Strong-property-fluctuation theory for homogenizing chiral particulate composites

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A statistical description of the electromagnetic wave propagation in a two-component chiral composite is presented. We develop the strong-property-fluctuation theory which is a generalization of the strong-permittivity-fluctuation theory for nonhomogeneous chiral media. The Dyson equation for the exciting electromagnetic field is solved in the bilocal approximation. Wave propagation in the composite can be described in this manner by a nonlocal effective medium containing information about the spatial correlations of the material properties. For length scales larger than the correlation length, the system may be homogenized and we obtain a local effective medium theory.

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I. INTRODUCTION

Isotropic chiral media are ubiquitous in the literature on physical chemistry [1] and have gained prominent mention in the areas of electromagnetics and optics, too [2]. Naturally occurring, isotropic chiral media have molecules with mirror-asymmetric conformations. When an electromagnetic plane wave traverses a chiral medium of a certain thickness, it emerges with its vibration ellipse completely altered: The handedness of the chiral molecules causes the rotation of the major axis and changes the eccentricity of the vibration ellipse. These two phenomena—named *optical rotation* and *circular dichroism,* respectively—are together called *natural optical activity.*

In nature, isotropic chiral media display their unique behavior only at optical frequencies. Naturally occurring materials have not been found to be appreciably optically active at the lower microwave frequencies. Recent advances in the construction of artificial chiral materials in the form of particulate composites are, however, promising [3] and have motivated the formulation of *effective medium* or *homogenization* theories.

Several effective medium rules have been set up for chiral composites [4], which are the analogs to the Maxwell-Garnett and the Bruggeman mixing rules for dielectric composites. These rules are derived under certain assumptions that may not always be justified. Let f denote the volumetric proportion of the particulate material $0 \leq f \leq 1$. Single-scattering mixing rules are valid only for very small f [5,6]. Although it has never been experimentally ascertained that the Maxwell-Garnett (MG) rule for chiral composites is valid only for dilute suspensions of chiral particles in an achiral matrix medium, we suspect it to be thus from studies on dielectric composites [5]. For high values of f it will fail to give reliable results, because of the neglect of correlation effects between neighboring particles; see, e.g., [7]. In the extreme case $f=1$ where the particulate medium alone is present, the effective medium and the particulate medium should be the same; this turns out to be true for the MG rule [8], but not for an extended MG rule [9] wherein the effect of the finite sizes of the particles have been meaningfully considered.

Moreover, the MG rule and its variants cannot predict the percolation threshold at all, because the electromagnetic properties of the particulate and the matrix media are incorporated asymmetrically therein [10]. That is not a problem with the Bruggeman rule, in which the two sets of properties are treated at par with one another [11]. However, there is a major drawback in the Bruggeman rule: it always yields $f=\frac{1}{3}$ for the percolation threshold in dielectric composites, without regard to the shapes and the sizes of the particles. This cannot be true, e.g., for a composite with filamentary particles; we should expect percolation well below $f=\frac{1}{3}$.

What one apparently needs to do, in order to improve the quality of homogenization predictions for chiral and other composites, is to take into account the microstructural topology in a more detailed fashion, but without inadvertently subverting the concept of homogenization [9,12]. Beyond the filling factor f, which gives only the minimal geometric information, one should at least include the effects of spatial correlations between the particles relative to the electromagnetically significant length scales for dense composites.

An attractive theoretical approach often used to describe wave propagation in nonhomogeneous dielectric media, which includes these correlation effects rigorously, is the Feynman-diagrammatic technique of Frisch [13]. In this approach for the homogenization of particulate composites, one must make a preliminary ansatz about the nature of the homogenized medium, and then use this approach to perturbatively calculate corrections in orders of the statistical cumulants of the spatial distribution of the particles. However, the straightforward application of this approach is limited to very small spatial fluctuations of the material properties, i.e., when the contrast between the particulate and the matrix permittivities is small. This limitation is due to secular terms in the per-

turbation expansion of the electric field equation. The secular terms are generated by the singularity of the dyadic Green function [14]. But, by properly treating this singularity, one can reformulate the approach to make it applicable for strong spatial fluctuations of the permittivity. This theoretical approach is called the strong-permittivity-fluctuation theory, and so far it has only been applied to dielectric mixtures [15].

Our goal in this paper is to generalize this approach to chiral-in-chiral composites, i.e., particulate composites in which the particulate, as well as the matrix media, are chiral. We shall call this approach the strong-property-fluctuation theory (SPFT). For the derivation of the SPFT, we shall closely follow the argumentation in [15], translating it step by step into a formalism for chiral-in-chiral composites. Let us also observe that, as the particulate and matrix materials are treated at par with one another in the SPFT, it is sensible to refer to a chiral-in-chiral composite simply as a *two-component mixture.* This term is particularly relevant when f is not small. Parenthetically, we note that our approach can also be used for homogenizing bi-isotropic–in–bi-isotropic composites [16]; however, it has been recently shown that bi-isotropic media are not allowed in modern electromagnetic theory [17].

This paper is organized as follows: After some general statements about chiral media and particulate composites, we explain the structures of the various dyadic Green functions in a homogeneous chiral medium. We then discuss the Dyson equation for the average electromagnetic field in a two-component mixture and obtain the lowest-order estimate of the effective medium properties. This lowest-order estimate is exactly the same as the one from the Bruggeman rule for chiral-in-chiral composites [11], and it also serves as the preliminary ansatz for our perturbative approach. We derive the relation between the exact effective medium, which includes all correlation effects, and the so-called mass operator of the Dyson equation. Then we investigate the behavior of the mixture on a length scale that is larger than the correlation length, so that the two-component mixture can be regarded as a homogeneous medium. Finally, we implement the SPFT in the bilocal approximation [18]. This paper is exclusively devoted to the formulation of the SPFT for two-component chiral mixtures, and applications are planned for future papers.

II. GENERAL

We start with the frequency-dependent version of the source-free Maxwell curl postulates,

$$\nabla\times\mathbf{E}(\mathbf{r})=i\omega\mathbf{B}(\mathbf{r}) , \tag{1}$$

$$\nabla\times\mathbf{H}(\mathbf{r})=-i\omega\mathbf{D}(\mathbf{r}) , \tag{2}$$

where we have assumed an $\exp(-i\omega t)$ time dependence. The Drude-Born-Fedorov constitutive relations of a nonhomogeneous chiral medium are given as [4]

$$\mathbf{D}(\mathbf{r})=\epsilon(\mathbf{r})[\mathbf{E}(\mathbf{r})+\beta(\mathbf{r})\nabla\times\mathbf{E}(\mathbf{r})] , \tag{3}$$

$$\mathbf{B}(\mathbf{r})=\mu(\mathbf{r})[\mathbf{H}(\mathbf{r})+\beta(\mathbf{r})\nabla\times\mathbf{H}(\mathbf{r})] , \tag{4}$$

where $\epsilon(\mathbf{r})$ is the permittivity scalar, $\mu(\mathbf{r})$ is the permeability scalar, and $\beta(\mathbf{r})$ is the chirality pseudoscalar, all three being implicit functions of the circular frequency ω. In Eqs. (3) and (4) we explicitly allow for position-dependent material properties for notational compactness.

From the Maxwell postulates (1) and (2) and the constitutive relations (3) and (4), we find the first-order field equation [19]

$$\nabla\times\mathbf{F}(\mathbf{r})=\mathcal{K}(\mathbf{r})\mathbf{F}(\mathbf{r}) , \tag{5}$$

where

$$\mathbf{F}(\mathbf{r})=\begin{bmatrix}\mathbf{E}(\mathbf{r})\\ \mathbf{H}(\mathbf{r})\end{bmatrix} , \tag{6}$$

$$\mathcal{K}(\mathbf{r})=\gamma^{+}(\mathbf{r})\gamma^{-}(\mathbf{r})\begin{bmatrix}\beta(\mathbf{r}) & \dfrac{i\omega\mu(\mathbf{r})}{k(\mathbf{r})^2}\\ -\dfrac{i\omega\epsilon(\mathbf{r})}{k(\mathbf{r})^2} & \beta(\mathbf{r})\end{bmatrix} , \tag{7}$$

$k(\mathbf{r})=\omega\sqrt{\epsilon(\mathbf{r})\mu(\mathbf{r})}$ and $\gamma^{\pm}(\mathbf{r})=k(\mathbf{r})/[1\mp k(\mathbf{r})\beta(\mathbf{r})]$. For later convenience, we rewrite the constitutive relations (3) and (4) in compact matrix notation as

$$\mathbf{C}(\mathbf{r})=\mathcal{J}\mathcal{K}(\mathbf{r})\mathbf{F}(\mathbf{r}) , \tag{8}$$

with

$$\mathbf{C}(\mathbf{r})=\begin{bmatrix}\mathbf{D}(\mathbf{r})\\ \mathbf{B}(\mathbf{r})\end{bmatrix} , \quad \mathcal{J}=\frac{1}{\omega}\begin{bmatrix}0 & i\\ -i & 0\end{bmatrix} . \tag{9}$$

III. TWO-COMPONENT MIXTURE

We now look at a two-component mixture consisting of two chiral components mixed at the microscopic, but not molecular, length scale. Let all space be divided into two disjoint parts V_a and V_b containing the components labeled a and b, respectively. For $\mathbf{r}\in V_p$ $(p=a,b)$ we write

$$\epsilon(\mathbf{r})=\epsilon_p , \quad \beta(\mathbf{r})=\beta_p , \quad \mu(\mathbf{r})=\mu_p , \tag{10}$$

so that

$$\gamma^{\pm}(\mathbf{r})=\gamma_p^{\pm} , \quad k(\mathbf{r})=k_p , \tag{11}$$

$$\mathcal{K}(\mathbf{r})=\mathcal{K}_p . \tag{12}$$

We introduce the characteristic functions θ_p as

$$\theta_p(\mathbf{r})=\begin{cases}1 & \text{for } \mathbf{r}\in V_p\\ 0 & \text{elsewhere ;}\end{cases} \tag{13}$$

thus,

$$\theta_a(\mathbf{r})+\theta_b(\mathbf{r})=1 . \tag{14}$$

Any of the $\mathbf{r}$-dependent quantities occurring in Eqs. (10)–(12) can be expressed *everywhere* in terms of the characteristic functions $\theta_p(\mathbf{r})$; for example,

$$\mathcal{K}(\mathbf{r})=\mathcal{K}_a\theta_a(\mathbf{r})+\mathcal{K}_b\theta_b(\mathbf{r}) . \tag{15}$$

Let us now turn to a statistical description of the mixture. Throughout the paper we shall use the concept of ensemble averaging, i.e., averaging over a large number of different samples of the two-component mixture. We denote ensemble averages by $\langle\ \rangle$. Only in Sec. IV shall we also need *spatially* averaged quantities in order to define macroscopic fields. The complete statistical information about the mixture is contained in the *moments* of the characteristic function $\theta_a(\mathbf{r})$. The nth moment is the expectation value $\langle \theta_a(\mathbf{r}_1), \ldots, \theta_a(\mathbf{r}_n) \rangle$ and is the probability for $\mathbf{r}_1, \ldots, \mathbf{r}_n$ being inside the component a (equivalently we may use b instead of a). We assume that, on average, the mixture is homogeneous and isotropic.

The first moment for the component a is the filling factor

$$f_a = \langle \theta_a(\mathbf{r}) \rangle \ , \tag{16}$$

which is constant with respect to $\mathbf{r}$. The same holds for the filling factor of the component b, which is defined by $f_b = \langle \theta_b(\mathbf{r}) \rangle$. Obviously,

$$f_a + f_b = 1 \ . \tag{17}$$

These two filling factors contain only the minimal geometrical information about the mixture: namely, the volume fractions of the two components. A more detailed description is provided by the second moment of the characteristic function, $\langle \theta_a(\mathbf{r}) \theta_a(\mathbf{r}') \rangle$, or, equivalently, by the second *cumulant* or covariance

$$\tau_a(R) = \langle \theta_a(\mathbf{r}) \theta_a(\mathbf{r}') \rangle - \langle \theta_a(\mathbf{r}) \rangle \langle \theta_a(\mathbf{r}') \rangle \ . \tag{18}$$

Because the two components are supposed to be isotropically mixed, $\tau_a(R)$ depends only on the distance $R = |\mathbf{r} - \mathbf{r}'|$. If the mixture is disordered—and for most practical applications of the SPFT this assumption is justified—it is usually possible to define a correlation length L such that $\tau_a(L)/\tau_a(0) = e^{-1}$ and that $\tau_a(R)$ is negligible for $R \gg L$. On scales larger than L the mixture may be considered homogeneous.

Higher-order cumulants can be defined in a similar way, as in Eq. (18); see [13]. In fact, for a rigorous statistical treatment of the effective medium problem, cumulants of arbitrarily high order are required, but in this paper we assume that the filling factor (16) and its covariance (18) suffice to describe the mixture.

IV. EQUATIONS FOR A LOCAL EFFECTIVE MEDIUM

We assume that electromagnetic wave propagation in $V_a \cup V_b$ can be approximately described with the help of an *effective medium* with constitutive parameters ϵ_{Br}, β_{Br}, and μ_{Br}. These parameters are not $\mathbf{r}$ dependent; hence this effective medium is not only homogeneous but also spatially *local*. This effective medium will later on serve as the preliminary ansatz for the SPFT and will be shown in the next section to actually be in complete agreement with the Bruggeman rule [11].

In analogy to the formulas given earlier, we use the notation $k_{\mathrm{Br}} = \omega \sqrt{\epsilon_{\mathrm{Br}} \mu_{\mathrm{Br}}}$, $\gamma_{\mathrm{Br}}^{\pm} = k_{\mathrm{Br}} / (1 \mp k_{\mathrm{Br}} \beta_{\mathrm{Br}})$, and

$$\mathcal{K}_{\mathrm{Br}} = \gamma_{\mathrm{Br}}^{+} \gamma_{\mathrm{Br}}^{-} \begin{bmatrix} \beta_{\mathrm{Br}} & \dfrac{i\omega\mu_{\mathrm{Br}}}{k_{\mathrm{Br}}^2} \\ -\dfrac{i\omega\epsilon_{\mathrm{Br}}}{k_{\mathrm{Br}}^2} & \beta_{\mathrm{Br}} \end{bmatrix} . \tag{19}$$

For later convenience, we also introduce the intrinsic impedance $\eta_{\mathrm{Br}} = \sqrt{\mu_{\mathrm{Br}}/\epsilon_{\mathrm{Br}}}$. Propagation in this local effective medium can be described by

$$\nabla \times \mathbf{F}_{\mathrm{Br}}(\mathbf{r}) = \mathcal{K}_{\mathrm{Br}} \mathbf{F}_{\mathrm{Br}}(\mathbf{r}) \ , \tag{20}$$

with

$$\mathbf{F}_{\mathrm{Br}}(\mathbf{r}) = \begin{bmatrix} \mathbf{E}_{\mathrm{Br}}(\mathbf{r}) \\ \mathbf{H}_{\mathrm{Br}}(\mathbf{r}) \end{bmatrix} \tag{21}$$

denoting the spatially averaged electromagnetic field. At this stage, it is appropriate to define the Green matrix [20]

$$\mathcal{G}_{\mathrm{Br}}(\mathbf{r} - \mathbf{r}') = \begin{bmatrix} G_{\mathrm{Br}}^{\mathrm{EE}}(\mathbf{r} - \mathbf{r}') & G_{\mathrm{Br}}^{\mathrm{EH}}(\mathbf{r} - \mathbf{r}') \\ G_{\mathrm{Br}}^{\mathrm{HE}}(\mathbf{r} - \mathbf{r}') & G_{\mathrm{Br}}^{\mathrm{HH}}(\mathbf{r} - \mathbf{r}') \end{bmatrix} , \tag{22}$$

which corresponds to the differential equation (20) with the usual radiation conditions fulfilled; that is,

$$\nabla \times \mathcal{G}_{\mathrm{Br}}(\mathbf{r} - \mathbf{r}') - \mathcal{K}_{\mathrm{Br}} \mathcal{G}_{\mathrm{Br}}(\mathbf{r} - \mathbf{r}') = \begin{bmatrix} \mathbf{I} & 0 \\ 0 & \mathbf{I} \end{bmatrix} \delta(\mathbf{r} - \mathbf{r}') \ . \tag{23}$$

The four components of the Green matrix $\mathcal{G}_{\mathrm{Br}}(\mathbf{r} - \mathbf{r}')$ are dyadic Green functions, $\mathbf{I}$ is the identity dyadic, and $\delta(\mathbf{r} - \mathbf{r}')$ is the Dirac delta.

In order to obtain the explicit form of the Green matrix $\mathcal{G}_{\mathrm{Br}}(\mathbf{r} - \mathbf{r}')$, we transform the matrix equation (23) into the so-called Beltrami representation [4]. We introduce the transformation matrix [19]

$$\mathcal{A} = \frac{1}{2} \begin{bmatrix} 1 & i\eta_{\mathrm{Br}} \\ i/\eta_{\mathrm{Br}} & 1 \end{bmatrix} , \tag{24}$$

which has the inverse

$$\mathcal{A}^{-1} = \begin{bmatrix} 1 & -i\eta_{\mathrm{Br}} \\ -i/\eta_{\mathrm{Br}} & 1 \end{bmatrix} . \tag{25}$$

The transformed Green matrix

$$\mathcal{G}_Q(\mathbf{r} - \mathbf{r}') = \mathcal{A} \mathcal{G}_{\mathrm{Br}}(\mathbf{r} - \mathbf{r}') \mathcal{A}^{-1} \tag{26}$$

satisfies the Beltrami field equation [4] with a point-source excitation as per

$$\nabla \times \mathcal{G}_Q(\mathbf{r} - \mathbf{r}') - \Lambda \mathcal{G}_Q(\mathbf{r} - \mathbf{r}') = \begin{bmatrix} \mathbf{I} & 0 \\ 0 & \mathbf{I} \end{bmatrix} \delta(\mathbf{r} - \mathbf{r}') \ . \tag{27}$$

The transformation is such that

$$\Lambda = \mathcal{A} \mathcal{K}_{\mathrm{Br}} \mathcal{A}^{-1} = \begin{bmatrix} \gamma_{\mathrm{Br}}^{+} & 0 \\ 0 & -\gamma_{\mathrm{Br}}^{-} \end{bmatrix} \tag{28}$$

is diagonal, and so is

$$\mathcal{G}_Q(\mathbf{R})=\begin{bmatrix} G^+(\gamma_{\rm Br}^+|\mathbf{R}) & 0 \\ 0 & G^-(\gamma_{\rm Br}^-|\mathbf{R}) \end{bmatrix} . \tag{29}$$

The Beltrami-Green dyadics $G^{\pm}(\sigma|\mathbf{R})$ outside the source region ($R\neq 0$) can be stated as [4]

$$G^{\pm}(\sigma|\mathbf{R})=\pm\frac{\sigma}{4\pi}\left\{\frac{2}{3}\mathbf{I}\pm\left[i-\frac{1}{\sigma R}\right]\mathbf{u}_{\mathbf{R}}\times\mathbf{I} - \left[1+\frac{3i}{\sigma R}-\frac{3}{(\sigma R)^2}\right]\times[\mathbf{u}_{\mathbf{R}}\mathbf{u}_{\mathbf{R}}-\tfrac{1}{3}\mathbf{I}]\right\}g(\sigma;R) , \tag{30}$$

with the unit vector $\mathbf{u}_{\mathbf{R}}=\mathbf{R}/R$ and the scalar Green function $g(\sigma;R)=e^{i\sigma R}/R$.

The singular behavior of the Beltrami-Green dyadics may be formally expressed through [21]

$$G^{\pm}(\sigma|\mathbf{R})=\mathcal{P}G^{\pm}(\sigma|\mathbf{R})\mp\frac{\mathbf{I}}{3\sigma}\delta(\mathbf{R}) , \tag{31}$$

where $\mathcal{P}$ is the principal value operation excluding an infinitesimal spherical region around $R=0$. Inserting Eq. (31) into Eq. (29), we find the matrix identity

$$\mathcal{G}_Q(\mathbf{R})=\mathcal{P}\mathcal{G}_Q(\mathbf{R})-\tfrac{1}{3}\Lambda^{-1}\delta(\mathbf{R}) . \tag{32}$$

The Green matrix $\mathcal{G}_{\rm Br}(\mathbf{r}-\mathbf{r}')$ can be calculated using Eq. (26) as

$$G_{\rm Br}^{\rm EE}(\mathbf{R})=\tfrac{1}{2}[G^+(\gamma_{\rm Br}^+|\mathbf{R})+G^-(\gamma_{\rm Br}^-|\mathbf{R})] , \tag{33}$$

$$G_{\rm Br}^{\rm EH}(\mathbf{R})=\frac{i\eta_{\rm Br}}{2}[G^+(\gamma_{\rm Br}^+|\mathbf{R})-G^-(\gamma_{\rm Br}^-|\mathbf{R})] , \tag{34}$$

$$G_{\rm Br}^{\rm HE}(\mathbf{R})=\frac{1}{2i\eta_{\rm Br}}[G^+(\gamma_{\rm Br}^+|\mathbf{R})-G^-(\gamma_{\rm Br}^-|\mathbf{R})] , \tag{35}$$

$$G_{\rm Br}^{\rm HH}(\mathbf{R})=G_{\rm Br}^{\rm EE}(\mathbf{R}) , \tag{36}$$

and we thus establish that the singular behavior of $\mathcal{G}_{\rm Br}(\mathbf{r},\mathbf{r}')$ at $\mathbf{r}=\mathbf{r}'$ can be accommodated through

$$\mathcal{G}_{\rm Br}(\mathbf{R})=\mathcal{P}\mathcal{G}_{\rm Br}(\mathbf{R})-\tfrac{1}{3}\mathcal{K}_{\rm Br}^{-1}\delta(\mathbf{R}) , \tag{37}$$

a result of great importance in the following section.

V. DYSON EQUATION

The stage is now set to obtain the Dyson equation for the ensemble-averaged electromagnetic field. We begin by combining Eqs. (5) and (20) as

$$\nabla\times\mathbf{F}(\mathbf{r})-\mathcal{K}_{\rm Br}\mathbf{F}(\mathbf{r})=[\mathcal{K}(\mathbf{r})-\mathcal{K}_{\rm Br}]\mathbf{F}(\mathbf{r}) . \tag{38}$$

Using $\mathcal{G}_{\rm Br}(\mathbf{r}-\mathbf{r}')$ and $\mathbf{F}_{\rm Br}(\mathbf{r})$, next we transform this equation to the following integral equation:

$$\mathbf{F}(\mathbf{r})=\mathbf{F}_{\rm Br}(\mathbf{r})+\int d^3r'\mathcal{G}_{\rm Br}(\mathbf{r}-\mathbf{r}')[\mathcal{K}(\mathbf{r}')-\mathcal{K}_{\rm Br}]\mathbf{F}(\mathbf{r}') . \tag{39}$$

Equation (39) cannot be evaluated perturbatively when the constitutive parameters $\epsilon(\mathbf{r})$, $\mu(\mathbf{r})$, and $\beta(\mathbf{r})$ fluctuate strongly. This is due to secular terms produced by the singularities of the dyadic Green functions in the source region [14]. The singularities can be removed from the right side of Eq. (39) by taking advantage of Eq. (37); thus,

$$\mathbf{F}(\mathbf{r})=\mathbf{F}_{\rm Br}(\mathbf{r})+\mathcal{P}\int d^3r'\mathcal{G}_{\rm Br}(\mathbf{r}-\mathbf{r}')[\mathcal{K}(\mathbf{r}')-\mathcal{K}_{\rm Br}]\mathbf{F}(\mathbf{r}') -\tfrac{1}{3}\mathcal{K}_{\rm Br}^{-1}[\mathcal{K}(\mathbf{r})-\mathcal{K}_{\rm Br}]\mathbf{F}(\mathbf{r}) . \tag{40}$$

Next, after introducing the *exciting field*

$$\mathbf{F}_{\rm exc}(\mathbf{r})=\tfrac{1}{3}\mathcal{K}_{\rm Br}^{-1}[\mathcal{K}(\mathbf{r})+2\mathcal{K}_{\rm Br}]\mathbf{F}(\mathbf{r}) , \tag{41}$$

we rewrite the integral equation (39) as

$$\mathbf{F}_{\rm exc}(\mathbf{r})=\mathbf{F}_{\rm Br}(\mathbf{r})+\mathcal{P}\int d^3r'\mathcal{G}_{\rm Br}(\mathbf{r}-\mathbf{r}')\mathcal{X}(\mathbf{r}')\mathbf{F}_{\rm exc}(\mathbf{r}') , \tag{42}$$

with a generalized *polarizability matrix* defined as

$$\mathcal{X}(\mathbf{r})=3[\mathcal{K}(\mathbf{r})-\mathcal{K}_{\rm Br}][\mathcal{K}(\mathbf{r})+2\mathcal{K}_{\rm Br}]^{-1}\mathcal{K}_{\rm Br} . \tag{43}$$

Equations (42) and (43) together constitute the basis for the strong-property-fluctuation theory for two-component chiral mixtures.

The next steps are canonical: We calculate the ensemble averages of the exciting field $\langle\mathbf{F}_{\rm exc}(\mathbf{r})\rangle$ by ensemble averaging both sides of the integral equation (42). For this purpose, we formally represent this equation in terms of a Born series and average each term of the series separately. Details of this procedure are available in the literature [13,22].

Now let us fix the lowest-order estimate of the effective medium properties, mentioned in the previous section, by demanding that

$$\langle\mathcal{X}(\mathbf{r})\rangle=0 . \tag{44}$$

This condition may look somewhat arbitrary at first glance, but it can be shown [14] to be necessary for removing the secular terms from the Born series expansion. Inserting Eqs. (15) and (16) into Eq. (44), we obtain

$$[\mathcal{K}_a-\mathcal{K}_{\rm Br}][\mathcal{K}_a+2\mathcal{K}_{\rm Br}]^{-1}f_a +[\mathcal{K}_b-\mathcal{K}_{\rm Br}][\mathcal{K}_b+2\mathcal{K}_{\rm Br}]^{-1}f_b=0 , \tag{45}$$

which is nothing but the Bruggeman mixing rule for chiral media, first given in [11].

Equation (42) may now be ensemble averaged using the Feynman-diagrammatic technique introduced by Frisch to arrive at the *Dyson equation,*

$$\langle\mathbf{F}_{\rm exc}(\mathbf{r})\rangle=\mathbf{F}_{\rm Br}(\mathbf{r})+\mathcal{P}\int d^3r'd^3r''\mathcal{G}_{\rm Br}(\mathbf{r}-\mathbf{r}')\boldsymbol{\Sigma}(\mathbf{r}'-\mathbf{r}'')\times\langle\mathbf{F}_{\rm exc}(\mathbf{r}'')\rangle , \tag{46}$$

where the quantity $\boldsymbol{\Sigma}(\mathbf{r}'-\mathbf{r}'')$ is called the *mass operator,* a name coming from quantum field theory. The mass operator consists of an infinite series, each term of which contains products over $\mathcal{P}\mathcal{G}_{\rm Br}(\mathbf{r}-\mathbf{r}')$ and the statistical cumulants of $\mathcal{X}(\mathbf{r})$ [13].

For pragmatic reasons, approximations to the Dyson equation are unavoidable. They are usually implemented by truncating the series expansion of the mass operator $\boldsymbol{\Sigma}$. To the lowest (i.e., second) order in $\mathcal{X}$ we have

$$\boldsymbol{\Sigma}(\mathbf{r}-\mathbf{r}')=\langle\mathcal{X}(\mathbf{r})\mathcal{P}\mathcal{G}_{\rm Br}(\mathbf{r}-\mathbf{r}')\mathcal{X}(\mathbf{r}')\rangle , \tag{47}$$

which is called the *bilocal approximation*. Now,

$$\mathcal{X}(\mathbf{r})=\mathcal{X}_a\theta_a(\mathbf{r})+\mathcal{X}_b\theta_b(\mathbf{r}) \tag{48}$$

for all $\mathbf{r}$. Inserting this expression into Eq. (47) and using the condition (44), we obtain

$$\mathbf{\Sigma}(\mathbf{R})=(\mathcal{X}_a-\mathcal{X}_b)\mathcal{P}\mathcal{G}_{\mathrm{Br}}(\mathbf{R})\tau_a(R)(\mathcal{X}_a-\mathcal{X}_b) \tag{49}$$

after some algebraic manipulations, with the covariance $\tau_a(R)$ having been introduced in Eq. (18).

VI. EQUATIONS FOR THE NONLOCAL EFFECTIVE MEDIUM

In order to complete the SPFT formulation, we go on to determine the relation between the ensemble-averaged fields $\langle\mathbf{C}(\mathbf{r})\rangle$ and $\langle\mathbf{F}(\mathbf{r})\rangle$. The ensemble average of the constitutive relation (8) may be stated as

$$\langle\mathbf{C}(\mathbf{r})\rangle=\mathcal{J}\langle\mathcal{K}(\mathbf{r})\mathbf{F}(\mathbf{r})\rangle \ . \tag{50}$$

Because we are examining *linear* matter, there must exist a linear relation between $\langle\mathcal{K}(\mathbf{r})\mathbf{F}(\mathbf{r})\rangle$ and $\langle\mathbf{F}(\mathbf{r})\rangle$. Furthermore, this relation has to be of the form

$$\langle\mathcal{K}(\mathbf{r})\mathbf{F}(\mathbf{r})\rangle=\int d^3R\,\mathcal{K}_{\mathrm{Dy}}(\mathbf{R})\langle\mathbf{F}(\mathbf{r}-\mathbf{R})\rangle \tag{51}$$

due to translational invariance. The matrix $\mathcal{K}_{\mathrm{Dy}}(\mathbf{R})$ contains the constitutive properties of the effective medium consistent with the SPFT. In general, this matrix is *nonlocal* and, therefore, signifies spatial dispersion. All we have to do now is to find the connection between the quantities discussed in the preceding section and $\mathcal{K}_{\mathrm{Dy}}(\mathbf{R})$.

Let us note from Eqs. (41) and (43) that

$$\mathbf{F}_{\mathrm{exc}}(\mathbf{r})=\tfrac{2}{3}\mathbf{F}(\mathbf{r})+\tfrac{1}{3}\mathcal{K}_{\mathrm{Br}}^{-1}\mathcal{K}(\mathbf{r})\mathbf{F}(\mathbf{r}) \ , \tag{52}$$

$$\mathcal{X}(\mathbf{r})\mathbf{F}_{\mathrm{exc}}(\mathbf{r})=\mathcal{K}(\mathbf{r})\mathbf{F}(\mathbf{r})-\mathcal{K}_{\mathrm{Br}}\mathbf{F}(\mathbf{r}) \ , \tag{53}$$

their respective ensemble-averaged counterparts being

$$\langle\mathbf{F}_{\mathrm{exc}}(\mathbf{r})\rangle=\tfrac{2}{3}\langle\mathbf{F}(\mathbf{r})\rangle+\tfrac{1}{3}\mathcal{K}_{\mathrm{Br}}^{-1}\langle\mathcal{K}(\mathbf{r})\mathbf{F}(\mathbf{r})\rangle \ , \tag{54}$$

$$\langle\mathcal{X}(\mathbf{r})\mathbf{F}_{\mathrm{exc}}(\mathbf{r})\rangle=\langle\mathcal{K}(\mathbf{r})\mathbf{F}(\mathbf{r})\rangle-\mathcal{K}_{\mathrm{Br}}\langle\mathbf{F}(\mathbf{r})\rangle \ . \tag{55}$$

Furthermore, on taking the ensemble average of Eq. (42) and comparing it with the Dyson equation (46), we get

$$\langle\mathcal{X}(\mathbf{r})\mathbf{F}_{\mathrm{exc}}(\mathbf{r})\rangle=\int d^3r'\mathbf{\Sigma}(\mathbf{r}-\mathbf{r}')\langle\mathbf{F}_{\mathrm{exc}}(\mathbf{r}')\rangle \ . \tag{56}$$

Finally, after rearranging Eqs. (54)–(56) and inserting the constitutive relations (50), we obtain

$$\mathcal{J}^{-1}\langle\mathbf{C}(\mathbf{r})\rangle-\tfrac{1}{3}\int d^3r'\mathbf{\Sigma}(\mathbf{r}-\mathbf{r}')\mathcal{K}_{\mathrm{Br}}^{-1}\mathcal{J}^{-1}\langle\mathbf{C}(\mathbf{r}')\rangle$$
$$=\mathcal{K}_{\mathrm{Br}}\langle\mathbf{F}(\mathbf{r})\rangle+\tfrac{2}{3}\int d^3r'\mathbf{\Sigma}(\mathbf{r}-\mathbf{r}')\langle\mathbf{F}(\mathbf{r}')\rangle \ . \tag{57}$$

This integral equation gives a linear relation between $\langle\mathbf{C}(\mathbf{r})\rangle$ and $\langle\mathbf{F}(\mathbf{r})\rangle$. After solving this integral equation for $\langle\mathbf{C}(\mathbf{r})\rangle$, we shall be able to obtain the desired effective medium matrix $\mathcal{K}_{\mathrm{Dy}}(\mathbf{R})$.

Since the integral equation (57) is of the convolution type, it can be solved by the Fourier transform technique. Therefore, we define the following quantities:

$$\hat{\mathbf{F}}(\mathbf{q})=\int d^3r\langle\mathbf{F}(\mathbf{r})\rangle e^{-i\mathbf{q}\cdot\mathbf{r}} \ , \tag{58}$$

$$\hat{\mathbf{C}}(\mathbf{q})=\int d^3r\langle\mathbf{C}(\mathbf{r})\rangle e^{-i\mathbf{q}\cdot\mathbf{r}} \ , \tag{59}$$

$$\hat{\mathbf{\Sigma}}(\mathbf{q})=\int d^3r\,\mathbf{\Sigma}(\mathbf{r})e^{-i\mathbf{q}\cdot\mathbf{r}} \ , \tag{60}$$

$$\hat{\mathcal{K}}_{\mathrm{Dy}}(\mathbf{q})=\int d^3r\,\mathcal{K}_{\mathrm{Dy}}(\mathbf{r})e^{-i\mathbf{q}\cdot\mathbf{r}} \ , \tag{61}$$

$\mathbf{q}$ being the three-dimensional spatial frequency vector. The Fourier-transformed version of Eq. (57) reads as

$$[\mathcal{K}_{\mathrm{Br}}-\tfrac{1}{3}\hat{\mathbf{\Sigma}}(\mathbf{q})]\mathcal{K}_{\mathrm{Br}}^{-1}\mathcal{J}^{-1}\hat{\mathbf{C}}(\mathbf{q})=[\mathcal{K}_{\mathrm{Br}}+\tfrac{2}{3}\hat{\mathbf{\Sigma}}(\mathbf{q})]\hat{\mathbf{F}}(\mathbf{q}) \ . \tag{62}$$

Solving this equation for $\hat{\mathbf{C}}(\mathbf{q})$ gives

$$\begin{aligned}\hat{\mathbf{C}}(\mathbf{q})&=\mathcal{J}\mathcal{K}_{\mathrm{Br}}[\mathcal{K}_{\mathrm{Br}}-\tfrac{1}{3}\hat{\mathbf{\Sigma}}(\mathbf{q})]^{-1}[\mathcal{K}_{\mathrm{Br}}+\tfrac{2}{3}\hat{\mathbf{\Sigma}}(\mathbf{q})]\hat{\mathbf{F}}(\mathbf{q})\\&=\mathcal{J}\mathcal{K}_{\mathrm{Br}}[\mathcal{K}_{\mathrm{Br}}-\tfrac{1}{3}\hat{\mathbf{\Sigma}}(\mathbf{q})]^{-1}[\mathcal{K}_{\mathrm{Br}}-\tfrac{1}{3}\hat{\mathbf{\Sigma}}(\mathbf{q})+\hat{\mathbf{\Sigma}}(\mathbf{q})]\hat{\mathbf{F}}(\mathbf{q})\\&=\mathcal{J}\{\mathcal{K}_{\mathrm{Br}}+\mathcal{K}_{\mathrm{Br}}[\mathcal{K}_{\mathrm{Br}}-\tfrac{1}{3}\hat{\mathbf{\Sigma}}(\mathbf{q})]^{-1}\hat{\mathbf{\Sigma}}(\mathbf{q})\}\hat{\mathbf{F}}(\mathbf{q}) \ .\end{aligned} \tag{63}$$

This can be rewritten as

$$\mathbf{C}(\mathbf{q})=\mathcal{J}\hat{\mathcal{K}}_{\mathrm{Dy}}(\mathbf{q})\hat{\mathbf{F}}(\mathbf{q}) \ , \tag{64}$$

where

$$\hat{\mathcal{K}}_{\mathrm{Dy}}(\mathbf{q})=\mathcal{K}_{\mathrm{Br}}+\mathcal{K}_{\mathrm{Br}}[\mathcal{K}_{\mathrm{Br}}-\tfrac{1}{3}\hat{\mathbf{\Sigma}}(\mathbf{q})]^{-1}\hat{\mathbf{\Sigma}}(\mathbf{q}) \ . \tag{65}$$

The effective medium matrix $\mathcal{K}_{\mathrm{Dy}}(\mathbf{r})$ is then obtainable as an integral, i.e.,

$$\mathcal{K}_{\mathrm{Dy}}(\mathbf{r})=\frac{1}{(2\pi)^3}\int d^3q\,\hat{\mathcal{K}}_{\mathrm{Dy}}(\mathbf{q})e^{i\mathbf{q}\cdot\mathbf{r}} \ , \tag{66}$$

The Dyson equation (46) involves the ensemble-averaged exciting field $\langle\mathbf{F}_{\mathrm{exc}}(\mathbf{r})\rangle$. In order to determine the ensemble-averaged electromagnetic field $\langle\mathbf{F}(\mathbf{r})\rangle$ itself, we take the ensemble average of Eq. (5) and use Eq. (51) to get

$$\nabla\times\langle\mathbf{F}(\mathbf{r})\rangle-\int d^3R\,\mathcal{K}_{\mathrm{Dy}}(\mathbf{R})\langle\mathbf{F}(\mathbf{r}-\mathbf{R})\rangle=0 \ . \tag{67}$$

The Fourier-transformed version of this equation is

$$i\mathbf{q}\times\hat{\mathbf{F}}(\mathbf{q})-\hat{\mathcal{K}}_{\mathrm{Dy}}(\mathbf{q})\hat{\mathbf{F}}(\mathbf{q})=0 \ , \tag{68}$$

which can be solved with standard dyadic techniques [4].

VII. LONG-WAVELENGTH APPROXIMATION

In principle, the electromagnetic field equation (67) gives an exact description of the electromagnetic properties of the mixture. As mentioned in Sec. III, disorder usually introduces an intrinsic length scale, viz., the correlation length L. Let us recall that there are four basic wave numbers in a two-component chiral mixtures, two in each component. Hence, we can think of four wavelengths. When the maximum of these four wavelengths is smaller than or comparable with L in magnitude—i.e., the scale is *microscopic*—the details of the particulate geometry are resolvable and make the effective medium nonlocal.

However, when the minimum of the four wavelengths is much larger than L, we can achieve a *macroscopic* description of the mixture. Although it has a different provenance, this description is conceptually no different from that available from the Maxwell-Garnett and the Bruggeman mixing rules: the mixture is considered

homogeneous in the so-called *long-wavelength approximation* [9].

Suppose the long-wavelength approximation is appropriate. Let us then introduce the macroscopic fields $\mathbf{C}_{\text{macro}}(\mathbf{r})$ and $\mathbf{F}_{\text{macro}}(\mathbf{r})$ by *spatially* averaging the microscopic fields $\langle \mathbf{C}(\mathbf{r}) \rangle$ and $\langle \mathbf{F}(\mathbf{r}) \rangle$ over a volume V; thus,

$$\mathbf{F}_{\text{macro}}(\mathbf{r}) = \frac{1}{V} \int_V d^3r'' \langle \mathbf{F}(\mathbf{r}+\mathbf{r}'') \rangle \ , \tag{69}$$

$$\mathbf{C}_{\text{macro}}(\mathbf{r}) = \frac{1}{V} \int_v d^3r'' \langle \mathbf{C}(\mathbf{r}+r'') \rangle \ . \tag{70}$$

The minimum linear cross-sectional extent of the volume V must be much larger than L, but much smaller than the minimum of the four wavelengths in the two components. Inserting Eqs. (50) and (51) into (70), we find

$$\begin{aligned} \mathbf{C}_{\text{macro}}(\mathbf{r}) &= \mathscr{A} \int d^3R \, \mathcal{H}_{\text{Dy}}(\mathbf{R}) \frac{1}{V} \int_V d^3r'' \langle \mathbf{F}(\mathbf{r}+\mathbf{r}''-\mathbf{R}) \rangle \\ &= \mathscr{A} \int d^3R \, \mathcal{H}_{\text{Dy}}(\mathbf{R}) \mathbf{F}_{\text{macro}}(\mathbf{r}+\mathbf{R}) \\ &\approx \mathscr{A} \int d^3R \, \mathcal{H}_{\text{Dy}}(\mathbf{R}) \cdot \mathbf{F}_{\text{macro}}(\mathbf{r}) \ . \end{aligned} \tag{71}$$

This leads to the macroscopic constitutive relation

$$\mathbf{C}_{\text{macro}}(\mathbf{r}) = \mathscr{A} \hat{\mathcal{H}}_{\text{Dy}}(\mathbf{q}=\mathbf{0}) \mathbf{F}_{\text{macro}}(\mathbf{r}) \ , \tag{72}$$

where $\mathbf{0}$ is the null vector. In deriving this estimate of the effective medium properties, we used the fact that macroscopic fields can be regarded as uniform on microscopic length scales. In a long-wavelength description, the mixture is treated as homogeneous; so we obtained a *local* effective medium.

We must pause here to note that $\hat{\mathcal{H}}_{\text{Dy}}(\mathbf{q}=\mathbf{0}) \neq \mathcal{H}_{\text{Br}}$. Hence, the estimate of the effective medium properties obtained in this section differs from the Bruggeman estimate. The matrix $\hat{\mathcal{H}}_{\text{Dy}}(\mathbf{q}=\mathbf{0})$ contains, via its relation to the mass operator (65), information about spatial correlations in the two-component mixture. It therefore gives a better description of the two-component mixture than the Bruggeman rule, which only contains the filling factors [Eq. (45)].

VIII. BILOCAL APPROXIMATION

We now go on to determine the effective medium properties using the bilocal approximation. For this purpose, we have to calculate the Fourier transform (60) of the mass operator given in equation (49). Therefore, we commence by writing

$$\hat{\mathbf{\Sigma}}(\mathbf{q}) = (\mathcal{X}_a - \mathcal{X}_b) \hat{\mathcal{S}}(\mathbf{q}) (\mathcal{X}_a - \mathcal{X}_b) \ , \tag{73}$$

where

$$\hat{\mathcal{S}}(\mathbf{q}) = \int d^3R \, \mathcal{P} \mathcal{G}_{\text{Br}}(\mathbf{R}) \tau_a(R) e^{-i\mathbf{q}\cdot\mathbf{R}} \ . \tag{74}$$

Inserting the matrix elements of $\mathcal{G}_{\text{Br}}$ as given in Eqs. (33) *et seq.*, we get

$$\hat{\mathcal{S}}(\mathbf{q}) = \begin{Bmatrix} \frac{1}{2}[\hat{\mathcal{S}}^+(\gamma_{\text{Br}}^+|\mathbf{q}) + \hat{\mathcal{S}}^-(\gamma_{\text{Br}}^-|\mathbf{q})] & \dfrac{i\eta_{\text{Br}}}{2}[\hat{\mathcal{S}}^+(\gamma_{\text{Br}}^+|\mathbf{q}) - \hat{\mathcal{S}}^-(\gamma_{\text{Br}}^-|\mathbf{q})] \\ \dfrac{1}{2i\eta_{\text{Br}}}[\hat{\mathcal{S}}^+(\gamma_{\text{Br}}^+|\mathbf{q}) - \hat{\mathcal{S}}^-(\gamma_{\text{Br}}^-|\mathbf{q})] & \frac{1}{2}[\hat{\mathcal{S}}^+(\gamma_{\text{Br}}^+|\mathbf{q}) + \hat{\mathcal{S}}^-(\gamma_{\text{Br}}^-|\mathbf{q})] \end{Bmatrix} , \tag{75}$$

with

$$\hat{\mathcal{S}}^{\pm}(\gamma_{\text{Br}}^{\pm}|\mathbf{q}) = \int d^3R \, \mathcal{P} \mathcal{G}^{\pm}(\gamma_{\text{Br}}^{\pm}|\mathbf{R}) \tau_a(R) e^{-i\mathbf{q}\cdot\mathbf{R}} \ . \tag{76}$$

Let us recall the coordinate-free form of the Beltrami-Green dyadics as

$$\begin{aligned} G^{\pm}(\sigma|\mathbf{R}) = \pm i(\sigma^2/4\pi) \{ &\tfrac{2}{3} h_0^{(1)}(\sigma R) \mathcal{T}_0(\mathbf{u}_{\mathbf{R}}) \\ &\mp h_1^{(1)}(\sigma R) \mathcal{T}_1(\mathbf{u}_{\mathbf{R}}) \\ &+ h_2^{(1)}(\sigma R) \mathcal{T}_2(\mathbf{u}_{\mathbf{R}}) \} \ , \end{aligned} \tag{77}$$

where we introduce the spherical Hankel functions

$$h_0^{(1)}(z) = -iz^{-1}e^{iz} \ , \quad h_1^{(1)}(z) = -(iz^{-2}+z^{-1})e^{iz} \ ,$$
$$h_2^{(1)}(z) = i(-3z^{-3}+3iz^{-2}+z^{-1})e^{iz}$$

and the irreducible dyadics

$$\mathcal{T}_0(\mathbf{u}_{\mathbf{R}}) = \mathbf{I} \ , \quad \mathcal{T}_1(\mathbf{u}_{\mathbf{R}}) = \mathbf{u}_{\mathbf{R}} \times \mathbf{I} , \quad \mathcal{T}_2(\mathbf{u}_{\mathbf{R}}) = \mathbf{u}_{\mathbf{R}}\mathbf{u}_{\mathbf{R}} - \tfrac{1}{3}\mathbf{I} \ .$$

Because of their irreducibility, these tensors can be written in terms of spherical harmonic functions $Y_{lm}(\mathbf{u}_{\mathbf{R}})$ as

$$\mathcal{T}_l(\mathbf{u}_{\mathbf{R}}) = \sum_{m=-l}^{l} \mathcal{T}_{lm} Y_{lm}(\mathbf{u}_{\mathbf{R}}) \ , \tag{78}$$

with the dyadics $\mathcal{T}_{lm}$ independent of $\mathbf{u}_{\mathbf{R}}$. The partial wave decomposition of the plane wave $e^{-i\mathbf{q}\cdot\mathbf{R}}$ is [23]

$$e^{-i\mathbf{q}\cdot\mathbf{R}} = 4\pi \sum_{l=0}^{\infty} (-i)^l j_l(qR) \sum_{m=-l}^{l} Y_{lm}^*(\mathbf{u}_{\mathbf{R}}) Y_{lm}(\mathbf{u}_{\mathbf{q}}) \ , \tag{79}$$

with the spherical Bessel functions $j_l(qR)$, $\mathbf{u}_{\mathbf{q}} = \mathbf{q}/q$, and the asterisk denoting complex conjugation.

We insert Eqs. (77)–(79) into (76) and use the orthonormality properties of $Y_{lm}(\mathbf{u}_{\mathbf{R}})$ on the unit sphere to obtain

$$\begin{aligned} \hat{\mathcal{S}}^{\pm}(\gamma_{\text{Br}}^{\pm}|\mathbf{q}) = \pm(\gamma_{\text{Br}}^{\pm})^2 \{ &\tfrac{2}{3} i s_0(\gamma_{\text{Br}}^{\pm}|q) \mathcal{T}_0(\mathbf{u}_{\mathbf{q}}) \\ &\mp s_1(\gamma_{\text{Br}}^{\pm}|q) \mathcal{T}_1(\mathbf{u}_{\mathbf{q}}) \\ &- i s_2(\gamma_{\text{Br}}^{\pm}|q) \mathcal{T}_2(\mathbf{u}_{\mathbf{q}}) \} \ , \end{aligned} \tag{80}$$

where the integrals

$$s_l(\gamma_{\text{Br}}^{\pm}|q) = \int_0^{\infty} dR \, R^2 h_l^{(1)}(\gamma_{\text{Br}}^{\pm} R) j_l(qR) \tau_a(R) \tag{81}$$

have to be evaluated numerically. We plan to discuss nu-

merical results in a future paper.

Let us return to the long-wavelength approximation discussed in the preceding section; i.e., $q=0$ and $\gamma_{\rm Br}^{\pm}L \ll 1$. Correct to the lowest order in $\gamma_{\rm Br}^{+}L$, we get

$$s_0(\gamma_{\rm Br}^{\pm}|0) \approx -\frac{i}{\gamma_{\rm Br}^{\pm}} \int_0^{\infty} dR\ R\tau_a(R)\ , \tag{82}$$

$$s_1(\gamma_{\rm Br}^{\pm}|0)=s_2(\gamma_{\rm Br}^{\pm}|0)=0\ . \tag{83}$$

Thus, the only surviving integral does not depend on the constitutive parameters of the two chiral components, but only on the mixture's microscopic structure. Using the foregoing expressions, we obtain the simple result

$$\hat{\mathcal{S}}^{\pm}(\gamma_{\rm Br}^{\pm}|\mathbf{q}=\mathbf{0}) \approx \pm\frac{2\gamma_{\rm Br}^{\pm}}{3} \int_0^{\infty} dR\ R\tau_a(R)\mathbf{I}\ . \tag{84}$$

Inserting into the Eq. (75) and using the definitions from Sec. III we obtain

$$\hat{\mathcal{S}}(\mathbf{q}=\mathbf{0}) \approx \tfrac{2}{3}\mathcal{H}_{\rm Br} \int_0^{\infty} dR\ R\tau_a(R)\ . \tag{85}$$

Finally, we get the following result for the mass operator:

$$\hat{\Sigma}(\mathbf{q}=\mathbf{0}) = U(\mathcal{X}_a-\mathcal{X}_b)\mathcal{H}_{\rm Br}(\mathcal{X}_a-\mathcal{X}_b)\ , \tag{86}$$

where

$$U=\tfrac{2}{3}\int_0^{\infty} dR\ R\tau_a(R)\ . \tag{87}$$

Hence, the macroscopic constitutive relation in Sec. VII can now be ascertained in terms of the spatial correlations in the two-component mixtures.

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[1] *Chirality—From Weak Bosons to the α-Helix* edited by R. Janoschek (Springer, Berlin, 1991).
[2] *Selected Papers on Natural Optical Activity,* edited by A. Lakhtakia (SPIE Optical Engineering, Bellingham, WA, 1991).
[3] *Proceedings of CHIRAL'94: 3rd International Workshop on Chiral, Bi-isotropic an Bi-anisotropic Media, edited by F. Marriotte and J. P. Parniex* (French Atomic Energy Commission, Le Barp, France, 1994).
[4] A. Lakhtakia, *Beltrami Fields in Chiral Media* (World Scientific, Singapore, 1994).
[5] C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles* (Wiley, New York, 1983).
[6] A. Lakhtakia, Int. J. Electron. **75**, 1243 (1993).
[7] M. E. Edwards, Y. H. Hwang, and X.-L. Wu, *Mossotti equation in a model microemission,* Phys. Rev. E **49**, 4263 (1994). The Maxwell-Garnett rule is identical to the so-called Lorentz-Lorentz and the Mossotti-Clausius rules.
[8] A. Lakhtakia, V. K. Varadan and V. V. Varadan, J. Mater. Res. **8**, 917 (1993).
[9] B. Shanker and A. Lakhtakia, J. Phys. D **26**, 1746 (1993).
[10] A. Sihvola, S. Saastamoinen, and K. Heiska, Remote Sensing Rev. **9**, 39 (1993).
[11] R. D. Kampia and A. Lakhtakia, J. Phys. D **25**, 1390 (1992).
[12] C. F. Bohren, J. Atmos. Sci. **43**, 468 (1986).
[13] U. Frisch, in *Probabilistic Methods in Applied Mathematics,* edited by A. T. Bharucha-Reid (Academic, London, 1970), Vol. 1.
[14] V. V. Tamoikin, Radiophys. Quantum Electron. **14**, 228 (1971).
[15] L. Tsang and J. A. Kong, Radio Sci. **16**, 303 (1981).
[16] A. Lakhtakia, Int. J. Infrared Millim. Waves **13**, 551 (1992).
[17] A. Lakhtakia and W. S. Weiglhofer, IEEE Trans. Microwave Theory Tech. **42**, 1715 (1994).
[18] A. Stogryn, IEEE Trans. Antennas Propag. **31**, 985 (1983).
[19] C. F. Bohren, Chem. Phys. Lett. **29**, 458 (1974).
[20] W. S. Weiglhofer, Radio Sci. **28**, 847 (1993).
[21] W. S. Weiglhofer, Am. J. Phys. **57**, 455 (1989).
[22] L. Tsang, J. A. Kong, and R. T. Shin, *Theory of Microwave Remote Sensing* (Wiley, New York, 1985).
[23] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1975).