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Homogenization Formalisms for Nonlinear Particulate Composite Mediums

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Abstract

The Maxwell Garnett (MG) and the Bruggeman (Br) formalisms are extended to homogenize *nonlinear*, two–component, composite mediums. The chosen material topology is ellipsoidal and weak nonlinearity is assumed. The MG formalism is illustrated by a case in which both component materials are bianisotropic, but only the inclusion component is nonlinear. Both component materials are isotropic dielectric in the following case, but only one has an intensity–dependent permittivity scalar; and the Br formalism is applied to show that the homogenized composite medium is anisotropic and has cubically nonlinear dielectric properties. Enhancement of nonlinearity emerges as a significant possibility.

1. Introduction

The Maxwell Garnett (MG) and the Bruggeman (Br) formalisms for the homogenization of linear composite mediums formed by randomly dispersing electrically small, bianisotropic, ellipsoidal inclusions in bianisotropic host materials can be extended to composite mediums comprising nonlinear materials. We illustrate this extension with an example for each formalism. The nonlinearity is assumed as weak and, therefore, perturbatively tractable.

In the first example, both component materials are bianisotropic, but only the inclusion material is nonlinear. The effective constitutive properties of the homogenized composite medium (HCM) are estimated using the MG formalism. The linear and nonlinear properties of the HCM are estimated separately in consequence of two assumptions: the nonlinearity of the inclusion material is weak, and the composite medium is dilute [1].

In the second example, both component materials are isotropic dielectric, but just one of them has an intensity–dependent permittivity scalar. Application of the Br formalism shows that the HCM is anisotropic and has cubically nonlinear dielectric properties. The anisotropy of nonlinearity can be considerably different from the anisotropy of the linearity; and the possibility of nonlinearity enhancement exists [2].

2. Nonlinear Bianisotropic Composite Medium

Consider a countably infinite number of identical, electrically small, ellipsoidal inclusions that are similarly oriented but randomly embedded in a host material. Each inclusion has a volume

v; the number density of inclusions is denoted by N; while $f = Nv$, $0 \leq f \leq 1$, is the volumetric proportion of the inclusion medium. Typically, $f < 0.2$ for the MG formalism to yield adequate results. The inclusions are described by a shape dyadic which is real symmetric with positive eigenvalues [3].

The use of 6–vectors and 6×6 dyadics is very convenient for bianisotropic mediums as it permits a compact notation. In this notation, the constitutive properties of the host material are expressed as

$$\underline{\mathbf{G}}(\omega) = \begin{pmatrix} \underline{\underline{\epsilon}}^h(\omega) & \underline{\underline{\xi}}^h(\omega) \\ \underline{\underline{\zeta}}^h(\omega) & \underline{\underline{\mu}}^h(\omega) \end{pmatrix} \bullet \underline{\mathbf{F}}(\omega) = \underline{\underline{\mathbf{C}}}^h(\omega) \bullet \underline{\mathbf{F}}(\omega)\,, \tag{1}$$

where the 6–vectors $\underline{\mathbf{F}}(\omega) = [\underline{E}(\omega),\, \underline{H}(\omega)]^T$ and $\underline{\mathbf{G}}(\omega) = [\underline{D}(\omega),\, \underline{B}(\omega)]^T$, the superscript T denoting the transpose.

With the 6×6 constitutive dyadic of free space denoted by $\underline{\underline{\mathbf{C}}}^v$ (which contains ϵ_0 and μ_0), the linear and the nonlinear constitutive properties of the inclusion material are best expressed through the 6–vector $\underline{\mathbf{Q}}(\omega) = [\underline{P}(\omega)\,, \underline{M}(\omega)]^T = \underline{\mathbf{G}}(\omega) - \underline{\underline{\mathbf{C}}}^v \bullet \underline{\mathbf{F}}(\omega)$, which contains both the polarization field $\underline{P}(\omega)$ and the magnetization field $\underline{M}(\omega)$. This 6–vector is split into linear and nonlinear parts as $\underline{\mathbf{Q}}(\omega) = \underline{\mathbf{Q}}^\ell(\omega) + \underline{\mathbf{Q}}^{nl}(\omega)$. Its linear part $\underline{\mathbf{Q}}^\ell(\omega)$ obeys the constitutive relation $\underline{\mathbf{Q}}^\ell(\omega) = \left[\underline{\underline{\mathbf{C}}}^{in}(\omega) - \underline{\underline{\mathbf{C}}}^v\right] \bullet \underline{\mathbf{F}}(\omega)$, with $\underline{\underline{\mathbf{C}}}^{in}(\omega)$ analogous to $\underline{\underline{\mathbf{C}}}^h(\omega)$.

The nonlinear properties of the inclusion material are described as follows: Let $\mathcal{W} = \{\omega_1, \omega_2, \omega_3, \ldots, \omega_M\}$ be a set of $M > 1$ angular frequencies, there being no requirement that all members of $\mathcal{W}$ be distinct. Suppose there exists an ensemble of M fields $\underline{\mathbf{F}}(\omega_m)$, $(1 \leq m \leq M)$. Then the simultaneous action of this ensemble of fields in the inclusion material gives rise to the nonlinear part of $\underline{\mathbf{Q}}(\omega)$ at $\omega = \omega^{nl}$. The j–th element of this 6–vector is given by

$$Q_j^{nl}(\omega^{nl}) = \sum_{j_1=1}^{6} \sum_{j_2=1}^{6} \cdots \sum_{j_m=1}^{6} \cdots \sum_{j_M=1}^{6} \left\{ \chi^{nl}_{jj_1j_2\ldots j_m\ldots j_M}(\omega^{nl}; \mathcal{W}) \prod_{n=1}^{M} F_{j_n}(\omega_n) \right\}, \quad 1 \leq j \leq 6\,, \tag{2}$$

where $\chi^{nl}_{jj_1j_2\ldots j_m\ldots j_M}(\omega^{nl}; \mathcal{W})$ represents the nonlinear susceptibilities of the inclusion material. The angular frequency ω^{nl} is simply related to all members of $\mathcal{W}$ as $\omega^{nl} = a_1\,\omega_1 + a_2\,\omega_2 + \ldots + a_M\,\omega_M$, with $a_m = \pm 1$, $(1 \leq m \leq M)$; furthermore, ω^{nl} may or or may not lie in $\mathcal{W}$. When $a_n = -1$, $F_{j_n}(\omega_n)$ must be replaced by its complex conjugate on the right side of (2) and in subsequent derivative expressions.

Details of the implementation of the MG formalism to homogenize the described composite medium were given elsewhere [1]. In summary, the constitutive relation of the HCM is given by

$$\underline{\mathbf{G}}(\omega) = \underline{\underline{\mathbf{C}}}^{MG}(\omega) \bullet \underline{\mathbf{F}}(\omega) + s(\omega, \omega^{nl})\, \underline{\mathbf{Q}}^{nls}(\omega)\,, \quad \omega \in \left(\mathcal{W} \cup \left\{\omega^{nl}\right\}\right)\,, \tag{3}$$

where the switching function $s(\omega, \omega^{nl})$ equals unity when both of its arguments are the same, but is null–valued otherwise. Expressions for $\underline{\underline{\mathbf{C}}}^{MG}(\omega)$ have been available for about three years [3], and need not be reproduced here. The nonlinear source polarization–magnetization field is expressed through [1]

$$\underline{\mathbf{Q}}^{nls}(\omega^{nl}) = \left\{ \underline{\underline{\mathbf{I}}} - i\omega^{nl} \left[\underline{\underline{\mathbf{I}}} - i\omega^{nl} f \underline{\underline{\tilde{\mathbf{D}}}}^{\,in/h}(\omega^{nl}) \bullet \underline{\underline{\mathbf{a}}}^{in/h}(\omega^{nl}) \right]^{-1} \bullet \underline{\underline{\tilde{\mathbf{D}}}}^{\,in/h}(\omega^{nl}) \right\} \bullet \underline{\mathbf{Q}}^{nl}_{eff}(\omega^{nl})\,, \tag{4}$$

where

$$\begin{aligned} \left(Q^{nl}_{eff}\right)_j(\omega^{nl}) \;=\; & f \sum_{j_1=1}^{6} \sum_{j_2=1}^{6} \cdots \sum_{j_m=1}^{6} \cdots \sum_{j_M=1}^{6} \Bigg\{ \chi^{nl}_{jj_1j_2\ldots j_m\ldots j_M}(\omega^{nl}; \mathcal{W}) \\ & \prod_{n=1}^{M} \left[\left(\underline{\underline{\mathbf{Y}}}^{in/MG}(\omega_n) \bullet \underline{\mathbf{F}}(\omega_n) \right)_{j_n} \right] \Bigg\}, \quad 1 \leq j \leq 6\,. \end{aligned} \tag{5}$$

In these expressions, the 6×6 dyadics

$$\underline{\underline{\mathbf{a}}}^{in/h}(\omega) = \left[\underline{\underline{\mathbf{C}}}^{in}(\omega) - \underline{\underline{\mathbf{C}}}^{h}(\omega)\right] \bullet \underline{\underline{\mathbf{Y}}}^{in/h}(\omega), \tag{6}$$

$$\underline{\underline{\mathbf{Y}}}^{in/p}(\omega) = \left\{\underline{\underline{\mathbf{I}}} + i\omega\, \underline{\underline{\mathbf{D}}}^{in/p}(\omega) \bullet \left[\underline{\underline{\mathbf{C}}}^{in}(\omega) - \underline{\underline{\mathbf{C}}}^{p}(\omega)\right]\right\}^{-1}, \quad p = h,\, MG, \tag{7}$$

while $\underline{\underline{\mathbf{D}}}^{in/p}$ (resp. $\tilde{\underline{\underline{\mathbf{D}}}}^{in/p}$) is the 6×6 depolarization dyadic of an ellipsoidal (resp. spherical) exclusion region in a linear medium with $\underline{\underline{\mathbf{C}}}^{p}$ as its constitutive dyadic [3].

The presented formalism is general in that it can be used to examine harmonic generation, parametric oscillation, self–focusing, stimulated Raman scattering, and a multitude of nonlinear phenomenons. It also updates and extends our previous work on the MG formalism for complex nonlinear composite mediums [4, 5].

3. Anisotropic Dielectric Composite Medium with Intensity–Dependent Permittivity Dyadic

Next, we implement the Br formulation for homogenizing a mixture of two dielectric materials. For the sake of illustration, here only one component material is assumed to be nonlinear: it possesses an intensity–dependent permittivity scalar. Accordingly, $\underline{\underline{\epsilon}}^{h} = \epsilon^{h}\,\underline{\underline{I}}$, $\underline{\underline{\epsilon}}^{in} = \epsilon^{in}\underline{\underline{I}} = (\epsilon^{in}_{\ell} + \epsilon^{in}_{nl}|\underline{E}|^2)\,\underline{\underline{I}}$, $\underline{\underline{\mu}}^{h} = \underline{\underline{\mu}}^{in} = \mu_0\underline{\underline{I}}$, and the remaining components of $\underline{\underline{\mathbf{C}}}^{h}$ and $\underline{\underline{\mathbf{C}}}^{in}$ are null–valued. The ω–dependences are not explicitly identified, as both the linear and the nonlinear fields vibrate at the same frequency. Both component materials are assumed to have parallel ellipsoidal topologies. As both component materials are treated in the same manner in the Br formalism, the labels *in* and *h* lose the meanings they have for the MG formalism, and the results are valid *prima facie* for $f \in [0,\, 1]$.

The Br formalism requires the solution of the dyadic equation [3]

$$f\,\left(\epsilon^{in}\,\underline{\underline{I}} - \underline{\underline{\epsilon}}^{Br}\right) \bullet \left(\underline{\underline{X}}^{in/Br}\right)^{-1} + (1-f)\,\left(\epsilon^{h}\,\underline{\underline{I}} - \underline{\underline{\epsilon}}^{Br}\right) \bullet \left(\underline{\underline{X}}^{h/Br}\right)^{-1} = \underline{\underline{0}}, \tag{8}$$

where

$$\underline{\underline{X}}^{in/Br} = \underline{\underline{I}} + i\omega\, \underline{\underline{D}}^{in/Br} \bullet \left(\epsilon^{in}\,\underline{\underline{I}} - \underline{\underline{\epsilon}}^{Br}\right) \tag{9}$$

and $\underline{\underline{X}}^{h/Br}$ is defined similarly; while $\underline{\underline{D}}^{in/Br}$ and $\underline{\underline{D}}^{h/Br}$ are 3×3 depolarization dyadics.

The HCM is anisotropic, and the Br formalism predicts its permittivity dyadic as [3, 2]

$$\underline{\underline{\epsilon}}^{Br} = \epsilon^{Br}_{x}\,\underline{u}_x\underline{u}_x + \epsilon^{Br}_{y}\,\underline{u}_y\underline{u}_y + \epsilon^{Br}_{z}\,\underline{u}_z\underline{u}_z\,. \tag{10}$$

A perturbative treatment permits the *ansatz* $\underline{\underline{\epsilon}}^{Br} \simeq \underline{\underline{\epsilon}}^{Br}_{\ell} + \underline{\underline{\epsilon}}^{Br}_{nl}\left|\underline{E}\right|^2$, consistently with our assumption that the nonlinearity of ϵ^{in} is weak; hence,

$$\epsilon^{Br}_{x} \simeq \epsilon^{Br}_{x_\ell} + \epsilon^{Br}_{x_{nl}}\left|\underline{E}\right|^2, \quad \epsilon^{Br}_{y} \simeq \epsilon^{Br}_{y_\ell} + \epsilon^{Br}_{y_{nl}}\left|\underline{E}\right|^2, \quad \epsilon^{Br}_{z} \simeq \epsilon^{Br}_{z_\ell} + \epsilon^{Br}_{z_{nl}}\left|\underline{E}\right|^2. \tag{11}$$

Therefore, the Taylor expansions

$$\underline{\underline{D}}^{p/Br} \simeq \underline{\underline{D}}^{p/Br}_{\ell} + \underline{\underline{D}}^{p/Br}_{nl}\left|\underline{E}\right|^2, \quad p = in,\, h\,, \tag{12}$$

emerge. Expressions for $\underline{\underline{D}}^{p/Br}_{nl}$ are available in Ref. [2]. Accordingly,

$$\underline{\underline{X}}^{p/Br} \simeq \underline{\underline{X}}^{p/Br}_{\ell} + \underline{\underline{X}}^{p/Br}_{nl}\left|\underline{E}^{Br}\right|^2, \quad p = in,\, h\,, \tag{13}$$

where

$$\underline{\underline{X}}_\ell^{in/Br} = \underline{\underline{I}} + i\omega\, \underline{\underline{D}}_\ell^{in/Br} \cdot \left(\epsilon_\ell^{in}\, \underline{\underline{I}} - \underline{\underline{\epsilon}}_\ell^{Br}\right), \tag{14}$$

$$\underline{\underline{X}}_\ell^{h/Br} = \underline{\underline{I}} + i\omega\, \underline{\underline{D}}_\ell^{h/Br} \cdot \left(\epsilon^{h}\, \underline{\underline{I}} - \underline{\underline{\epsilon}}_\ell^{Br}\right), \tag{15}$$

$$\underline{\underline{X}}_{nl}^{in/Br} = i\omega\left[\underline{\underline{D}}_\ell^{in/Br} \cdot \left(g\, \epsilon_{nl}^{in}\, \underline{\underline{I}} - \underline{\underline{\epsilon}}_{nl}^{Br}\right) + \underline{\underline{D}}_{nl}^{in/Br} \cdot \left(\epsilon_\ell^{in}\, \underline{\underline{I}} - \underline{\underline{\epsilon}}_\ell^{Br}\right)\right], \tag{16}$$

$$\underline{\underline{X}}_{nl}^{h/Br} = i\omega\left[-\,\underline{\underline{D}}_\ell^{h/Br} \cdot \underline{\underline{\epsilon}}_{nl}^{Br} + \underline{\underline{D}}_{nl}^{h/Br} \cdot \left(\epsilon^{h}\, \underline{\underline{I}} - \underline{\underline{\epsilon}}_\ell^{Br}\right)\right]. \tag{17}$$

The local field factor g is estimated, as a first approximation, as $g \simeq (1/9)|\mathrm{trace}\left\{\underline{\underline{Y}}_\ell^{in/Br}\right\}|^2$ where $\underline{\underline{Y}}_\ell^{in/Br} = \left(\underline{\underline{X}}_\ell^{in/Br}\right)^{-1}$ [2].

With the foregoing developments, the nonlinear dyadic equation (8) separates into two parts [2]: (i) $\underline{\underline{\epsilon}}_\ell^{Br}$ is the solution of

$$\underline{\underline{\epsilon}}_\ell^{Br} = \left[f\, \underline{\underline{X}}_\ell^{h/Br} + (1-f)\, \underline{\underline{X}}_\ell^{in/Br}\right]^{-1} \cdot \left[f\epsilon_\ell^{in}\, \underline{\underline{X}}_\ell^{h/Br} + (1-f)\epsilon^{h}\, \underline{\underline{X}}_\ell^{in/Br}\right], \tag{18}$$

while (ii) $\underline{\underline{\epsilon}}_{nl}^{Br}$ has to obtained from

$$\begin{aligned}\underline{\underline{\epsilon}}_{nl}^{Br} \;=\; & \left[f\, \underline{\underline{X}}_\ell^{h/Br} + (1-f)\, \underline{\underline{X}}_\ell^{in/Br}\right]^{-1} \cdot \Big[f g \epsilon_{nl}^{in}\, \underline{\underline{X}}_\ell^{h/Br} + \\ & f(\epsilon_\ell^{in}\, \underline{\underline{I}} - \underline{\underline{\epsilon}}_\ell^{Br}) \cdot \underline{\underline{X}}_{nl}^{h/Br} + (1-f)(\epsilon^{h}\, \underline{\underline{I}} - \underline{\underline{\epsilon}}_\ell^{Br}) \cdot \underline{\underline{X}}_{nl}^{in/Br}\Big]. \end{aligned} \tag{19}$$

These two equations were solved iteratively on a computer. The obtained numerical results allowed us to conclude the following [2]:

(i) The anisotropy of nonlinearity can be considerably different from the anisotropy of the linearity in the chosen HCM.

(ii) Enhancement of nonlinearity over that of the inclusion material is possible, the enhancement being anisotropic too.

In closing, we note that the algorithm developed can be easily generalized when *both* component materials have intensity–dependent permittivity scalars.

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