma_x^\ell_1 \Gamma_y^\ell_0 \Gamma_z^\ell_0 . \] (25)
Combining (23) and (24) with (15), and separating linear and nonlinear terms, provides
\[ X_\ell^0 = -i \omega \left( \epsilon_\ell^0 I - \epsilon_{Br0} \right) \cdot \Gamma_\ell^{-1} \]
\[ X_\ell^1 = -i \omega \left[ (g_\ell \chi I - \chi_{Br}) \cdot \Gamma_\ell^{-1} + \left( \epsilon_\ell^0 I - \epsilon_{Br0} \right) \cdot \Lambda_\ell \right], \] \( \ell = a, b. \) (26)

### 2.4 Bruggeman homogenization

The Bruggeman estimates of the HCM linear permittivity \( \epsilon_{Br0} \) and nonlinear susceptibility \( \chi_{Br} \) are delivered through solving the nonlinear equations [3, 6, 31]
\[ f_o X_{aj} + f_b X_{bj} = 0, \] (27)
\( j = 0, 1. \)
Recursive procedures for this purpose provide the \( p^{th} \) iterates \([4, 24]\)

\[
\begin{align*}
\xi_{ba0}[p] &= T_\varepsilon \left\{ \xi_{ba0}[p-1] \right\} \\
\chi_{ba}[p] &= T_\chi \left\{ \chi_{ba}[p-1] \right\}
\end{align*}
\] (28)

in terms of the \((p-1)^{th}\) iterates, wherein the operators \( T_\varepsilon, T_\chi \) are defined by

\[
\begin{align*}
T_\varepsilon \left\{ \xi_{ba0} \right\} &= \left( f_a \varepsilon_{a0} \Gamma_{a0}^{-1} + f_b \varepsilon_{b0} \Gamma_{b0}^{-1} \right) \cdot \left( f_a \xi_{a0} + f_b \xi_{b0} \right)^{-1}, \\
T_\chi \left\{ \chi_{ba0} \right\} &= \left\{ f_a \left[ \varepsilon_{a0} \Gamma_{a0}^{-1} + \left( \varepsilon_{a0} I - \xi_{ba0} \right) \cdot \Lambda_a \right] + f_b \left[ \varepsilon_{b0} \Gamma_{b0}^{-1} + \left( \varepsilon_{b0} I - \xi_{ba0} \right) \cdot \Lambda_b \right] \right\} \\
&\quad \cdot \left( f_a \xi_{a0} + f_b \xi_{b0} \right)^{-1},
\end{align*}
\] (29)

while suitable initial values are given by

\[
\begin{align*}
\xi_{ba0}[0] &= (f_a \varepsilon_{a0} + f_b \varepsilon_{b0}) \frac{I}{L} \\
\chi_{ba}[0] &= (f_a \chi_{a0} + f_b \chi_{b0}) \frac{I}{L}
\end{align*}
\] (30)

### 3 The bilocally-approximated SPFT

The bilocally-approximated SPFT estimate of the HCM permittivity dyadic, as derived elsewhere [21], is given by

\[
\xi_{ba} = \xi_{Br} - \frac{1}{i \omega} \left( \frac{L}{L} + \sum_{ba} \cdot \mathbb{D} \right)^{-1} \cdot \sum_{ba};
\] (31)

the mass operator term

\[
\sum_{ba} = \left( X_a - X_b \right) \cdot \mathbb{W} \cdot \left( X_a - X_b \right)
\] (32)

is specified in terms of the principal value integral

\[
\mathbb{W} = \mathcal{P} \int_{\sigma \leq L} d^3 R \ G_{Br}(R),
\] (33)

with \( G_{Br}(R) \) being the unbounded dyadic Green function of the comparison medium. Here we develop expressions for the linear and nonlinear contributions of the mass operator, appropriate to the component phases specified in §2.

Under the assumption of weak nonlinearity, we express \( \mathbb{W} = \mathbb{W}_0 + \mathbb{W}_1 |\mathbf{E}_{HCM}|^2 \); integral expressions for \( \mathbb{W}_0 \) and \( \mathbb{W}_1 \) are provided in the Appendix. Thereby, the linear and nonlinear terms in the mass operator decomposition \( \sum_{ba} = \sum_{ba0} + \sum_{ba1} |\mathbf{E}_{HCM}|^2 \) are given as

\[
\sum_{ba0} = \left( X_a - X_b \right) \cdot \mathbb{W}_0 \cdot \left( X_a - X_b \right),
\] (34)

\[
\sum_{ba1} = 2 \left( X_a - X_b \right) \cdot \mathbb{W}_0 \cdot \left( X_a - X_b \right) + \left( X_a - X_b \right) \cdot \mathbb{W}_1 \cdot \left( X_a - X_b \right),
\] (35)

respectively, correct to the second order in \( |\mathbf{E}_{HCM}| \). Now, let us introduce the dyadic quantity

\[
\mathbb{\Omega} = I + \sum_{ba} \cdot \mathbb{D} = \mathbb{\Omega}_0 + \mathbb{\Omega}_1 |\mathbf{E}_{HCM}|^2,
\] (36)
such that

\[
\begin{align*}
\Omega_0 &= \text{diag} (\Omega_0^x, \Omega_0^y, \Omega_0^z) = I + \sum_{ba0} \cdot D_{ba0}, \\
\Omega_1 &= \text{diag} (\Omega_1^x, \Omega_1^y, \Omega_1^z) = \sum_{ba0} \cdot D_{ba0} + \sum_{ba1} \cdot D_{ba1}.
\end{align*}
\] (37) (38)

We may then express the inverse dyadic in the form

\[
\Omega^{-1} = \Omega_{0}^{-1} + \Pi |E_{HCM}|^2,
\] (39)

with nonlinear part

\[
\Pi = \frac{1}{\det(\Omega_0)} \left[ \text{diag} \left( \Omega_0^x \Omega_0^y, \Omega_0^y \Omega_0^z, \Omega_0^z \Omega_0^x + \Omega_0^y \Omega_0^z \right) - \nu \Omega_0^{-1} \right],
\] (40)

where

\[
\nu = \Omega_0^x \Omega_0^y \Omega_0^z + \Omega_0^y \Omega_0^z \Omega_0^x + \Omega_0^z \Omega_0^x \Omega_0^y.
\] (41)

Thus, the linear and nonlinear contributions of the SPFT estimate \( \varepsilon_{ba} \) are delivered, respectively, as

\[
\varepsilon_{ba0} = \varepsilon_{Br0} - \frac{1}{i \omega} \Omega_0^{-1} \cdot \sum_{ba0},
\] (42)

\[
\varepsilon_{ba} = \chi_{Br} - \frac{1}{i \omega} \left( \Omega_0^{-1} \cdot \sum_{ba1} + \Pi \cdot \sum_{ba0} \right).
\] (43)

4 Numerical results and discussion

Let us explore the HCM constitutive parameter space by means of a representative numerical example: Consider the homogenization of a cubically nonlinear phase \( a \) with linear permittivity \( \varepsilon_{a0} = 2 \varepsilon_0 \) and nonlinear susceptibility \( \chi_a \equiv 9 \times 10^{-12} \varepsilon_0 \) m² V⁻² (\( \equiv 6.5 \times 10^{-4} \) esu) and a linear phase \( b \) with permittivity \( \varepsilon_{b0} = 12 \varepsilon_0 \). Note that the selected nonlinear susceptibility value corresponds to that of gallium arsenide [25], while selected the linear permittivity values are typical of a wide range of insulating crystals [32]. We assume the ellipsoidal component phase topology specified by \( U_x = 1, U_y = 3 \) and \( U_z \in [0.5, 15] \). The angular frequency \( \omega \) is fixed at \( 2\pi \times 10^{10} \) rad s⁻¹ for all calculations reported here.

The Bruggeman estimates of the HCM relative linear and nonlinear constitutive parameters are plotted in figure 1 as functions of \( f_a \) and \( U_z \). The calculated constitutive parameters presented in figure 1 are consistent with those calculated by Lakhtakia and Lakhtakia [31] in a study pertaining to the Bruggeman homogenization of ellipsoidal inclusions with a host medium comprising spherical particles. The linear parameters follow an approximately linear progression between their constraining values at \( f_a = 0 \) and \( f_a = 1 \). Furthermore, for the range \( U_z \in [0.5, 15] \), the linear parameters are largely (but not completely) independent of the particulate geometry of the component phases. This is in contrast to the nonlinear parameters which are acutely sensitive to \( U_z \). Of special significance is the nonlinearity enhancement (i.e., the manifestation of a higher degree of nonlinear susceptibility in the HCM than is present in its component phases) which is particularly observed at high values of \( U_z \) for \( \chi_{Br} \) and at low values of \( U_z \) for \( \chi_{Br} \). This phenomenon and its possible technological exploitation are described elsewhere [3, 16, 17, 24, 31, 33]. In order
to best consider nonlinearity enhancement, we fix the shape parameter \( U_z = 15 \) for all remaining calculations.

We turn our attention now to the bilocally-approximated SPFT calculations. Let

\[
\epsilon_{nrx}^{br0} = \frac{\epsilon^n_{ba0} - \epsilon^n_{Br0}}{\epsilon_0}, \quad \chi_{nrx}^{br0} = \frac{\chi^n_{ba} - \chi^n_{Br}}{\chi_a}, \quad (n = x, y, z).
\]

The SPFT estimates of the HCM relative linear constitutive parameters \( \epsilon_{nrx}^{br0} \) and nonlinear constitutive parameters \( \chi_{nrx}^{br0} \) are plotted in figures 2 and 3, respectively, as functions of \( f_a \) and \( k_0L \). Significant differences are clear between the Bruggeman-estimated values and the SPFT-estimated values: The SPFT estimates of linear constitutive parameters provide an additive correction to the corresponding Bruggeman parameters, whereas for the nonlinear constitutive parameters the SPFT estimates provide a subtractive correction to the corresponding Bruggeman parameters. Furthermore, the magnitudes of these differences exhibit local maxima which occur at progressively higher values of \( f_a \) as one compares the constitutive parameter components aligned with the \( x \), \( y \) and \( z \) coordinate axes, respectively. This trend holds for both the real and the imaginary parts of both the linear permittivity and the nonlinear susceptibility parameters. However, it is less pronounced for the nonlinear constitutive parameters.

Coherent interactions between scattering centres enclosed within a region of linear dimensions \( L \) are accommodated in the bilocally-approximated SPFT via the two-point covariance function (5) (see figure 1). Thus, since neither component phase \( a \) nor component phase \( b \) is dissipative, the nonzero imaginary parts of the SPFT constitutive parameters in figures 2 and 3 are attributable entirely to scattering losses. Furthermore, the magnitudes of the imaginary parts of the constitutive parameters are observed in figures 2 and 3 to increase as \( L \) increases, due to the actions of greater numbers of scattering centres becoming correlated.

### 5 Concluding remarks

The bilocally-approximated SPFT for weakly nonlinear isotropic HCMs, based on spherical particulate geometry, has been recently established [16, 17, 24]. In the present study we further advance the theory through considering anisotropic, cubically nonlinear HCMs, arising from isotropic component phases with ellipsoidal particulate geometries. Significant differences between the bilocally-approximated SPFT (i.e., second-order theory) and the Bruggeman homogenization formalism (i.e., zeroth-order theory) — which depend upon the underlying particulate geometry — have emerged. In particular, nonlinearity enhancement is predicted to a lesser degree with the SPFT than with the Bruggeman homogenization formalism. The importance of taking into account the distributional statistics of the HCM component phases is thereby further emphasized.

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Consider the principal value integral term $\mathbb{W}$ (33) which was expressed as the sum $\mathbb{W}_0 + \mathbb{W}_1 | \mathbf{E}_{HCM} |^2$ in §3. Here we develop expressions for the linear component $\mathbb{W}_0$ and the nonlinear component $\mathbb{W}_1$, appropriate to the homogenization scenario of §2.

Let us begin with the following straightforward specialization of the evaluation of $\mathbb{W}$ for bianisotropic HCMs [21, 22]

$$\mathbb{W} = \frac{f_a f_b}{2 \pi^2 k} \int d^3 \mathbf{q} \frac{(q/\omega)^2 \mathbf{a} + \beta}{(q/\omega)^2 t_C + (q/\omega)^2 t_B + t_A} \left( \frac{\sin qL - L \cos qL}{q} \right). \quad (45)$$

For the weakly nonlinear homogenization outlined in §2, the scalar terms $t_A$, $t_B$ and $t_C$ in (45), along with their linear and nonlinear decompositions, are given by

$$t_A = \mu_0^3 \det \left[ \varepsilon_{Br} \right] = t_{A0} + t_{A1} | \mathbf{E}_{HCM} |^2, \quad (46)$$

$$t_B = \mu_0^2 \left\{ \left[ \varepsilon_{adj} \cdot \mathbf{A} \right] - \left( \varepsilon_{Br0} \right) \mathbf{A} - \left[ \varepsilon_{Br} \cdot \mathbf{A} \right] \right\}, \quad (47)$$

$$t_C = \mu_0 \varepsilon_{Br0} \left[ \varepsilon_{Br} \cdot \mathbf{A} \right] = t_{C0} + t_{C1} | \mathbf{E}_{HCM} |^2, \quad (48)$$

wherein

$$t_{A0} = \mu_0^3 \det \left[ \varepsilon_{Br0} \right], \quad t_{A1} = \mu_0^3 \left( \varepsilon_{Br0} \varepsilon_{Br} \right) \cdot \varepsilon_{Br0} + \varepsilon_{Br0} \varepsilon_{Br} \cdot \varepsilon_{Br0} + \varepsilon_{Br0} \varepsilon_{Br} \cdot \varepsilon_{Br0} \varepsilon_{Br}, \quad (49)$$

$$t_{B0} = \mu_0^2 \left\{ \left[ \varepsilon_{adj} \cdot \mathbf{A} \right] - \left( \varepsilon_{Br0} \right) \mathbf{A} - \left[ \varepsilon_{Br} \cdot \mathbf{A} \right] \right\}, \quad (50)$$

$$t_{B1} = \mu_0^2 \left\{ \left[ \Xi \cdot \mathbf{A} \right] - \left( \varepsilon_{Br0} \right) \mathbf{A} - \left[ \varepsilon_{Br} \cdot \mathbf{A} \right] \right\}, \quad (51)$$

$$\Xi = \varepsilon_{Br0} \varepsilon_{Br} \varepsilon_{Br0} + \varepsilon_{Br0} \varepsilon_{Br} \varepsilon_{Br0} + \varepsilon_{Br0} \varepsilon_{Br} \varepsilon_{Br0} \varepsilon_{Br}, \quad (52)$$

$$t_{C0} = \mu_0 \varepsilon_{Br0} \left[ \varepsilon_{Br} \cdot \mathbf{A} \right], \quad t_{C1} = \mu_0 \varepsilon_{Br0} \left[ \varepsilon_{Br} \cdot \mathbf{A} \right]. \quad (53)$$

Similarly, the dyadic quantities $\underline{\alpha}$ and $\beta$ in (45), along with their linear and nonlinear decompositions, are given by

$$\underline{\alpha} = \mu_0^2 \left( 2 \varepsilon_{Br} - \left[ \varepsilon_{Br} \right] \right) \cdot \mathbf{A} - \mu_0 t_{A0} \mathbf{A} = \underline{\alpha}_0 + \underline{\alpha}_1 | \mathbf{E}_{HCM} |^2, \quad (54)$$

$$\underline{\beta} = \mu_0^3 \varepsilon_{adj} - \mu_0 t_{A0} \mathbf{A} = \underline{\beta}_0 + \underline{\beta}_1 | \mathbf{E}_{HCM} |^2, \quad (55)$$

with

$$\underline{\alpha}_0 = \mu_0^2 \left( 2 \varepsilon_{Br0} - \left[ \varepsilon_{Br0} \right] \right) \cdot \mathbf{A} - \mu_0 t_{B0} \mathbf{A} = \underline{\alpha}_0 + \underline{\alpha}_1 | \mathbf{E}_{HCM} |^2, \quad (56)$$

$$\underline{\alpha}_1 = \mu_0^2 \left( 2 \varepsilon_{Br} - \left[ \varepsilon_{Br} \right] \right) \cdot \mathbf{A} - \mu_0 t_{B1} \mathbf{A} = \underline{\alpha}_0 + \underline{\alpha}_1 | \mathbf{E}_{HCM} |^2, \quad (57)$$

$$\underline{\beta}_0 = \mu_0^3 \varepsilon_{adj} - \mu_0 t_{A0} \mathbf{A} = \underline{\beta}_0 + \underline{\beta}_1 | \mathbf{E}_{HCM} |^2, \quad (58)$$

$$\underline{\beta}_1 = \mu_0^3 \Xi - \mu_0 t_{A1} \mathbf{A} = \underline{\beta}_0 + \underline{\beta}_1 | \mathbf{E}_{HCM} |^2, \quad (59)$$

In the long-wavelength regime, i.e., $|d_\pm| \ll 1$, the application of residue calculus to (45) delivers

$$W = \frac{f_a f_b \omega}{4 \pi i} \int_0^{2\pi} d\phi \int_0^\pi d\theta \frac{\sin \theta}{3 \Delta} \left\{ \frac{1}{\omega^2} \left[ \frac{3}{2} \left( d_+^2 - d_-^2 \right) + i \left( d_+^3 - d_-^3 \right) \right] \tilde{\alpha} - i \left( \frac{d_+^3}{\kappa_+} - \frac{d_-^3}{\kappa_-} \right) \tilde{\beta} \right\},$$

where we have introduced

$$\Delta = \sqrt{t_B^2 - 4t_A t_C} = \Delta_0 + \Delta_1 |E_{HCM}|^2,$$  \hspace{1cm} (60)

$$\kappa_\pm = \omega^2 \frac{t_B \pm \Delta}{2 t_C} = \kappa_{0\pm} + \kappa_{1\pm} |E_{HCM}|^2,$$  \hspace{1cm} (61)

$$d_\pm = L \sqrt{\kappa_\pm} = d_{0\pm} + d_{1\pm} |E_{HCM}|^2,$$  \hspace{1cm} (62)

with linear and nonlinear parts

$$\Delta_0 = \sqrt{t_{B0}^2 - 4t_{A0} t_{C0}}, \hspace{1cm} \Delta_1 = \frac{t_{B0} t_{B1} - 2 \left( t_{A1} t_{C0} + t_{A0} t_{C1} \right)}{\Delta_0},$$  \hspace{1cm} (63)

$$\kappa_{0\pm} = \omega^2 \frac{-t_B \pm \Delta_0}{2 t_{C0}}, \hspace{1cm} \kappa_{1\pm} = \omega^2 \left( -t_{B1} \pm \Delta_1 \right) - \frac{2 t_{C1} \left( \kappa_{0\pm}/\omega^2 \right)}{2 t_{C0}},$$  \hspace{1cm} (64)

$$d_{0\pm} = L \sqrt{\kappa_{0\pm}}, \hspace{1cm} d_{1\pm} = L \frac{\kappa_{1\pm}}{2 \sqrt{\kappa_{0\pm}}}.$$  \hspace{1cm} (65)

The linear and nonlinear components of $W$ are thereby given as

$$W_0 = \frac{f_a f_b \omega}{4 \pi i} \int_0^{2\pi} d\phi \int_0^\pi d\theta \frac{\sin \theta}{3 \Delta_0} \left( \frac{\tau_\alpha}{\omega^2} \tilde{\alpha}_0 + \tau_\beta \tilde{\beta}_0 \right),$$  \hspace{1cm} (66)

and

$$W_1 = \frac{f_a f_b \omega}{4 \pi i} \int_0^{2\pi} d\phi \int_0^\pi d\theta \sin \theta \times \left\{ \frac{1}{3 \Delta_0} \left[ \frac{\tau_\alpha}{\omega^2} \tilde{\alpha}_1 + \tau_\beta \tilde{\beta}_1 + \frac{3}{\omega^2} \left( d_{0+} d_{1+} (1 + i d_{0+}) - d_{0-} d_{1-} (1 + i d_{0-}) \right) \tilde{\alpha}_0 + i \left[ \frac{d_0^3}{\kappa_{0\pm}} \left( \frac{3 d_{1+}}{d_{0+} - \kappa_{1\pm}} - \frac{3 d_{1-}}{d_{0-} - \kappa_{1\pm}} \right) - \frac{d_{0\pm}^3}{\kappa_{0\pm}} \left( \frac{\kappa_{1+}}{\kappa_{0+}} - \frac{\kappa_{1-}}{\kappa_{0-}} \right) \right] \tilde{\beta}_0 + \frac{\Delta_1}{3 \Delta_0} \left( \frac{\tau_\alpha}{\omega^2} \tilde{\alpha}_0 + \tau_\beta \tilde{\beta}_0 \right) \right\},$$  \hspace{1cm} (67)

respectively, where

$$\tau_\alpha = \frac{3}{2} \left( d_{0+}^2 - d_{0-}^2 \right) + i \left( d_{0+}^3 - d_{0-}^3 \right), \hspace{1cm} \tau_\beta = i \left( \frac{d_{0+}^3}{\kappa_{0\pm}} - \frac{d_{0-}^3}{\kappa_{0\pm}} \right).$$  \hspace{1cm} (68)

The integrals (66) and (67) are straightforwardly evaluated by standard (e.g., Gaussian) numerical methods [34]. In the degenerate isotropic case $U_x = U_y = U_z$, the integrals (66) and (67) yield the analytic results of [17].
Figure 1: Schematic diagram illustrating the bilocally-approximated SPFT for ellipsoidal component phase geometry: pair-wise scattering interactions are accommodated between ellipsoidal scattering centres contained within an ellipsoidal correlation region of linear dimensions $L$. 
Figure 2: HCM relative linear permittivity and nonlinear susceptibility parameters calculated using the Bruggeman homogenization formalism. Component phase parameter values: $\epsilon_{a0} = 2\epsilon_0$, $\chi_a = 9.07571 \times 10^{-12}\epsilon_0\text{m}^2\text{V}^{-2}$, $\epsilon_b \equiv \epsilon_{b0} = 12\epsilon_0$, $U_x = 1$ and $U_y = 3$. 
Figure 3: Real and imaginary parts of the HCM linear permittivity parameters calculated using the SPFT homogenization formalism. Component phase parameter values: \( \epsilon_{a0} = 2\epsilon_0, \chi_a = 9.07571 \times 10^{-12}\epsilon_0 \text{m}^2\text{V}^{-2}, \epsilon_b \equiv \epsilon_{b0} = 12\epsilon_0, U_x = 1, U_y = 3 \) and \( U_z = 15. \)
Figure 4: As figure 3 but for the HCM nonlinear susceptibility parameters.