Multiple-scattering formalism for general discrete random composites

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A multiple-scattering formalism is developed for the determination of four effective parameters for bi-isotropic composites in the resonance range. Two models, the truncated quasicrystalline approximation and the dynamic Maxwell-Garnett model, are presented to calculate the effective parameters of general discrete random composites. All the results recover the Maxwell-Garnett mixing formula at low frequency. The degenerative two- and three-effective-parameter cases are also discussed in some detail due to the practical importance of these composites and the requirement of the inherent unity of the formalism.

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I. INTRODUCTION

The concept of chirality (or lack of inversion symmetry) has been the subject of study in many fields, such as chemistry, optics, physics, mathematics, biology, and life science [1]. This fundamental concept deals with the broken symmetry and addresses the handedness of an object or a medium, which is one of the fundamental notions in geometry [1]. Chirality, or handedness, is associated with optical activity, which includes optical rotation dispersion and circular dichroism, has been known for almost two centuries [2]. Recent measurements support that electromagnetic activity also occurs in artificial chiral composites at microwave frequencies [3]. Artificial chiral composites can be constructed by suspending randomly oriented short helices of the same handedness in a host isotropic medium [4]. The possibility of artificial chiral materials that are electromagnetically active in the 1-1000-GHz frequency range has led to predicting the effective medium properties of chiral composites [5-7].

In this paper we consider a discrete random composite medium formed by suspending n_0 identical spheres per unit volume in a host material with permittivity ϵ_0 and permeability μ_0 . The constitutive relations of each scatterer are as follows:

$$\mathbf{D} = \boldsymbol{\epsilon} (\mathbf{E} + \alpha \nabla \times \mathbf{E}) , \qquad (1a)$$

$$\mathbf{B} = \mu(\mathbf{H} + \beta \nabla \times \mathbf{H}) . \tag{1b}$$

Different from Ref. [5], which described the composite by an effective isotopic achiral medium, this paper extends the low-frequency analysis [6,7] to the multiplescattering formalism [5] in the resonance range with the following homogeneous effective medium characterized by [6,7]

$$\mathbf{D} = \epsilon_{\text{eff}} (\mathbf{E} + \alpha_{\text{eff}} \nabla \times \mathbf{E}) , \qquad (2a)$$

$$\mathbf{B} = \mu_{\text{eff}}(\mathbf{H} + \beta_{\text{eff}} \nabla \times \mathbf{H}) . \tag{2b}$$

In experimental work [3], a random suspension of chiral particles in an achiral isotropic medium is known to be of the above constitutive relations. The special case of $\alpha = \beta = \alpha_{\text{eff}} = \beta_{\text{eff}} = 0$ is also considered in some detail since the composites of this kind are also of great importance [8–10].

For wave propagation in a medium that consists of randomly distributed discrete scatterers, the classical assumption is that of independent scattering, which is not valid for a dense medium that contains particles occupying an appreciable fractional volume. This has been verified both theoretically and experimentally [11–13]. The effective-field approximation (EFA) [13], the effective-field approximation with coherent potential (EFA-CP) [13], the quasicrystalline approximation (QCA) [11,13,14], the quasicrystalline approximation with coherent potential (QCA-CP) [13], and the effective-medium approximation (EMA) [15] have been applied to calculate the effective permittivities of discrete dense media.

The purpose of this paper is to extend the multiple-scattering formalism [11,13] to the calculation of the general effective parameters given in Eq. (2). The extension includes two respects. One is the self-consistent multiple-scattering equations for the general discrete random composites. This is given in Sec. II. The other is the practically approximate calculation in the resonance range. Section III deals with this problem by the truncated QCA and dynamic Maxwell-Garnett model. Section IV concludes this paper with a discussion of relevant problems.

II. MULTIPLE-SCATTERING FORMULATION

In this section, the self-consistent multiple-scattering equations for coherent electromagnetic wave propagation through randomly distributed scatterers are derived for the general composites, which was described in the Introduction. We shall consider first the two-parameter formulation. Then we shall give the four-parameter formulation.

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A. Two-parameter formulation

If the scatterer's medium parameters are of the form

$$\epsilon \neq \epsilon_0, \quad \mu \neq \mu_0, \quad \alpha = \beta = 0$$
, (3)

the composite material will be of effective permittivity ϵ_{eff} and effective permeability μ_{eff} . To determine these two quantities, two self-consistent equations are required. Let \mathbf{u}^0 , \mathbf{u}_i^e and \mathbf{u}_i^s denote the incident field, the field exciting the *i*th scatterer, and the field scattered by the *i*th scatterer, respectively; then the self-consistency requires that [5]

$$\mathbf{E}_{i}^{e} = \mathbf{E}^{0} + \sum_{j \ (\neq i)} \mathbf{E}_{j}^{s}(\mathbf{E}_{j}^{e}) ,$$
 (4a)

$$\mathbf{H}_{i}^{e} = \mathbf{H}^{0} + \sum_{j \ (\neq i)} \mathbf{H}_{j}^{s}(\mathbf{H}_{j}^{e}) \ .$$
 (4b)

The incident, exciting, and scattered fields are expanded in terms of vector spherical wave functions as follows [16]:

$$\mathbf{E}^0 = \mathcal{R} \boldsymbol{\psi}^t(\mathbf{r}_i) \cdot \mathbf{a}_e^i , \qquad (5a)$$

$$\mathbf{H}^0 = \mathcal{R} \boldsymbol{\psi}^t(\mathbf{r}_i) \cdot \mathbf{a}_m^i , \qquad (5b)$$

$$\mathbf{E}_{i}^{e} = \mathcal{R} \boldsymbol{\psi}^{t}(\mathbf{r}_{i}) \cdot \boldsymbol{\alpha}_{e}^{i} , \qquad (5c)$$

$$\mathbf{H}_{i}^{e} = \mathcal{R} \boldsymbol{\psi}^{t}(\mathbf{r}_{i}) \cdot \boldsymbol{\alpha}_{m}^{i} , \qquad (5d)$$

$$\mathbf{E}_{i}^{s} = \boldsymbol{\psi}^{t}(\mathbf{r}_{i}) \cdot \mathbf{f}_{a}^{i} , \qquad (5e)$$

$$\mathbf{E}_{i}^{s} = \boldsymbol{\psi}^{t}(\mathbf{r}_{i}) \cdot \mathbf{f}_{m}^{i} , \qquad (5f)$$

where \mathbf{a}_e and \mathbf{a}_m are known coefficients, while α_e^i , α_m^i , \mathbf{f}_e^i , and \mathbf{f}_m^i are unknown coefficients; $\psi(\mathbf{r}_i)$ and $\mathcal{R}\psi(\mathbf{r}_i)$ are the column vectors containing the vector spherical wave functions and their regular parts, respectively; the superscript t denotes the matrix transposition. With these notations, the addition theorems of wave functions read [16]

$$\boldsymbol{\psi}^{t}(\mathbf{r}_{i}) = \mathcal{R}\boldsymbol{\psi}^{t}(\mathbf{r}_{i}) \cdot \underline{\boldsymbol{G}}_{ii}, \quad |\mathbf{r}_{i}| < |\mathbf{r}_{i} - \mathbf{r}_{i}| , \qquad (6a)$$

$$\psi^{t}(\mathbf{r}_{i}) = \psi^{t} \cdot \underline{S}_{ii}, \quad |\mathbf{r}_{i}| > |\mathbf{r}_{i} - \mathbf{r}_{i}| , \qquad (6b)$$

$$\mathcal{R}\boldsymbol{\psi}^{t}(\mathbf{r}_{i}) = \mathcal{R}\boldsymbol{\psi}^{t}(\mathbf{r}_{i}) \cdot \underline{\boldsymbol{S}}_{ii} . \tag{6c}$$

Using the extended boundary condition method [16], we can derive T matrices to relate the unknown coefficients α and f as follows:

$$\mathbf{f}_{e}^{i} = \underline{T}_{e}^{i} \cdot \boldsymbol{\alpha}_{e}^{i}, \quad \mathbf{f}_{m}^{i} = \underline{T}_{m}^{i} \cdot \boldsymbol{\alpha}_{m}^{i} . \tag{7}$$

Substituting Eqs. (5), (6a), and (7) into Eq. (4) we obtain [17]

$$\boldsymbol{\alpha}_{e}^{i} = \mathbf{a}_{e}^{i} + \sum_{j \ (\neq i)} \underline{\underline{G}}_{ij} \cdot \underline{\underline{T}}_{e}^{j} \cdot \boldsymbol{\alpha}_{e}^{j} , \qquad (8a)$$

$$\boldsymbol{\alpha}_{m}^{i} = \mathbf{a}_{m}^{i} + \sum_{j \ (\neq i)} \underline{\underline{G}}_{ij} \cdot \underline{\underline{T}}_{m}^{j} \cdot \boldsymbol{\alpha}_{m}^{j} . \tag{8b}$$

A configurational average is performed over the random positions of the scatterers and the QCA is used [11]. For identical scatterers, we obtain [5]

$$\alpha_e^i(\mathbf{r}_i) = n_0 \int d\mathbf{r}_j g(|\mathbf{r}_j - \mathbf{r}_i|) \underline{\underline{G}}(\mathbf{r}_i, \mathbf{r}_j) \cdot \underline{\underline{T}}_e^j \cdot \alpha_e^j(\mathbf{r}_j) , \qquad (9a)$$

$$\alpha_m^i(\mathbf{r}_i) = n_0 \int d\mathbf{r}_i g(|\mathbf{r}_i - \mathbf{r}_i|) \underline{G}(\mathbf{r}_i, \mathbf{r}_i) \cdot \underline{T}_m^j \cdot \alpha_m^j(\mathbf{r}_i) , \qquad (9b)$$

where n_0 is the number density of scatterers, and $g(|\mathbf{r}_j - \mathbf{r}_i|)$ is the radial distribution function for the spherical case [13].

For the spherical particles considered in this paper, $\underline{\underline{T}}_e$ are diagonal with the explicit expressions given in Ref. [2]. $\underline{\underline{T}}_m$ is obtainable via the replacements of ϵ by μ and μ by ϵ in $\underline{\underline{T}}_e$. When $\mu = \mu_0$ or $\epsilon = \epsilon_0$, Eqs. (9a) and (9b) become a single equation due to the following Maxwell's equations in both background medium and the scatterers:

$$\nabla \times \mathbf{E} = i\omega \mu_0 \mathbf{H} \quad \text{or} \quad \nabla \times \mathbf{H} = -i\omega \epsilon_0 \mathbf{E} . \tag{10}$$

However, when $\epsilon \neq \epsilon_0$ and $\mu \neq \mu_0$. Eqs. (9a) and (9b) do represent two different equations since $\underline{\underline{T}}_e \neq \underline{\underline{T}}_m$. For a conducting scatterer [8-10], $\underline{\underline{T}}^e$ is really different from $\underline{\underline{T}}^m$ [9], and we have two equations (9a) and (9b) to determine the effective permittivity and permeability [10].

Mathematically, the above derivation is trivial since the formalism of Ref. [5] is suitable for both E and H. However, it has the following features. First, the physical concept that the exciting electromagnetic fields of each scatterer are expanded by two unknown vectors is important and different from the classical formalism [11,13]. Second, these equations cannot be easily solved by the well-known approximation [5,13]

$$\boldsymbol{\alpha}^{j}(\mathbf{r}_{i}) = e^{i\mathbf{K}\cdot(\mathbf{r}_{i}-\mathbf{r}_{j})}\boldsymbol{\alpha}^{i}(\mathbf{r}_{i}). \tag{11}$$

Furthermore, one does not know how to determine $\epsilon_{\rm eff}$ and $\mu_{\rm eff}$ except of the effective wave number K [5]. Finally, it provides us a limit that the four-parameter formulation should satisfy. Moreover, the physical concept is easier to be understood than that of the four-parameter formulation in the next subsection.

B. Four-parameter formulation

The pioneering papers [6,18] inspired the present author to extend the low-frequency analysis of the four-parameter composites [6,7] to the multiple-scattering formalism in the resonance range. On the basis of these papers [6,18], the extension is much simpler than five years ago [5].

Following the work [2], the electromagnetic fields in the medium characterized by Eq. (1) can be written as [18]

$$\mathbf{E}(\mathbf{r}) = \mathbf{Q}_1(\mathbf{r}) - i\eta_2 \mathbf{Q}_2(\mathbf{r}) , \qquad (12a)$$

$$\mathbf{H}(\mathbf{r}) = -(i/\eta_1)\mathbf{Q}_1(\mathbf{r}) + \mathbf{Q}_2(\mathbf{r})$$
, (12b)

$$\eta_1 = \eta / \{ \sqrt{1 + k^2 (\alpha - \beta)^2 / 4} - k(\alpha - \beta) / 2 \},$$
(12c)

$$\eta_2 = \eta / \{ \sqrt{1 + k^2 (\alpha - \beta)^2 / 4} + k(\alpha - \beta) / 2 \},$$
(12d)

$$\eta = \sqrt{\mu/\epsilon}, \quad k = \omega \sqrt{\mu \epsilon},$$
(12e)

where the left-handed field Q_1 and the right-handed field Q_2 satisfy the equations [12]

$$\nabla \times \mathbf{Q}_1(\mathbf{r}) = \gamma_1 \mathbf{Q}_1(\mathbf{r}) , \qquad (13a)$$

$$\nabla \times \mathbf{Q}_2(\mathbf{r}) = -\gamma_2 \mathbf{Q}_2(\mathbf{r}) , \qquad (13b)$$

$$\gamma_1 = \{k/(1-k^2\alpha\beta)\}\{\sqrt{1+k^2(\alpha-\beta)^2} + k(\alpha+\beta)/2\}$$
, (13c)

 $\gamma_2 \! = \! \{ k / (1 \! - \! k^2 \alpha \beta) \} \{ \sqrt{1 \! + \! k^2 (\alpha \! - \! \beta)^2} \! - \! k (\alpha \! + \! \beta) / 2 \} \ .$

(13d)

So it is more natural to develop the self-consistent equations using the left-handed field \mathbf{Q}_1 and the right-handed field \mathbf{Q}_2 . Again \mathbf{u}^0 , \mathbf{u}_i^e , and \mathbf{u}_i^s specify the incident field, the field exciting the *i*th scatterer, and the field scattered by the *i*th scatterer, respectively. As shown in Sec. II A, the self-consistency requires that both the left-handed field and the right-handed field must be self-consistent, namely [6],

$$\mathbf{Q}_{1i}^{e} = \mathbf{Q}_{1i}^{0} + \sum_{\substack{i \ (\neq i)}} (\mathbf{Q}_{11j}^{s} + \mathbf{Q}_{12j}^{s}) , \qquad (14a)$$

$$\mathbf{Q}_{2i}^{e} = \mathbf{Q}_{2i}^{0} + \sum_{j \ (\neq i)} (\mathbf{Q}_{21j}^{s} + \mathbf{Q}_{22j}^{s}) , \qquad (14b)$$

with

$$\mathbf{Q}_{11i}^s = T_{11}(\mathbf{Q}_{1i}^e) , \qquad (14c)$$

$$\mathbf{Q}_{12i}^{s} = T_{12}(\mathbf{Q}_{1i}^{e}) , \qquad (14d)$$

$$\mathbf{Q}_{21i}^{s} = T_{21}(\mathbf{Q}_{2i}^{e}) , \qquad (14e)$$

$$\mathbf{Q}_{22i}^{s} = T_{22}(\mathbf{Q}_{2i}^{e}) , \qquad (14f)$$

where subscripts p and q in \mathbf{Q}_{pqj}^s (p,q=1,2) denote the exciting and scattering fields, being \mathbf{Q}_p and \mathbf{Q}_q , respectively, and T_{pq} (p,q=1,2) are the corresponding linear operators transforming the exciting field \mathbf{Q}_p to the scattering field \mathbf{Q}_q . The two conditions (14a) and (14b) can be guaranteed to hold provided [6]

$$\mathbf{Q}_{1i}^{e} = \mathbf{Q}_{1i}^{0} + \sum_{j \ (\neq i)} T_{11}(\mathbf{Q}_{1j}^{e}) , \qquad (15a)$$

$$0 = \sum_{i \ (\neq i)} T_{12}(\mathbf{Q}_{1j}^e) , \qquad (15b)$$

$$0 = \sum_{j \ (\neq i)} T_{21}(\mathbf{Q}_{2j}^e) , \qquad (15c)$$

$$\mathbf{Q}_{2i}^{e} = \mathbf{Q}_{2i}^{0} + \sum_{j \ (\neq i)} T_{22}(\mathbf{Q}_{2j}^{e}) \ . \tag{15d}$$

The physical meaning of Eqs. (15a) and (15b) is that when the exciting field of the *i*th scatterer is the left-handed field only, the consistency requires that the left-handed exciting field of the *i*th scatterer must be equal to the superposition of other scatterer's left-handed fields; meanwhile, the superposition of the right-handed fields of other scatterers must be equal to the right-handed exciting field of the *i*th scatterer, which is zero according to our assumption. Equations (15c) and (15d) can be similarly interpreted.

Notice that when $\alpha = \beta$, Eqs. (15a) and (15b) are not independent [18], then we have the three-parameter formulation to determine (ϵ_{eff} , μ_{eff}).

By the representation theory of linear transformation [19], we can write Eqs. (15a)-(15d) in matrix forms. This can be shown as follows.

The incident, exciting and scattered fields are expanded in terms of the handed spherical vector wave functions [18] [see also Eq. (5)]:

$$\mathbf{Q}_{1i}^{0} = \mathcal{R} \mathbf{L}^{t}(\mathbf{r}_{i}) \cdot \mathbf{a}_{1}^{i} , \qquad (16a)$$

$$\mathbf{Q}_{2i}^{0} = \mathcal{R}\mathbf{R}^{t}(\mathbf{r}_{i}) \cdot \mathbf{a}_{2}^{i} , \qquad (16b)$$

$$\mathbf{Q}_{1i}^{e} = \mathcal{R} \mathbf{L}^{t}(\mathbf{r}_{i}) \cdot \boldsymbol{\alpha}_{1}^{i} , \qquad (16c)$$

$$\mathbf{Q}_{2i}^{e} = \mathcal{R}\mathbf{R}^{t}(\mathbf{r}_{i}) \cdot \boldsymbol{\alpha}_{2}^{i} , \qquad (16d)$$

$$\mathbf{Q}_{1i}^{s} = \mathbf{L}^{t}(\mathbf{r}_{i}) \cdot \mathbf{f}_{1}^{i} , \qquad (16e)$$

$$\mathbf{Q}_{2i}^{s} = \mathbf{R}^{t}(\mathbf{r}_{i}) \cdot \mathbf{f}_{2}^{i} . \tag{16f}$$

Using the T matrices, we can relate the unknowns α and f such that [18]

$$\mathbf{f}_{1i} = \underline{T}_{11} \cdot \boldsymbol{\alpha}_1^i + \underline{T}_{12} \cdot \boldsymbol{\alpha}_2^i , \qquad (17a)$$

$$\mathbf{f}_{2i} = \underline{\underline{T}}_{21} \cdot \boldsymbol{\alpha}_1^i + \underline{\underline{T}}_{22} \cdot \boldsymbol{\alpha}_2^i . \tag{17b}$$

The translation addition theorems for handed spherical wave functions [20] are

$$\mathbf{L}^{t}(\mathbf{r}_{i}) = \mathcal{R}\mathbf{L}^{t}(\mathbf{r}_{i}) \cdot \underline{G}_{1ii}, \quad |\mathbf{r}_{i}| < |\mathbf{r}_{i} - \mathbf{r}_{i}| , \qquad (18a)$$

$$\mathbf{L}^{t}(\mathbf{r}_{j}) = \mathbf{L}^{t}(\mathbf{r}_{i}) \cdot \underline{\underline{S}}_{1ij}, \quad |\mathbf{r}_{i}| > |\mathbf{r}_{j} - \mathbf{r}_{i}| , \qquad (18b)$$

$$\mathbf{R}^{t}(\mathbf{r}_{i}) = \mathcal{R}\mathbf{R}^{t}(\mathbf{r}_{i}) \cdot \underline{\mathbf{G}}_{2ii}, \quad |\mathbf{r}_{i}| < |\mathbf{r}_{i} - \mathbf{r}_{i}| , \qquad (18c)$$

$$\mathbf{R}^{t}(\mathbf{r}_{i}) = \mathbf{R}^{t}(\mathbf{r}_{i}) \cdot \underline{\mathbf{S}}_{2ii}, \quad |\mathbf{r}_{i}| > |\mathbf{r}_{i} - \mathbf{r}_{i}| . \tag{18d}$$

Substituting Eqs. (16)–(18) into Eqs. (16a)–(16d), we obtain

$$\boldsymbol{\alpha}_{1}^{i} = \boldsymbol{a}_{1}^{i} + 6 \sum_{j \ (\neq i)} \underline{\underline{\boldsymbol{G}}}_{1ij} \cdot \underline{\underline{\boldsymbol{T}}}_{11}^{j} \cdot \boldsymbol{\alpha}_{1}^{j} , \qquad (19a)$$

$$0 = \sum_{i \ (\neq i)} \underline{\underline{G}}_{1ij} \cdot \underline{\underline{T}}_{12}^{j} \cdot \alpha_{2}^{j} , \qquad (19b)$$

$$0 = \sum_{\substack{i \ (\neq i)}} \underline{\underline{G}}_{2ij} \cdot \underline{\underline{T}}_{21}^{j} \cdot \alpha_{1}^{j} , \qquad (19c)$$

$$\boldsymbol{\alpha}_{2}^{i} = \mathbf{a}_{2}^{i} + \sum_{j \ (\neq i)} \underline{\underline{G}}_{2ij} \cdot \underline{\underline{T}}_{22}^{j} \cdot \boldsymbol{\alpha}_{2}^{j} \ . \tag{19d}$$

From Eqs. (A6b) and (A7a) of Ref. [21], we see that $\underline{\underline{T}}_{12}^{j} = \underline{\underline{T}}_{21}^{j}$ for the $\alpha = \beta$ case in the notation of Ref. [21]. Substituting the explicit forms of $\underline{\underline{G}}^{1ij}$ and $\underline{\underline{G}}^{2ij}$ into Eqs. (19b) and (19c), we find that for the case of $\alpha = \beta$, these two equations do become a single one.

Similar to the two-parameter case, the configurational average and QCA result in the following equations for identical scatterers:

$$\alpha_1^i(\mathbf{r}_i) = n_0 \int d\mathbf{r}_j g(|\mathbf{r}_j - \mathbf{r}_i|) \underline{\underline{G}}_{1ij} \cdot \underline{\underline{T}}_{11}^j \cdot \alpha_1^j(\mathbf{r}_j)$$
, (20a)

$$0 = \int d\mathbf{r}_{i} g(|\mathbf{r}_{i} - \mathbf{r}_{i}|) \underline{G}_{1ii} \cdot \underline{T}_{12}^{i} \cdot \boldsymbol{\alpha}_{2}^{j}(\mathbf{r}_{i}) , \qquad (20b)$$

$$0 = \int d\mathbf{r}_j \mathbf{g}(|\mathbf{r}_j - \mathbf{r}_i|) \underline{\underline{\mathbf{G}}}_{2ij} \cdot \underline{\underline{T}}_{21}^j \cdot \boldsymbol{\alpha}_1^j(\mathbf{r}_j) , \qquad (20c)$$

$$\alpha_2^i(\mathbf{r}_j) = n_0 \int d\mathbf{r}_j g(|\mathbf{r}_j - \mathbf{r}_i|) \underline{\underline{G}}_{2ij} \cdot \underline{\underline{T}}_{22}^j \cdot \alpha_2^j(\mathbf{r}_j)$$
. (20d)

It is very difficult to solve Eqs. (20a)–(20d) by means of the well-known approximation

$$\boldsymbol{\alpha}_{1}^{i}(\mathbf{r}_{i}) = e^{i\mathbf{K}_{1}\cdot(\mathbf{r}_{i}-\mathbf{r}_{j})}\boldsymbol{\alpha}_{1}^{i}(\mathbf{r}_{i}), \qquad (21a)$$

$$\boldsymbol{\alpha}_{2}^{i}(\mathbf{r}_{i}) = e^{i\mathbf{K}_{2}\cdot(\mathbf{r}_{i}-\mathbf{r}_{j})}\boldsymbol{\alpha}_{2}^{i}(\mathbf{r}_{i}), \qquad (21b)$$

where K_1 and K_2 are the effective left- and right-handed wave numbers. It is possible to introduce the four effective parameters by Eqs. (21) and (12a) and (12b). Also, it is not difficult to derive the multiple-scattering equations in terms of E and H instead of Q_1 and Q_2 . However, these formalism cannot recover the two-parameter limit discussed in Sec. II A. Therefore the approximation given by Eq. (21) are not so useful as that of the single-parameter case [11-13]. In Sec. III we make a try to solve Eqs. (20a)-(20d).

III. DISPERSION EQUATIONS FOR THE GENERAL DISCRETE RANDOM COMPOSITES

In this section, we shall give two physical models to derive the dispersion equations for the general composites. One is called the truncated QCA. Another is the dynamic Maxwell-Garnett model. These two models are discussed in Secs. III A and III B, respectively.

A. Truncated QCA

The truncated QCA presented in this section means that for the exciting field of a given scatterer, we use QCA in the range where the scatterers have a short-range order with their positions given by the pair distribution function, and replace the discrete random composites by an effective homogeneous medium in the range where the scatterers are disordered [22].

Since the discrete random composites is macroscopically homogeneous, for every scatterer the whole effect of the other scatterers in the discrete random composite have no difference if no scatterer has been fixed. In the other words, the exciting fields of every scatterer have the same expression in the scatterer coordinate, namely,

$$\mathbf{u}^{e}(\mathbf{r}_{i}) = \mathcal{R} \boldsymbol{\psi}^{t}(\mathbf{r}_{i}) \cdot \mathbf{a} , \qquad (22a)$$

$$\mathbf{u}^{e}(\mathbf{r}_{i}) = \mathcal{R} \boldsymbol{\psi}^{t}(\mathbf{r}_{i}) \cdot \mathbf{a} . \tag{22b}$$

When the *i*th scatterer is fixed, the positions of the other scatterers are given by the pair distribution function, and the exciting fields may vary. But by the QCA, we can assume

$$\langle \mathbf{u}^{e}(\mathbf{r}_{i})\rangle_{ii} = \langle \mathbf{u}^{e}(\mathbf{r}_{i})\rangle_{i} = \mathbf{u}^{e}(\mathbf{r}_{i}) = \mathcal{R}\psi^{t}(\mathbf{r}) \cdot \mathbf{a}$$
 (23)

Notice that [13] if $|\mathbf{r}_j - \mathbf{r}_i| \ge 10a$, where a is the radius of the scatterer, the pair distribution function $g(|\mathbf{r}_j - \mathbf{r}_i|) \equiv 1$. This means that for the exciting fields of the *i*th scatterer $\mathbf{u}^e(\mathbf{r}_i)$, all the scatterers outside the sphere $|\mathbf{r}_j - \mathbf{r}_i| = 10a$ are uncorrelated with the *i*th scatterer. Therefore, for the consideration of $\mathbf{u}^e(\mathbf{r}_i)$, the composite can be subdivided into two regions, as shown in Fig. 1. Region I contains the scatterers whose positions are given by the pair distribution function [13], whereas all the interactions of the scatterers in region II are replaced by the effective medium [22,23]. This is the model that we use to calculate the medium's effective parameters.

The truncated QCA model can be further interpreted as follows.

(i) The outer radius 10a is unchanged in our model

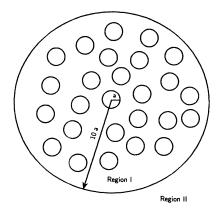


FIG. 1. The effective model for the exciting field of the *i*th scatterer.

whether the scatterers in region I is in order or disorder. It therefore includes the case when all the scatterers in region I are also uncorrelated. For this case, this model recovers the low-frequency limit. This can be proved by comparing the present model with the Maxwell-Garnett model [24] and remembering the discussion on these two complementary models by Born and Wolf in their well-known treatise [23]. Notice that there are two steps in the proof: one is the effectiveness of the composite at low frequency and in the Maxwell-Garnett model [24]; another is the effectiveness of the Maxwell-Garnett model and its complementary configuration [23,24].

- (ii) Since the multiple-scattering formulation is developed by the self-consistency of the *i*th scatterer, this model is for this purpose only.
- (iii) It is well known that the effective parameters of the discrete random composites depend on the microstructure of the composite. The truncated QCA uses the short-range order of the medium to obtain the medium's effective parameters. By this model, the medium's effective parameters are naturally introduced.

Equation (9a) is taken as an illustrative example for the derivation of dispersion equations.

From Eqs. (5) and (9), setting $\mathbf{r}_i = 0$, we obtain

$$\mathbf{a} = n_0 \int_{2a}^{10a} d\mathbf{r} \, g(r) \underline{\underline{G}}(r) \cdot \underline{\underline{T}}_e \cdot \mathbf{a} + \underline{\underline{t}}_e \cdot \left[\mathbf{a} + n_0 \int_{2a}^{10a} d\mathbf{r} \, g(r) \underline{\underline{S}}(r) \cdot \underline{\underline{T}}_e \cdot \mathbf{a} \right] . \tag{24}$$

Thus for a nontrivial solution to a, we obtain the dispersion equation

$$\det \left| \underline{\underline{I}} - \underline{\underline{t}}_{e} - n_{0} \int_{2a}^{10a} d\mathbf{r} g(\mathbf{r}) [\underline{\underline{G}}(\mathbf{r}) + \underline{\underline{t}}_{e} \cdot \underline{\underline{S}}(\mathbf{r})] \cdot \underline{\underline{T}}_{e} \right| = 0 ,$$
(25)

where the addition theorem (6a) has been used, the effective medium's exciting field is obtained by the addition theorem (6b), the scattering fields of the effective medium exciting on the *i*th scatterer is obtained by the reflective matrix \underline{t}_e [16,25], and \underline{I} is the unit matrix [5].

Similarly, we obtain another dispersion equation for the two-parameter case:

$$\det \left| \underline{\underline{I}} - \underline{\underline{t}}_m - n_0 \int_{2a}^{10a} d\mathbf{r} \, g(r) [\underline{\underline{G}}(\mathbf{r}) + \underline{\underline{t}}_m \cdot \underline{\underline{S}}(r)] \cdot \underline{\underline{T}}_m \right| = 0 \ . \tag{26}$$

Let \underline{t}_{11} , \underline{t}_{12} , \underline{t}_{21} , and \underline{t}_{22} indicate the reflective matrices [16,25] in the four-parameter case. The addition theorems (18a) and (18c) and (18b) and (18d) are invoked. Finally, we obtain the dispersion equations for the four-parameter case as follows:

$$\det \left| \underline{\underline{I}} - \underline{\underline{t}}_{11} - n_0 \int_{2a}^{10a} d\mathbf{r} g(\mathbf{r}) [\underline{\underline{G}}_{1}(\mathbf{r}) - \underline{\underline{t}}_{11} \cdot \underline{\underline{S}}_{1}(\mathbf{r})] \cdot \underline{\underline{T}}_{11} \right| = 0 ,$$
(27a)

$$\det \left| \int_{2a}^{10a} d\mathbf{r} g(\mathbf{r}) [\underline{\underline{G}}_{1}(\mathbf{r}) - \underline{\underline{t}}_{12} \cdot \underline{\underline{S}}_{1}(\mathbf{r})] \cdot \underline{\underline{T}}_{12} \right| = 0 , \qquad (27b)$$

$$\det \left| \int_{2a}^{10a} d\mathbf{r} \, g(r) [\underline{\underline{G}}_{2}(r) - \underline{\underline{t}}_{21} \cdot \underline{\underline{S}}_{2}(r)] \cdot \underline{\underline{T}}_{21} \right| = 0 , \qquad (27c)$$

$$\det \left| \underline{\underline{I}} - \underline{\underline{t}}_{22} - n_0 \int_{2a}^{10a} d\mathbf{r} g(\mathbf{r}) [\underline{\underline{G}}_2(\mathbf{r}) - \underline{\underline{t}}_{22} \cdot \underline{\underline{S}}_2(\mathbf{r})] \cdot \underline{\underline{T}}_{22} \right| = 0 \ .$$

(27d)

From Eqs. (44b) and (45a) of Ref. [21], it is evident that for the $\alpha = \beta$ case, Eqs. (27b) and (27c) merge into a single equation. This is as expected. As pointed out by Schwartz [8], the pair distribution function cannot include exactly the interactions of dipoles at low frequency since it is the function of r only. Therefore, in the low-frequency limit, all the formulas derived by OCA with the pair distribution function become the Maxwell-Garnett [24] mixing formula for the disorder case. The same conclusion is suitable for the present formulation.

Notice that instead of Eq. (22), we can also use the approximation (11) or (21a) and (21b) to derive the dispersion equations under the present model. So we have two methods to derive the dispersion equations. Which method is better depends on the future experiments.

B. Dynamic Maxwell-Garnett model

Recently, the Maxwell-Garnett model [24] has been used to calculate the effective dielectric constant at low frequency via the Monte Carlo simulation [24,26]. In this subsection, we generalize the well-known Maxwell-Garnett model to the resonance range. We call this model as dynamic Maxwell-Garnett model [9].

If the cluster of spheres is viewed as a scatterer with effective parameter [24], then the incident and scattering fields can be written as

$$\mathbf{u} = \mathcal{R} \boldsymbol{\psi}_t(\mathbf{r}_0) \cdot \mathbf{a} , \qquad (28a)$$

$$\mathbf{u}^{s} = \psi_{t}(\mathbf{r}_{0}) \cdot \underline{T}^{\text{eff}} \cdot \mathbf{a} , \qquad (28b)$$

where $\underline{\underline{T}}^{\text{eff}}$ is the T matrix of the effective scatterer. Meanwhile, the scattering field can also be expressed as [16,24]

$$\mathbf{u}^{s} = \boldsymbol{\psi}_{t}(\mathbf{r}_{0}) \cdot \underline{\tau}_{(N)} \cdot \mathbf{a} , \qquad (29)$$

where $\underline{\tau}_{(N)}$ is the average aggregate T matrix for N scatterers. The calculation is repeated by the recursive T-matrix algorithms [16,24] for each realization and the

 $\underline{\underline{\tau}}_{(N)}$ is averaged over N, realizations; each realization is generated by the Monte Carlo simulation [24,26].

The self-consistency requires that

$$\underline{T}^{\text{eff}} \cdot \mathbf{a} = \underline{\tau}_{(N)} \cdot \mathbf{a} . \tag{30}$$

The nontrivial solution to a leads to the dispersion equation

$$\det |\underline{T}^{\text{eff}} - \underline{\tau}_{(N)}| = 0 . \tag{31}$$

This result recovers the low-frequency limit [24]. When the scatterers become sparse and disordered, the above tedious calculation can be avoided. Instead, after the EFA is used for the scatterers in a spherical region [27], the closed form of the average aggregate T matrix for all the scatterers inside the sphere can be derived [27].

The above procedure can be easily generalized to the two- and four-parameter cases. The results are

$$\det \left| \underline{\underline{T}}_{e}^{\text{eff}} - \underline{\underline{\tau}}_{e(N)} \right| = 0 , \qquad (32a)$$

$$\det \left| \underline{\underline{T}}_{m}^{\text{eff}} - \underline{\underline{\tau}}_{m(N)} \right| = 0 \tag{32b}$$

for the two-parameter case and

$$\det |\underline{T}_{pq}^{\text{eff}} - \underline{\tau}_{pq(N)}| = 0, \quad p, q = 1, 2$$
(33)

for the four-parameter case.

IV. CONCLUSION

In this paper, the problem of how to calculate the four effective parameters for the bi-isotropic composites in the resonance range has been presented. The multiple-scattering self-consistent equations are obtained. Two approaches of deriving the dispersion equations are suggested. All the results recover the Maxwell-Garnett mixing formula at low frequency. The degenerative two- and three-parameter cases are also discussed due to both their practical importance and the requirement of the inherent unity of our theory. The methods of this paper can be extended to the following case:

- (i) The background medium is also bi-isotropic.
- (ii) The scatterers are randomly distributed and oriented [28].
- (iii) The bianisotropic spheres are randomly distributed in a host bi-isotropic medium since the T matrix [28] of a bianisotropic sphere can be derived via the wave-function theory of anisotropic media [29].

This paper's formalism probably would be useful in the theoretical description of the discrete random composites with both electric and magnetic fluctuations, such as granular metals and unmagnetized ferrites, as well as the discrete random composites with cross coupling electric and magnetic fluctuations, such as chiral particulate composites.

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