Third-order implementation and convergence of the strong-property-fluctuation theory in electromagnetic homogenization

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The strong-property-fluctuation theory (SPFT) has been widely applied under the second-order approximation (also known as the bilocal approximation) to estimate the constitutive properties of effectively homogeneous composite mediums. A third-order mass operator approximation is developed here. The convergence of the long-wavelength, bilocally-approximated SPFT is demonstrated for isotropic chiral composite mediums, as well as for chiroferrite composite mediums.

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I. INTRODUCTION

In modeling the behavior of physical systems, the mathematical representation of solutions as infinite series is commonplace. However, first- or second-order approximations are often utilized, while the implementation of higher-order approximations may be too arduous in practice. Clearly, the issue of convergence is of fundamental importance in such cases. A prime example occurs in the strong-property-fluctuation theory (SPFT). The SPFT provides a formalism for estimating the constitutive properties of effectively homogeneous, linear composite mediums. It represents an advance over the more conventional approaches to homogenization, such as the Maxwell Garnett formalism (including its incremental and differential variants) and the Bruggeman formalism [1–6], by providing a more comprehensive description of the distributional statistics of the component material phases. The SPFT has been successively developed for the cases of isotropic dielectric [7], anisotropic dielectric [8], isotropic chiral [9], and, most recently, bianisotropic [10] mediums. Recent studies for the general bianisotropic case have highlighted the role of the component phase topology and correlation length [11], as well as covariance function [12].

In the SPFT, statistical cumulants of the spatial distribution of the component material phases are used perturbatively to refine an initial ansatz for the nature of the homogenized composite medium (HCM). Solutions are expressed in terms of a so-called *mass operator* that—due to the process of iteration—has an infinite series representation. Previous studies [7–12] have exclusively concentrated on the zeroth-, first-, and second-order truncations of the mass operator series. In the usual SPFT implementation, the second-order truncation—known as the *bilocal approximation*—is adopted. A covariance function and its associated correlation length L are used to characterize the distributional statistics of the component material phases under the bilocal approximation. Furthermore, the *long-wavelength* approximation is usually adopted in which the actions of scattering centres separated by distances much greater than the correlation length are assumed to be statistically independent. To our knowledge, higher-order approximations and the issue of convergence of the mass operator representation have not hitherto been addressed. In the present paper, we develop a third-order approximation, and investigate convergence of the mass operator series. The analysis is presented for isotropic chiral composite mediums. Additionally, we conjecture that our conclusions hold for more general mediums; this conjecture is substantiated by our numerical results.

In the notation adopted, six-vectors (three-vectors) are in bold (normal) face and underlined, whereas 6×6 (3×3) dyadics are in bold (normal) face and double underlined. The inverse and the ℓjth entry of a dyadic $\underline{\underline{\mathbf{Y}}}$ are given as $\underline{\underline{\mathbf{Y}}}^{-1}$ and $[\underline{\underline{\mathbf{Y}}}]_{\ell j}$, respectively. For dyadics and vectors, the dot product denotes contraction of indexes. The unit vector corresponding to a vector $\underline{n}$ is signified by $\hat{\underline{n}}$ and the 6×6 (3×3) identity dyadic by $\underline{\underline{\mathbf{I}}}$ $(\underline{\underline{I}})$. The permittivity and permeability of free space (i.e., vacuum) are denoted by ϵ_0 and μ_0, respectively. The Cartesian unit vector in the z direction is $\hat{\underline{u}}_z$. The ensemble average of a quantity ψ is represented by $\langle \psi \rangle$.

II. SPFT GENERALITIES

We consider the long-wavelength SPFT pertaining to a two-phase HCM. The component phases are designated a and b. All space is partitioned into the disjoint regions V_a and V_b that contain the phases a and b, respectively. The

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component phases are assumed to be randomly distributed as spheres; the distributions are specified in terms of characteristic functions given as

$$\theta_\ell(\underline{r}) = \begin{cases} 1, & \underline{r} \in V_\ell \\ & \qquad (\ell = a,b). \\ 0, & \underline{r} \notin V_\ell \end{cases} \tag{1}$$

In particular, statistical moments of the characteristic functions are utilized: The nth moment is the expectation value $\langle \theta_\ell(\underline{r}_1) \cdots \theta_\ell(\underline{r}_n) \rangle$ and represents the probability for $\underline{r}_1, \ldots, \underline{r}_n$ being inside V_ℓ $(\ell = a,b)$; $\langle \ \rangle$ denotes ensemble averaging. Thus, the volume fraction of phase ℓ is given by the first moment of $\theta_\ell(\underline{r})$; i.e.,

$$f_\ell = \langle \theta_\ell(\underline{r}) \rangle, \quad (\ell = a,b). \tag{2}$$

Further, the second moment of $\theta_\ell(\underline{r})$ is used to define a covariance function as

$$\tau(\underline{R}) = \langle \theta_a(\underline{r}) \theta_a(\underline{r}') \rangle - \langle \theta_a(\underline{r}) \rangle \langle \theta_a(\underline{r}') \rangle, \tag{3}$$

or, equivalently,

$$\tau(\underline{R}) = \langle \theta_b(\underline{r}) \theta_b(\underline{r}') \rangle - \langle \theta_b(\underline{r}) \rangle \langle \theta_b(\underline{r}') \rangle, \tag{4}$$

where $\underline{R} = \underline{r} - \underline{r}'$. We define a correlation length L such that the covariance function $\tau(\underline{R})$ vanishes for $|\underline{R}| \gg L$. In the long-wavelength regime, the principal electromagnetic wavelengths are assumed to be much larger than the correlation length L.

The component phases are characterized by linear bianisotropic constitutive relations, given in the frequency domain as

$$\underline{\mathbf{C}}(\underline{r}) = \underline{\underline{\mathbf{K}}}_\ell \cdot \underline{\mathbf{F}}(\underline{r}), \quad \underline{r} \in V_\ell, \quad (\ell = a,b), \tag{5}$$

where

$$\underline{\mathbf{C}}(\underline{r}) = \begin{bmatrix} \underline{D}(\underline{r}) \\ \underline{B}(\underline{r}) \end{bmatrix}, \quad \underline{\underline{\mathbf{K}}}_\ell = \begin{bmatrix} \underline{\underline{\epsilon}}_\ell & \underline{\underline{\xi}}_\ell \\ \underline{\underline{\zeta}}_\ell & \underline{\underline{\mu}}_\ell \end{bmatrix}, \quad \underline{\mathbf{F}}(\underline{r}) = \begin{bmatrix} \underline{E}(\underline{r}) \\ \underline{H}(\underline{r}) \end{bmatrix}. \tag{6}$$

The 3×3 dyadics $\underline{\underline{\epsilon}}_\ell$ and $\underline{\underline{\mu}}_\ell$ are the permittivity and the permeability dyadics, respectively, whereas $\underline{\underline{\xi}}_\ell$ and $\underline{\underline{\zeta}}_\ell$ are the magnetoelectric constitutive dyadics.

A key concept in the SPFT is the *bianisotropic comparison medium* (BCM), characterized by the constitutive dyadic $\underline{\underline{\mathbf{K}}}_{BCM}$. We denote the dyadic Green function of the BCM by $\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R})$ and note that the singular behavior of $\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R})$ in the limit $|\underline{R}| \to 0$ is conveniently isolated through

$$\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R}) = \mathcal{P} \underline{\underline{\mathbf{G}}}_{BCM}(\underline{R}) + \underline{\underline{\mathbf{D}}} \delta(\underline{R}), \tag{7}$$

where $\mathcal{P}$ is the principal value operation excluding a spherical region about $\underline{R} = \underline{0}$, $\underline{\underline{\mathbf{D}}}$ is the corresponding depolarization dyadic [13], and $\delta(\underline{R})$ is the Dirac delta function. Furthermore, we observe that [13]

$$\int \underline{\underline{\mathbf{G}}}_{BCM}(\underline{R}) d^3\underline{R} = \frac{1}{i\omega} \underline{\underline{\mathbf{K}}}_{BCM}^{-1}. \tag{8}$$

The basis of the SPFT lies in the introduction of the *exciting field*

$$\underline{\mathbf{F}}_{exc}(\underline{r}) = \{ \underline{\underline{\mathbf{I}}} + i\omega \underline{\underline{\mathbf{D}}} \cdot [\underline{\underline{\mathbf{K}}}(\underline{r}) - \underline{\underline{\mathbf{K}}}_{BCM}] \} \cdot \underline{\mathbf{F}}(\underline{r}), \tag{9}$$

where

$$\underline{\underline{\mathbf{K}}}(\underline{r}) = \underline{\underline{\mathbf{K}}}_a \theta_a(\underline{r}) + \underline{\underline{\mathbf{K}}}_b \theta_b(\underline{r}); \tag{10}$$

and the generalized polarizability dyadic

$$\underline{\underline{\boldsymbol{\chi}}}(\underline{r}) = \underline{\underline{\boldsymbol{\chi}}}_a \theta_a(\underline{r}) + \underline{\underline{\boldsymbol{\chi}}}_b \theta_b(\underline{r}), \tag{11}$$

with

$$\underline{\underline{\boldsymbol{\chi}}}_\ell = -i\omega [\underline{\underline{\mathbf{K}}}_\ell - \underline{\underline{\mathbf{K}}}_{BCM}] \cdot [\underline{\underline{\mathbf{I}}} + i\omega \underline{\underline{\mathbf{D}}} \cdot (\underline{\underline{\mathbf{K}}}_\ell - \underline{\underline{\mathbf{K}}}_{BCM})]^{-1}, \quad (\ell = a,b). \tag{12}$$

The quantities $\underline{\mathbf{F}}_{exc}(\underline{r})$ and $\underline{\underline{\boldsymbol{\chi}}}(\underline{r})$ are linked through the integral equation [10]

$$\underline{\mathbf{F}}_{exc}(\underline{r}) = \underline{\mathbf{F}}_{BCM}(\underline{r}) + \mathcal{P} \int \underline{\underline{\mathbf{G}}}_{BCM}(\underline{r} - \underline{r}') \cdot \underline{\underline{\boldsymbol{\chi}}}(\underline{r}') \cdot \underline{\mathbf{F}}_{exc}(\underline{r}') d^3\underline{r}', \tag{13}$$

wherein the local spatially averaged electromagnetic field is represented by $\underline{\mathbf{F}}_{BCM}(\underline{r})$. The integral equation (13) may be formally represented in terms of its Born series as

$$\begin{aligned} \underline{\mathbf{F}}_{exc}(\underline{r}) = {} & \underline{\mathbf{F}}_{BCM}(\underline{r}) + \mathcal{P} \int \underline{\underline{\mathbf{G}}}_{BCM}(\underline{r} - \underline{r}') \cdot \underline{\underline{\boldsymbol{\chi}}}(\underline{r}') \cdot \underline{\mathbf{F}}_{BCM}(\underline{r}') d^3\underline{r}' + \mathcal{P} \int \underline{\underline{\mathbf{G}}}_{BCM}(\underline{r} - \underline{r}') \cdot \underline{\underline{\boldsymbol{\chi}}}(\underline{r}') \\ & \cdot \left[\mathcal{P} \int \underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}' - \underline{r}'') \cdot \underline{\underline{\boldsymbol{\chi}}}(\underline{r}'') \cdot \underline{\mathbf{F}}_{BCM}(\underline{r}'') d^3\underline{r}'' \right] d^3\underline{r}' + \mathcal{P} \int \underline{\underline{\mathbf{G}}}_{BCM}(\underline{r} - \underline{r}') \cdot \underline{\underline{\boldsymbol{\chi}}}(\underline{r}') \\ & \cdot \left[\mathcal{P} \int \underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}' - \underline{r}'') \cdot \underline{\underline{\boldsymbol{\chi}}}(\underline{r}'') \cdot \left\{ \mathcal{P} \int \underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}'' - \underline{r}''') \cdot \underline{\underline{\boldsymbol{\chi}}}(\underline{r}''') \cdot \underline{\mathbf{F}}_{BCM}(\underline{r}''') d^3\underline{r}''' \right] d^3\underline{r}'' \right\} d^3\underline{r}' + \cdots. \end{aligned} \tag{14}$$

By ensemble averaging the terms of Eq. (14) separately and reordering terms using a Feynman-diagrammatic technique [14,15], the *Dyson equation* [10]

$$\langle \underline{\mathbf{F}}_{exc}(\underline{r})\rangle = \underline{\mathbf{F}}_{BCM}(\underline{r}) + \mathcal{P}\int \underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}-\underline{r}') \cdot \left[\int \underline{\underline{\boldsymbol{\Sigma}}}(\underline{r}'-\underline{r}'')\cdot\langle \underline{\mathbf{F}}_{exc}(\underline{r}'')\rangle d^3\underline{r}''\right] d^3\underline{r}' \tag{15}$$

is developed. The *mass operator* $\underline{\underline{\boldsymbol{\Sigma}}}(\underline{r}'-\underline{r}'')$ has an infinite series representation, the terms of which comprise products over $\mathrm{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}'-\underline{r}'')$ and the statistical cumulants of $\underline{\underline{\boldsymbol{\chi}}}(\underline{r}'-\underline{r}'')$. For later convenience, we express

$$\underline{\underline{\boldsymbol{\Sigma}}}(\underline{R}) = \underline{\underline{\boldsymbol{\Sigma}}}_0(\underline{R}) + \underline{\underline{\boldsymbol{\Sigma}}}_1(\underline{R}) + \underline{\underline{\boldsymbol{\Sigma}}}_2(\underline{R}) + \underline{\underline{\boldsymbol{\Sigma}}}_3(\underline{R}) + \cdots, \tag{16}$$

where the subscript j in $\underline{\underline{\boldsymbol{\Sigma}}}_j(\underline{R})$ refers to the order of $\underline{\underline{\boldsymbol{\chi}}}(\underline{R})$. The Dyson equation may be manipulated to deliver an estimate of $\underline{\underline{\mathbf{K}}}_{Dy0}$—the constitutive dyadic of the HCM arising in the long-wavelength SPFT. Thus, we have [10]

$$\underline{\underline{\mathbf{K}}}_{Dy0} = \underline{\underline{\mathbf{K}}}_{BCM} - \frac{1}{i\omega}(\underline{\underline{\mathbf{I}}} + \tilde{\underline{\underline{\boldsymbol{\Sigma}}}}^{\dagger}\cdot\underline{\underline{\mathbf{D}}})^{-1}\cdot\tilde{\underline{\underline{\boldsymbol{\Sigma}}}}^{\dagger}, \tag{17}$$

where $\tilde{\underline{\underline{\boldsymbol{\Sigma}}}}^{\dagger}$ is the Fourier transform of the mass operator evaluated at zero spatial frequency; i.e.,

$$\tilde{\underline{\underline{\boldsymbol{\Sigma}}}}^{\dagger} = \int \underline{\underline{\boldsymbol{\Sigma}}}(\underline{R})\, d^3\underline{R}. \tag{18}$$

III. MASS OPERATOR APPROXIMATIONS

For practical purposes, an approximate evaluation of the mass operator is necessary. The lowest-order truncation of the mass operator series, i.e.,

$$\underline{\underline{\boldsymbol{\Sigma}}}(\underline{R}) \approx \underline{\underline{\boldsymbol{\Sigma}}}_0(\underline{R}) = \underline{\underline{\mathbf{0}}}, \tag{19}$$

gives the trivial result $\underline{\underline{\mathbf{K}}}_{Dy0} = \underline{\underline{\mathbf{K}}}_{BCM}$. The BCM is conventionally chosen so that the first-order mass operator approximation does not add to the zero-order approximation [7,8,10]. Thus, we have the condition

$$\langle \underline{\underline{\boldsymbol{\chi}}}(\underline{r})\rangle = \underline{\underline{\mathbf{0}}} \tag{20}$$

and $\underline{\underline{\boldsymbol{\Sigma}}}_1(\underline{R}) = \underline{\underline{\mathbf{0}}}$ correspondingly. We reiterate here that through the condition of Eq. (20), $\underline{\underline{\mathbf{K}}}_{BCM}$ becomes identical to the HCM constitutive dyadic arising from the Bruggeman homogenization formalism.

The most widely adopted procedure is to implement the second-order truncation of the mass operator series, whence

$$\underline{\underline{\boldsymbol{\Sigma}}}(\underline{r}-\underline{r}') \approx \underline{\underline{\boldsymbol{\Sigma}}}_2(\underline{r}-\underline{r}') = \langle \underline{\underline{\boldsymbol{\chi}}}(\underline{r})\cdot \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}-\underline{r}')\cdot \underline{\underline{\boldsymbol{\chi}}}(\underline{r}')\rangle, \tag{21}$$

which is known as the *bilocal approximation*. After using Eqs. (11) and (20), the bilocally approximated mass operator may be conveniently expressed as

$$\underline{\underline{\boldsymbol{\Sigma}}}_2(\underline{R}) = \tau(\underline{R})(\underline{\underline{\boldsymbol{\chi}}}_a - \underline{\underline{\boldsymbol{\chi}}}_b)\cdot \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R})\cdot(\underline{\underline{\boldsymbol{\chi}}}_a - \underline{\underline{\boldsymbol{\chi}}}_b). \tag{22}$$

In order to evaluate $\int \underline{\underline{\boldsymbol{\Sigma}}}_2(\underline{R})d^3\underline{R}$, the second moment $\langle \theta_a(\underline{r})\theta_a(\underline{r}')\rangle$ must be specified. For the physically motivated form [16]

$$\langle \theta_a(\underline{r})\theta_a(\underline{r}')\rangle = \begin{cases} f_a, & |\underline{r}-\underline{r}'| \leq L \\ f_a^2, & |\underline{r}-\underline{r}'| > L, \end{cases} \tag{23}$$

we have

$$\int \underline{\underline{\boldsymbol{\Sigma}}}_2(\underline{R})\, d^3\underline{R} = (\underline{\underline{\boldsymbol{\chi}}}_a - \underline{\underline{\boldsymbol{\chi}}}_b)\cdot \underline{\underline{\mathbf{W}}}\cdot(\underline{\underline{\boldsymbol{\chi}}}_a - \underline{\underline{\boldsymbol{\chi}}}_b), \tag{24}$$

the evaluation of

$$\underline{\underline{\mathbf{W}}} = \int_{|\underline{R}|\leq L} \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R})\, d^3\underline{R}, \tag{25}$$

having been provided elsewhere [10,11]. Calculations of $\int \underline{\underline{\boldsymbol{\Sigma}}}_2(\underline{R})d^3\underline{R}$ for other choices of $\langle \theta_a(\underline{r})\theta_a(\underline{r}')\rangle$ have also been presented [12]; and we note here that the resulting estimates of $\underline{\underline{\mathbf{K}}}_{Dy0}$ were found to be comparatively insensitive to the form of $\langle \theta_a(\underline{r})\theta_a(\underline{r}')\rangle$ [12].

On retaining the next highest-order term (i.e., to third order in $\underline{\underline{\boldsymbol{\chi}}}$), the mass operator is approximated by [14,15]

$$\underline{\underline{\boldsymbol{\Sigma}}}(\underline{r}-\underline{r}') \approx \underline{\underline{\boldsymbol{\Sigma}}}_2(\underline{r}-\underline{r}') + \underline{\underline{\boldsymbol{\Sigma}}}_3(\underline{r}-\underline{r}'), \tag{26}$$

where

$$\underline{\underline{\boldsymbol{\Sigma}}}_3(\underline{r}-\underline{r}') = \int \langle \underline{\underline{\boldsymbol{\chi}}}(\underline{r})\cdot \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}-\underline{r}'')\cdot \underline{\underline{\boldsymbol{\chi}}}(\underline{r}'') \cdot \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}''-\underline{r}')\cdot \underline{\underline{\boldsymbol{\chi}}}(\underline{r}')\rangle d^3\underline{r}''. \tag{27}$$

Some algebraic manipulations utilizing Eqs. (11) and (20) lead to

$$\underline{\underline{\boldsymbol{\Sigma}}}_3(\underline{r}-\underline{r}') = \left(\frac{1}{1-f_a}\right)^3 \underline{\underline{\boldsymbol{\chi}}}_a\cdot[\underline{\underline{\mathbf{T}}} - f_a(\underline{\underline{\mathbf{M}}}_1 + \underline{\underline{\mathbf{M}}}_2 + \underline{\underline{\mathbf{N}}})]\cdot \underline{\underline{\boldsymbol{\chi}}}_a, \tag{28}$$

in which

$$\underline{\underline{\mathbf{T}}} = \int \langle \theta_a(\underline{r})\theta_a(\underline{r}')\theta_a(\underline{r}'')\rangle \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}-\underline{r}'')\cdot \underline{\underline{\boldsymbol{\chi}}}_a \cdot \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}''-\underline{r}')\, d^3\underline{r}'', \tag{29}$$

$$\underline{\underline{\mathbf{M}}}_1 = \int \langle \theta_a(\underline{r})\theta_a(\underline{r}'')\rangle \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}-\underline{r}'')\cdot \underline{\underline{\boldsymbol{\chi}}}_a \cdot \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}''-\underline{r}')\, d^3\underline{r}'', \tag{30}$$

$$\underline{\underline{\mathbf{M}}}_2 = \int \langle \theta_a(\underline{r}'')\theta_a(\underline{r}')\rangle \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}-\underline{r}'')\cdot \underline{\underline{\boldsymbol{\chi}}}_a \cdot \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}''-\underline{r}')\, d^3\underline{r}'', \tag{31}$$

$$\underline{\underline{\mathbf{N}}} = (\langle \theta_a(\underline{r})\theta_a(\underline{r}')\rangle - 2f_a^2)\int \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}-\underline{r}'')\cdot\underline{\underline{\boldsymbol{\chi}}}_a \cdot \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}''-\underline{r}')d^3\underline{r}'', \tag{32}$$

are all implicit functions of $\underline{R}$.

We now consider the evaluation of $\int \underline{\underline{\mathbf{T}}} d^3\underline{R}$, $\int \underline{\underline{\mathbf{M}}}_{1,2} d^3\underline{R}$ and $\int \underline{\underline{\mathbf{N}}} d^3\underline{R}$ for use in Eq. (18), and then in Eq. (17). To do so, we must first specify the second and third moments of $\theta_a(\underline{r})$. Consistent with [10], we choose Eq. (23) and

$$\langle \theta_a(\underline{r})\theta_a(\underline{r}')\theta_a(\underline{r}'')\rangle = \begin{cases} f_a^3, & \min\{L_{12},L_{13},L_{23}\} > L \\ f_a, & \max\{L_{12},L_{13},L_{23}\} \leqslant L \\ \frac{1}{3}(f_a+2f_a^3), & \text{one of } L_{12},L_{13},L_{23} \leqslant L \\ \frac{1}{3}(2f_a+f_a^3), & \text{two of } L_{12},L_{13},L_{23} \leqslant L, \end{cases} \tag{33}$$

where

$$L_{12} = |\underline{r}-\underline{r}'|, \quad L_{13} = |\underline{r}-\underline{r}''|, \quad L_{23} = |\underline{r}'-\underline{r}''|. \tag{34}$$

From Eq. (33) we have

$$\int \langle \theta_a(\underline{r})\theta_a(\underline{r}')\theta_a(\underline{r}'')\rangle d^3\underline{r}'' = \int_{L_{13}\leqslant L} h\, d^3\underline{r}'' + \int_{L_{23}\leqslant L} h\, d^3\underline{r}'' + \int [f_a^3 + h\, w(\underline{r}-\underline{r}')]d^3\underline{r}'', \tag{35}$$

where

$$w(\underline{r}-\underline{r}') = \begin{cases} 1, & |\underline{r}-\underline{r}'| \leqslant L \\ 0, & |\underline{r}-\underline{r}'| > L, \end{cases} \tag{36}$$

and $h=(f_a-f_a^3)/3$. Thus, selecting the origin of our coordinate system at $\underline{r}'$, we have

$$\underline{\underline{\mathbf{T}}} = \int \{f_a^3 + h[w(\underline{r}'') + w(\underline{R}-\underline{r}'') + w(\underline{R})]\} \times \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R}-\underline{r}'')\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}'')d^3\underline{r}''. \tag{37}$$

Focusing on the term $w(\underline{R})\mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R}-\underline{r}'')\cdot\underline{\underline{\boldsymbol{\chi}}}_a \cdot\mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}'')$ in Eq. (37), we assume that

$$w(\underline{R})\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}'')\cdot\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}''-\underline{R}) \approx \underline{\underline{\mathbf{0}}} \tag{38}$$

for $|\underline{r}''| \geqslant L$. This simplification is justified in the Appendix for isotropic chiral mediums. It is highly probable that Eq. (38) is also valid for weakly anisotropic mediums with diagonally dominant constitutive dyadics. However, in the absence of appropriate Green function representations for such general mediums, this remains a conjecture. Using Eq. (38), we make the approximation

$$\underline{\underline{\mathbf{T}}} \approx \int \{f_a^3 + h[w(\underline{r}'') + w(\underline{R}-\underline{r}'') + w(\underline{r}'')w(\underline{R}-\underline{r}'')]\} \times \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R}-\underline{r}'')\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}'')d^3\underline{r}''. \tag{39}$$

Taking the spatial Fourier transform of Eq. (39) and applying the convolution theorem [17], we find

$$\int \underline{\underline{\mathbf{T}}} d^3\underline{R} = h(\underline{\underline{\mathbf{V}}}\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\underline{\underline{\mathbf{W}}} + \underline{\underline{\mathbf{W}}}\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\underline{\underline{\mathbf{V}}} + \underline{\underline{\mathbf{W}}}\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\underline{\underline{\mathbf{W}}}) + f_a^3\underline{\underline{\mathbf{V}}}\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\underline{\underline{\mathbf{V}}}, \tag{40}$$

where

$$\underline{\underline{\mathbf{V}}} = \int \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R})d^3\underline{R} \tag{41}$$

$$= \frac{1}{i\omega}\underline{\underline{\mathbf{K}}}_{BCM}^{-1} - \underline{\underline{\mathbf{D}}}, \tag{42}$$

by Eqs. (7) and (8).

We consider now the integration of $\underline{\underline{\mathbf{M}}}_1$: Introducing

$$s(\underline{R}-\underline{r}'') = \begin{cases} f_a, & |\underline{R}-\underline{r}''| \leqslant L \\ f_a^2, & |\underline{R}-\underline{r}''| > L, \end{cases} \tag{43}$$

we have

$$\underline{\underline{\mathbf{M}}}_1 = \int s(\underline{R}-\underline{r}'')\mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R}-\underline{r}'')\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}'')d^3\underline{r}'', \tag{44}$$

where, as previously for $\underline{\underline{\mathbf{T}}}$, we have selected the origin of our coordinate system at $\underline{r}'$. By means of Fourier transformations and application of the convolution theorem again, the integral

$$\int \underline{\underline{\mathbf{M}}}_1 d^3\underline{R} = (f_a - f_a^2)\underline{\underline{\mathbf{W}}}\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\underline{\underline{\mathbf{V}}} + f_a^2\underline{\underline{\mathbf{V}}}\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\underline{\underline{\mathbf{V}}} \tag{45}$$

emerges. It follows similarly that

$$\int \underline{\underline{\mathbf{M}}}_2 d^3\underline{R} = (f_a - f_a^2)\underline{\underline{\mathbf{V}}}\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\underline{\underline{\mathbf{W}}} + f_a^2\underline{\underline{\mathbf{V}}}\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\underline{\underline{\mathbf{V}}}. \tag{46}$$

For $\underline{\underline{\mathbf{N}}}$, we have

$$\underline{\underline{\mathbf{N}}} = (f_a - f_a^2)\int w(\underline{R})\mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R}-\underline{r}''')\cdot\underline{\underline{\boldsymbol{\chi}}}_a \cdot\mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}''')d^3\underline{r}''' - f_a^2\int \mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R}-\underline{r}''')\cdot\underline{\underline{\boldsymbol{\chi}}}_a \cdot\mathcal{P}\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r}''')d^3\underline{r}'''. \tag{47}$$

Utilizing the approximation of Eq. (39) and repeating the procedure of Fourier transformations, we find

$$\int \underline{\underline{\mathbf{N}}} d^3\underline{R} = (f_a - f_a^2)\underline{\underline{\mathbf{W}}}\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\underline{\underline{\mathbf{W}}} - f_a^2\underline{\underline{\mathbf{V}}}\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\underline{\underline{\mathbf{V}}}. \quad (48)$$

Finally, combining Eqs. (40), (45), (46), and (48) into Eq. (28) and integrating, we find

$$\int \underline{\underline{\boldsymbol{\Sigma}}}_3(\underline{R}) d^3\underline{R} = \frac{f_a(1-2f_a)}{3(1-f_a)^2}\underline{\underline{\boldsymbol{\chi}}}_a\cdot(\underline{\underline{\mathbf{V}}}\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\underline{\underline{\mathbf{W}}} + \underline{\underline{\mathbf{W}}}\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\underline{\underline{\mathbf{V}}} + \underline{\underline{\mathbf{W}}}\cdot\underline{\underline{\boldsymbol{\chi}}}_a\cdot\underline{\underline{\mathbf{W}}})\cdot\underline{\underline{\boldsymbol{\chi}}}_a. \quad (49)$$

IV. NUMERICAL RESULTS

Using the zeroth-, second-, and third-order-approximated SPFT in the long-wavelength regime, we investigated the constitutive properties of two examples of HCM. The volume fraction $f_a = 0.3$ and an angular frequency $\omega = 2\pi \times 10^{10}$ rad s^{-1} were selected for all numerical results presented here.

A. Isotropic chiral HCM

In the first example, component phase *a* was chosen to be an isotropic chiral material with constitutive relations

$$\underline{\underline{\epsilon}}^a = \epsilon_0\epsilon^a\underline{\underline{I}}, \quad \underline{\underline{\xi}}^a = -\underline{\underline{\zeta}}^a = i\sqrt{\epsilon_0\mu_0}\,\xi^a\underline{\underline{I}}, \quad \underline{\underline{\mu}}^a = \mu_0\mu^a\underline{\underline{I}}, \quad (50)$$

and parameter values

$$\epsilon^a = \delta(3 + i\,1.5), \quad \xi^a = \delta(1.5 + i), \quad \mu^a = \delta(2 + i\,0.8), \quad (51)$$

where $\delta = 10,\ 20$, and 30. Component phase *b* was taken to be free space. The parameter δ provides the means to vary the constitutive contrast between the component phases. The relative permittivity ϵ^{HCM} of the resulting isotropic chiral HCM is plotted as a function of correlation length L in Fig. 1. The values for the zeroth-order approximation—which are identical to those values calculated using the Bruggeman homogenization formalism—are independent of the correlation length. Furthermore, the calculated values for all orders of approximation coincide at $L = 0$. The magnetoelectric parameter ξ^{HCM} and relative permeability μ^{HCM} of the HCM behave in a manner similar to the relative permittivity ϵ^{HCM} and are displayed in Figs. 2 and 3, respectively.

Under the long-wavelength regime, we require $Q \ll 1$, where

$$Q = \frac{\max W_k}{2\pi} L. \quad (52)$$

For the present case, in which the BCM is an *isotropic chiral* comparison medium,

$$W_k = \{|\gamma^+|, |\gamma^-|\}, \quad (53)$$

where $\gamma^{\pm}$ denote the left- and right-handed wave numbers in the BCM [22]. We find for the example illustrated in Figs. 1–3, $Q = 0.1$ at $L = 5.1 \times 10^{-4}$ m for $\delta = 30$; at $L = 5.7 \times 10^{-4}$ m for $\delta = 20$; and at $L = 7.3 \times 10^{-4}$ m for $\delta = 10$. These limits establish the applicability ranges of the presented formalism.

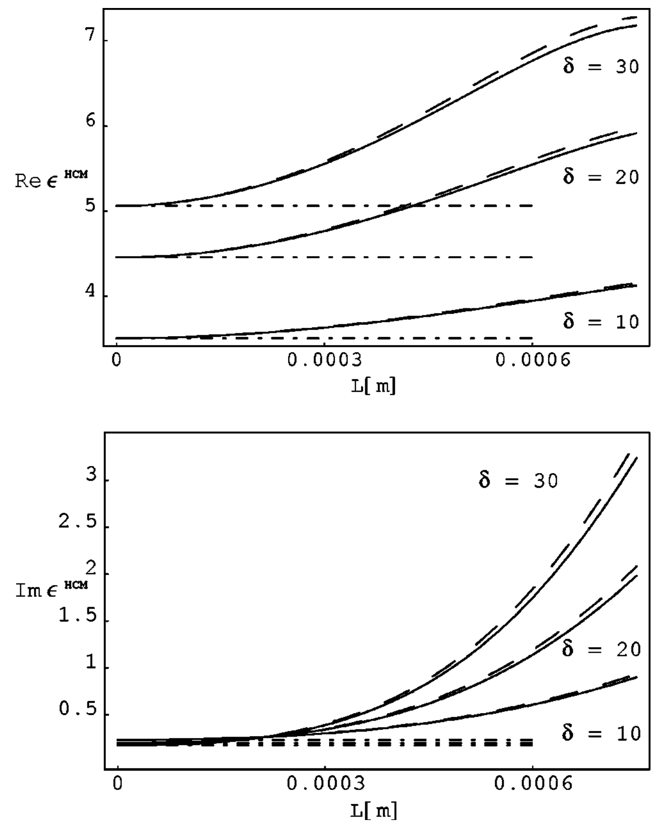

FIG. 1. Real and imaginary parts of the relative permittivity of an isotropic chiral HCM, calculated using the zeroth-, second-, and third-order mass operator approximations, plotted against correlation length L for $\delta = 10$, 20, and 30. At $L = 0$, the calculated values for all orders of approximation coincide. See Sec. IV A for the constitutive parameters of the component phases. Key: broken dashed lines indicate zeroth-order values; solid lines indicate second-order values; dashed lines indicate third-order values.

B. Faraday chiral HCM

For our second example, we again selected the isotropic chiral medium of Eqs. (50) and (51) as component phase *a*. A magnetically gyrotropic medium characterized by

$$\underline{\underline{\epsilon}}^b = \epsilon_0\epsilon^b\underline{\underline{I}}, \quad \underline{\underline{\xi}}^b = \underline{\underline{\zeta}}^b = \underline{\underline{0}}, \quad (54)$$

$$\underline{\underline{\mu}}^b = \mu_0[\mu^b\underline{\underline{I}} - i\mu_g^b\hat{\underline{u}}_z\times\underline{\underline{I}} + (\mu_u^b - \mu^b)\hat{\underline{u}}_z\hat{\underline{u}}_z], \quad (55)$$

was chosen as component phase *b*, along with the parameter values

$$\epsilon^b = 1.2 + i\,0.4, \quad \mu^b = 2.5 + i\,0.5, \quad \mu_u^b = 2.1 + i\,0.4,$$

$$\mu_g^b = 0.2 + i\,0.1. \quad (56)$$

The resulting HCM, with the constitutive relations

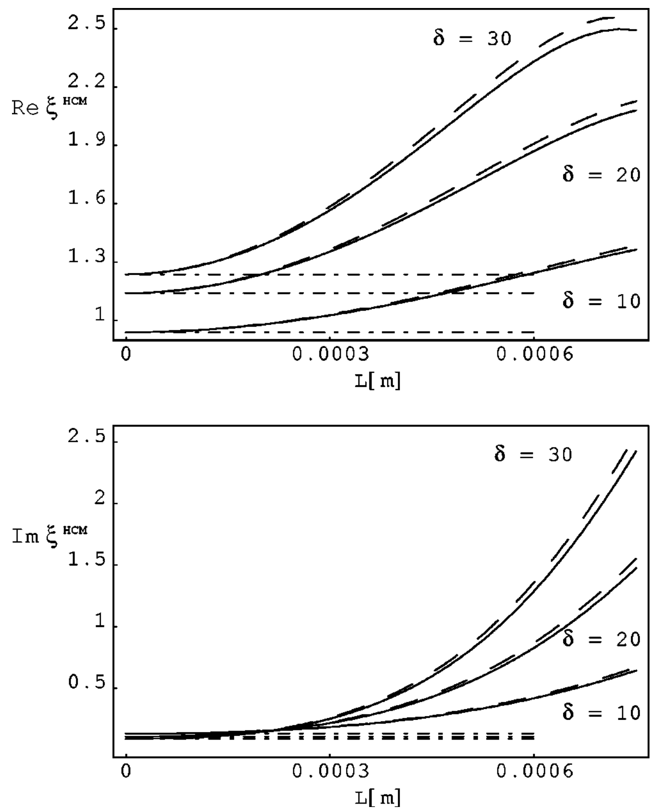

FIG. 2. Same as Fig. 1, but the plotted values are of the magnetoelectric parameter of the isotropic chiral HCM of Sec. IV A.

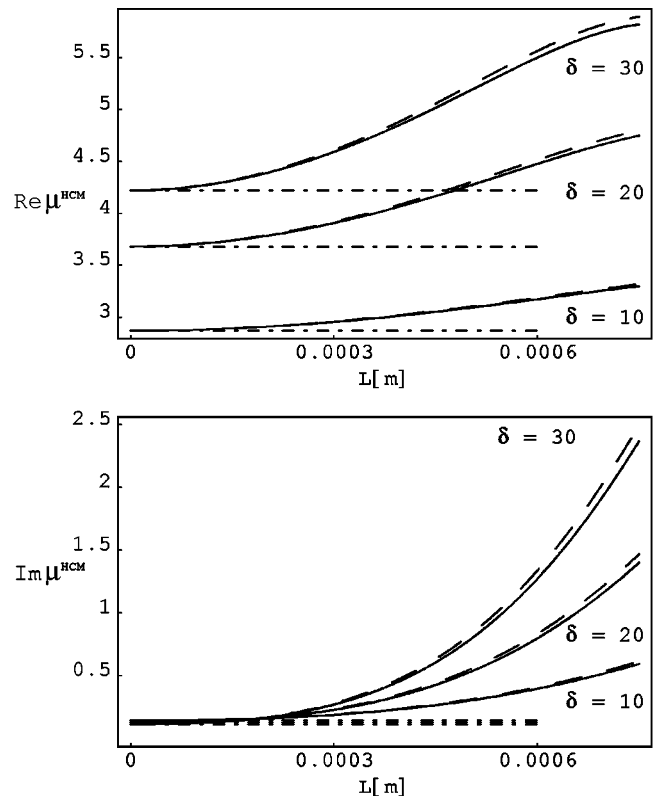

FIG. 3. Same as Fig. 1, but the plotted values are of the relative permeability of the isotropic chiral HCM of Sec. IV A.

$$\underline{\underline{\epsilon}}^{HCM} = \epsilon_0 [\, \epsilon^{HCM} \underline{\underline{I}} - i\, \epsilon_g^{HCM} \hat{\underline{u}}_z \times \underline{\underline{I}} + (\epsilon_u^{HCM} - \epsilon^{HCM}) \hat{\underline{u}}_z \hat{\underline{u}}_z], \tag{57}$$

$$\underline{\underline{\xi}}^{HCM} = i \sqrt{\epsilon_0 \mu_0} [\, \xi^{HCM} \underline{\underline{I}} - i\, \xi_g^{HCM} \hat{\underline{u}}_z \times \underline{\underline{I}} + (\xi_u^{HCM} - \xi^{HCM}) \hat{\underline{u}}_z \hat{\underline{u}}_z], \tag{58}$$

$$\underline{\underline{\zeta}}^{HCM} = - \underline{\underline{\xi}}^{HCM}, \tag{59}$$

$$\underline{\underline{\mu}}^{HCM} = \mu_0 [\, \mu^{HCM} \underline{\underline{I}} - i\, \mu_g^{HCM} \hat{\underline{u}}_z \times \underline{\underline{I}} + (\mu_u^{HCM} - \mu^{HCM}) \hat{\underline{u}}_z \hat{\underline{u}}_z], \tag{60}$$

belongs to the general class of *Faraday chiral mediums* [18,19]. Such HCMs have been comprehensively studied using both the Maxwell Garnett and Bruggeman formalisms [20,21], as well as the bilocally approximated SPFT [11]. The real and imaginary parts of the calculated relative permittivity parameters ϵ^{HCM}, ϵ_u^{HCM}, and ϵ_g^{HCM} are graphed as functions of correlation length L in Figs. 4 and 5, respectively. A similar close agreement between the second- and the third-order calculated values was also found for the magnetoelectric parameters ξ^{HCM}, ξ_u^{HCM}, and ξ_g^{HCM}, and the relative permeability parameters μ^{HCM}, μ_u^{HCM}, and μ_g^{HCM}; therefore, the corresponding graphs are not displayed. For clarity, the zeroth-order approximation values—which are constant with respect to L and are equal to the second- and third-order values in the limit $L \rightarrow 0$—are not displayed. The BCM lies in the category of weakly anisotropic mediums with diagonally dominant constitutive dyadics, for which we anticipate that the simplification (38) is valid.

Since the Faraday chiral HCM is a Lorentz-nonreciprocal medium [23], the corresponding dispersion equation yields four distinct wave numbers: γ_1, γ_2, γ_3, and γ_4. Accordingly, here we have

$$W_k = \{ |\gamma_1|, |\gamma_2|, |\gamma_3|, |\gamma_4| \}, \tag{61}$$

and we find $Q=0.1$ at $L=3.6\times10^{-4}$ m for $\delta=30$; at $L=4.3\times10^{-4}$ m for $\delta=20$; and at $L=5.5\times10^{-4}$ m for $\delta=10$.

V. CONCLUSION

It is clear from Secs. IV A and IV B that the third-order-approximated SPFT yields significantly different results from the bilocally approximated SPFT only as either (i) the correlation length L becomes electrically larger, and/or (ii) the constitutive contrast between the component phases a and b increases.

However, in case (i), the long-wavelength approximation begins to lose validity; while in case (ii), spatial fluctuations in the generalized polarizability $\underline{\underline{\chi}}(\underline{r})$ are likely to become strong. Thus, in either instance the addition of the third-order term $\underline{\underline{\Sigma}}_3(\underline{R})$ to the mass operator is not significant, provided the basic assumptions underlying the long-wavelength SPFT remain valid. We, therefore, conclude that the SPFT converges at the level of the bilocal approximation for isotropic

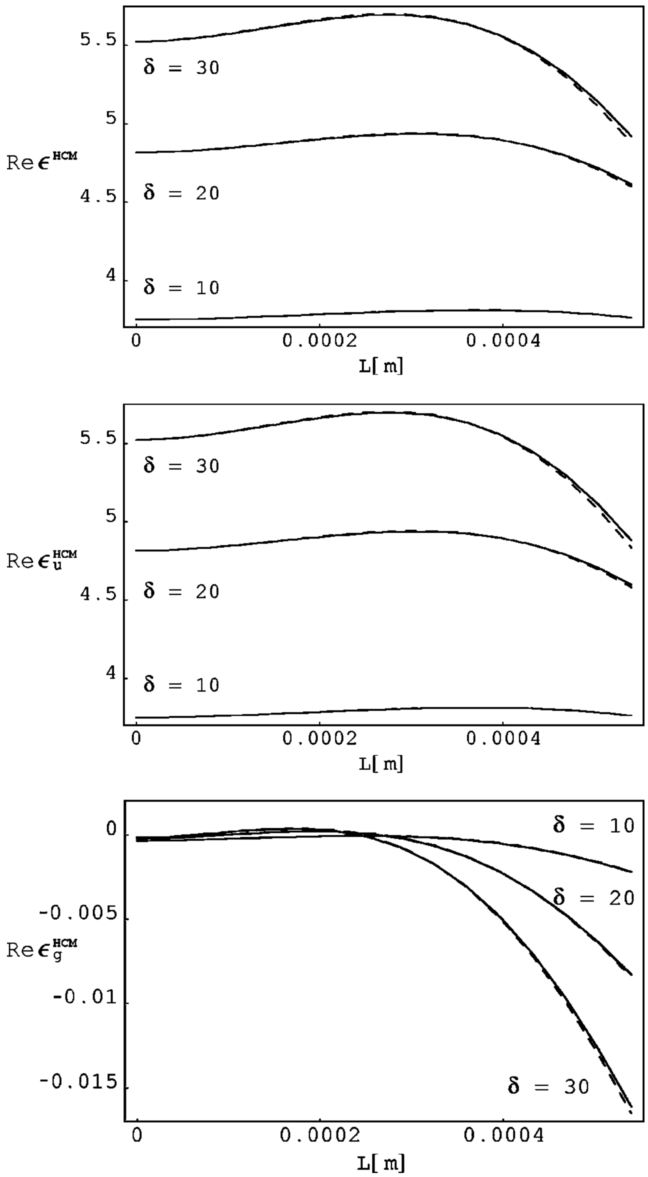

FIG. 4. Real parts of the relative permittivity parameters ϵ^{HCM}, ϵ_u^{HCM}, and ϵ_g^{HCM} of a Faraday chiral HCM, calculated using the second- and third-order mass operator approximations, plotted against correlation length L for $\delta=10$, 20, and 30. See Sec. IV B for the constitutive parameters of the component phases. Key: solid lines indicate second-order values; dashed lines indicate third-order values.

chiral mediums, as well as for chiroferrite mediums that are both weakly uniaxial and weakly gyrotropic.

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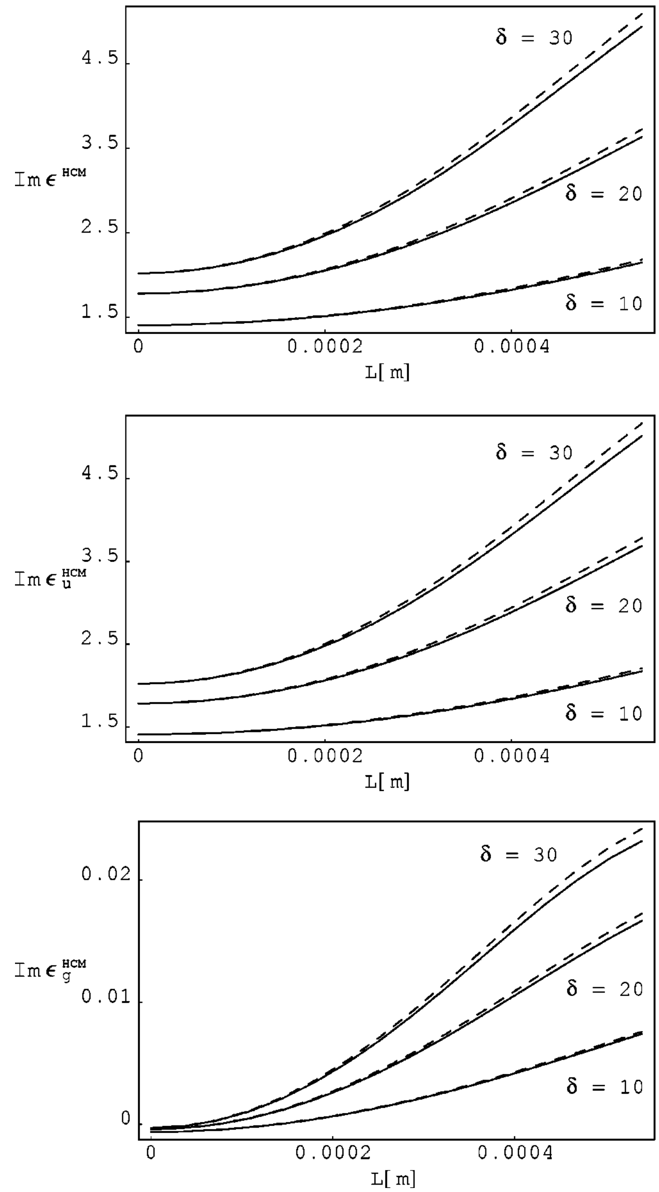

FIG. 5. Same as Fig. 4, but the plotted values are the imaginary parts of the relative permittivity parameters ϵ^{HCM}, ϵ_u^{HCM}, and ϵ_g^{HCM} of the Faraday chiral HCM of Sec. IV B.

APPENDIX

We show that $w(\underline{R})\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r})\cdot\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R}-\underline{r})$ for $|\underline{r}|\gg L$ is negligible in comparison with its evaluation for $|\underline{r}|<L$. To do so, we express the 6×6 dyadic Green function $\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R})$ in terms of 3×3 dyadics as

$$\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R})=\begin{pmatrix} \underline{\underline{G}}^{ee}_{BCM}(\underline{R}) & \underline{\underline{G}}^{em}_{BCM}(\underline{R}) \\ \underline{\underline{G}}^{me}_{BCM}(\underline{R}) & \underline{\underline{G}}^{mm}_{BCM}(\underline{R}) \end{pmatrix}. \tag{A1}$$

We begin by considering the isotropic dielectric-magnetic case: the 3×3 constitutive dyadics are

$$\underline{\underline{\epsilon}}_{BCM}=\epsilon\underline{\underline{I}},\quad \underline{\underline{\mu}}_{BCM}=\mu\underline{\underline{I}},\quad \underline{\underline{\xi}}_{BCM}=\underline{\underline{\zeta}}_{BCM}=\underline{\underline{0}}, \tag{A2}$$

and the wave number $k=\omega\sqrt{\epsilon\mu}$. An explicit representation of $\underline{\underline{G}}^{ee}_{BCM}(\underline{R})$ is available as [24]

$$\underline{\underline{G}}^{ee}_{BCM}(\underline{R})=\frac{1}{i\omega\epsilon}\Bigg[\frac{1}{3}\delta(\underline{R})\underline{\underline{I}}-\frac{1}{4\pi}(-1+ikR+k^2R^2)\times\frac{\exp(ikR)}{R^3}\underline{\underline{I}}+\frac{1}{4\pi}(3-3ikR-k^2R^2)\times\frac{\exp(ikR)}{R^3}\hat{\underline{R}}\hat{\underline{R}}\Bigg]. \quad \text{(A3)}$$

For $r\gg L$, and introducing constants α, β, and γ of order unity, we have

$$\frac{|[\underline{\underline{G}}^{ee}_{BCM}(\underline{r})]_{jj}|}{|[\underline{\underline{G}}^{ee}_{BCM}(L\hat{\underline{r}})]_{jj}|}\approx\frac{\dfrac{\alpha+\beta kr+\gamma k^2r^2}{r^3}}{\dfrac{\alpha+\beta kL+\gamma k^2L^2}{L^3}},\quad (j=1,2,3)$$
$$\approx\left(\frac{L^3}{r^3}\right)\frac{\alpha+\beta kr+\gamma k^2r^2}{\alpha}$$
$$=\frac{L^3}{r^3}+\beta(kL)\frac{L^2}{r^2}+\gamma(kL)^2\frac{L}{r}\approx 0, \quad \text{(A4)}$$

since $|kL|\ll 1$ in the long-wavelength regime.

For the case of the $\underline{\underline{G}}^{ee}_{BCM}(\underline{R}-\underline{r})$ term, we need only consider $R\leq L$ [since $w(\underline{R})=0$ for $R>L$]. Again, for $r\gg L$ and with constants α, β, and γ of order unity, we have

$$\frac{|[\underline{\underline{G}}^{ee}_{BCM}(\underline{R}-\underline{r})]_{jj}|}{|[\underline{\underline{G}}^{ee}_{BCM}(\underline{R}-L\hat{\underline{r}})]_{jj}|}\approx\frac{\dfrac{\alpha+\beta k(R-r)+\gamma k^2(R-r)^2}{(R-r)^3}}{\dfrac{\alpha+\beta k(R-L)+\gamma k^2(R-L)^2}{(R-L)^3}}\quad (j=1,2,3)$$
$$\approx\left(\frac{(R-L)^3}{(R-r)^3}\right)\frac{\alpha+\beta k(R-r)+\gamma k^2(R-r)^2}{\alpha}$$
$$=\frac{(R-L)^3}{(R-r)^3}+\beta[k(R-L)]\frac{(R-L)^2}{(R-r)^2}+\gamma[k(R-L)]^2\frac{R-L}{R-r}\approx 0, \quad \text{(A5)}$$

since $(L-R)\ll(r-R)$. For the isotropic dielectric-magnetic case, the corresponding terms for $\underline{\underline{G}}^{mm}_{BCM}(\underline{R})$ behave similarly to $\underline{\underline{G}}^{ee}_{BCM}(\underline{R})$; the corresponding terms for $\underline{\underline{G}}^{em}_{BCM}(\underline{R})$ and $\underline{\underline{G}}^{me}_{BCM}(\underline{R})$ do not contribute to the analysis of Sec. III as they disappear upon integration.

For an isotropic chiral medium, the diagonal terms of $\underline{\underline{G}}^{ee}_{BCM}(\underline{R})$, $\underline{\underline{G}}^{em}_{BCM}(\underline{R})$, $\underline{\underline{G}}^{me}_{BCM}(\underline{R})$ and $\underline{\underline{G}}^{mm}_{BCM}(\underline{R})$ are all of the same form as those in Eq. (A3) [22], while the integrals of the off-diagonal terms are null valued. Therefore, we have that $w(\underline{R})\underline{\underline{\mathbf{G}}}_{BCM}(\underline{r})\cdot\underline{\underline{\mathbf{G}}}_{BCM}(\underline{R}-\underline{r})$ for $|\underline{r}|\gg L$ is negligible in comparison with its evaluation for $|\underline{r}|<L$, for an isotropic chiral medium.

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