ABSTRACT: Two interpretations of the Bruggeman approach for the homogenization of isotropic chiral mixtures are shown to lead to different results. Whereas the standard interpretation is shown to yield the average polarizability density approach, a recent interpretation turns out to deliver a null excess polarization approach. The difference between the two interpretations arises from differing treatments of the local field.

Key Words: Bruggeman approach; chiral materials; composite materials; excess polarization; homogenization; local field; polarizability density

1 Introduction

Homogenization of particulate composite materials is at least a two–century–old theoretical problem; yet, it retains its freshness to this day. Indeed, it can be argued that, as theoretical approaches can — at best — only estimate the effective constitutive parameters of a mixture of two or more component materials but still viewed as being homogeneous, homogenization is unlikely to lose its charm for theorists for the foreseeable future Mbook,Mackay.

This viewpoint rose to the fore recently when we had occasion to look at the Bruggeman approach for the homogenization of an isotropic mixture of two isotropic chiral materials. In this approach, the volume fractions of both component materials are taken into account, but the particulate dimensions are effectively null–valued, as explicated by Kampia and Lakhtakia KL. In the extended Brugge-
man approach, the particulate dimensions are considered as electrically small but finite, as exemplified by Shanker Shan for the chosen mixture. However, we found that Shanker’s interpretation of the Bruggeman approach differs in an essential point from that of Kampaia and Lakhtakia — in addition to the differing treatments of the particulate dimensions. Our ruminations on the newly discovered difference led to this communication.

2 Theory in Brief

Let us consider an isotropic mixture of two isotropic chiral materials labeled $a$ and $b$. Their frequency–domain constitutive relations are stated as

$$
\begin{align*}
D &= \epsilon_0 \epsilon_p [E + \beta_p \nabla \times E], \\
B &= \mu_0 \mu_p [H + \beta_p \nabla \times H],
\end{align*}
$$

where $\epsilon_0$ and $\mu_0$ are the permittivity and the permeability of free space (i.e., vacuum); $\epsilon_{a,b}$ are the relative permittivity scalars, $\mu_{a,b}$ are the relative permeability scalars, and $\beta_{a,b}$ are the chirality pseudoscalars in the Drude–Born–Fedorov representation; and an $\exp(-j\omega t)$ time–dependence is implicit. The volumetric fractions of the two component materials are denoted by $f_a$ and $f_b = 1 - f_a$. The aim of any homogenization exercise is to predict the quantities $\epsilon_{HCM}$, $\mu_{HCM}$ and $\beta_{HCM}$ appearing in the constitutive relations

$$
\begin{align*}
D &= \epsilon_0 \epsilon_{HCM} [E + \beta_{HCM} \nabla \times E], \\
B &= \mu_0 \mu_{HCM} [H + \beta_{HCM} \nabla \times H]
\end{align*}
$$

that presumably hold for the homogenized composite material (HCM). The exercise is well–founded only if the particles of both component materials can be considered to be electrically small.

The Bruggeman approach for homogenization was initiated for isotropic mixtures of isotropic dielectric materials, but has been subsequently extended to far more complex situations. The general formulation of the approach is as follows: Suppose the composite material has been homogenized, and it obeys $\epsilon_0$. Disperse in it, homogeneously and randomly, a small number density of particles of both types of component materials in the volumetric ratio $f_a : f_b$; and then homogenize. The properties of the HCM could not have altered in consequence.

All particles of type $p$, $(p = a, b)$, are identical, and are equivalent to electric and magnetic dipole moments, $p_p$ and $m_p$, when immersed in the HCM. The
standard interpretation of the Bruggeman approach then requires the solution of the following two equations WLM97:

\[
\begin{align*}
fa \mathbf{p}_a + fb \mathbf{p}_b &= 0 \\
f_a \mathbf{m}_a + f_b \mathbf{m}_b &= 0
\end{align*}
\]

(3)

In the present context, Kampia and Lakhtakia KL solved eq3 for $$\epsilon_{HCM}$$, $$\mu_{HCM}$$ and $$\beta_{HCM}$$.

An alternative interpretation is that the dispersal of particles of component material $$p$$ is equivalent to the creation of excess polarization and excess magnetization, $$\mathbf{P}_p$$ and $$\mathbf{M}_p$$, ($$p = a, b$$), in the HCM. But the total excess polarization and magnetization must be null–valued. Then, the two equations

\[
\begin{align*}
\mathbf{P}_a + \mathbf{P}_b &= 0 \\
\mathbf{M}_a + \mathbf{M}_b &= 0
\end{align*}
\]

(4)

could be solved to determine $$\epsilon_{HCM}$$, $$\mu_{HCM}$$ and $$\beta_{HCM}$$. Although eq4 were stated by Kampia and Lakhtakia KL, these equations were not solved by them; indeed, expressions for $$\mathbf{P}_p$$ and $$\mathbf{M}_p$$ were not even provided by them. However, Shanker Shan did present expressions for $$\mathbf{P}_p$$ and $$\mathbf{M}_p$$, and then solved eq4.

3 Numerical Results

We decided to compare the implementations of eq3 and eq4. All particles of both component materials were treated as spheres of radius $$R$$. Expressions for the polarizability densities (relating electric and magnetic dipole moments to exciting electric and magnetic fields) and polarization densities (relating excess polarization and excess magnetization to electric and magnetic fields) were obtained from Shanker’s paper Shan.

Computed values of $$\epsilon_{HCM}$$, $$\mu_{HCM}$$ and $$\beta_{HCM}$$ as functions of $$f_b$$ are shown in Figures 1 and 2 for $$k_0R \to 0$$ and $$k_0R = 0.2$$, where $$k_0 = \omega(\epsilon_0\mu_0)^{1/2}$$ is the free–space wavenumber. The constitutive properties of the component materials for the two figures are the same as chosen by Shanker Shan.

Quite clearly, Figures 1 and 2 show that the incorporation of the finite size of the particles gives rise to a dissipative HCM, even when both component materials are nondissipative. This conclusion is true whenever a nonzero length–scale is considered in a homogenization approach — whether as the particle size Shan,D89,
or a correlation length for particle–distribution statistics Mačkay, or both M04. The incorporation of the length scale appears to account, in some manner, for the scattering loss.

More importantly, whether the length scale is neglected (Fig. 1) or considered (Fig. 2), estimates of $\epsilon_{HCM}$, $\mu_{HCM}$ and $\beta_{HCM}$ from $\text{eq}3$ and $\text{eq}4$ do not coincide. There seems to be a basic difference between $\text{eq}3$ and $\text{eq}4$, which persists even when $\mu_{a,b} = 1$, $\beta_{a,b} = 0$ and $R \to 0$. An explanation of this difference, in that simple context for the sake of clarity, is provided in the next section.

4 Explanation

4.1 Preliminaries

We begin with the derivation of an important equation. Let all space be occupied by a homogeneous dielectric material with relative permittivity $\epsilon_h$ at the frequency of interest; thus, its relevant frequency–domain constitutive relation is

$$D = \epsilon_0 \epsilon_h E.$$  \hfill (5)

Suppose that an electrically small sphere made of a dielectric material with relative permittivity $\epsilon_i$ were to be introduced. This particle would act as an electric dipole moment

$$p = \frac{v}{\epsilon_0} \alpha_{i/h} \hat{e},$$  \hfill (6)

where $v$ is the volume of the particle, $\hat{e}$ is the electric field at the location of the particle if the particle were to be removed and the resulting hole filled with the host material, and the product of $\epsilon_0$ and $\alpha_{i/h}$ is the polarizability density of the particle embedded in the specific host material. The exact expression of $\alpha_{i/h}$ does not matter for our purpose here Lak92; but we note that it is independent of $R$ for the Bruggeman approach, and dependent on $R$ for the extended Bruggeman approach PLS.

Let many identical particles be randomly dispersed in the host material, such that their number density $N$ is macroscopically uniform. Then, the particles can be replaced by an excess polarization

$$P = Nv\epsilon_0 \alpha_{i/h} \hat{E},$$  \hfill (7)
where

\[ \tilde{E} = E + \frac{P}{3\varepsilon_0\varepsilon_h} \tag{8} \]

is the local electric field \( \text{Lak92} \). The qualifier \textit{excess} is used here because this \( P \) is in addition to the polarization \( \varepsilon_0(\varepsilon_h - 1)E \) that indicates the presence of the host material.

By virtue of \text{eqq4} and \text{eqq5}, the excess polarization

\[ P = \varepsilon_0 \left( \frac{f_{\alpha_i/h}}{1 - f_{\alpha_i/h}/3\varepsilon_h} \right) E, \tag{9} \]

where \( f = Nv \) is the volumetric fraction of the particulate material. Hence, the constitutive relation of the HCM is

\[
\begin{align*}
\mathbf{D} &= \varepsilon_0\varepsilon_h\mathbf{E} + \mathbf{P} \\
&= \varepsilon_0 \left[ \varepsilon_h + \frac{f_{\alpha_i/h}}{1 - f_{\alpha_i/h}/3\varepsilon_h} \right] \mathbf{E} \\
&= \varepsilon_0\varepsilon_{HCM}\mathbf{E}, \tag{10}
\end{align*}
\]

so that

\[ \varepsilon_{HCM} = \varepsilon_h + \frac{f_{\alpha_i/h}}{1 - f_{\alpha_i/h}/3\varepsilon_h} \tag{11} \]

is the estimated relative permittivity of the HCM at the frequency of interest. The first rigorous derivation of the foregoing equation can be attributed to Faxén \( \text{Fg} \).

Parenthetically, a Maxwell Garnett formula for \( \varepsilon_{HCM} \) can be derived by setting \( \varepsilon_h = \varepsilon_a \) and \( \varepsilon_i = \varepsilon_b \) in basic, which is quite appropriate if \( f_b < f_a \); otherwise, the choice \( \{\varepsilon_h = \varepsilon_b, \varepsilon_i = \varepsilon_a\} \) should be made. These two Maxwell Garnett estimates also constitute the so-called Hashin–Shtrikman bounds on \( \varepsilon_{HCM} \) \( \text{HS} \).

4.2 Standard Interpretation of the Bruggeman approach: Eq. \text{eq3}

As stated in Section 2, let us imagine that the composite material has already been homogenized. Into this HCM, let spherical particles of both component materials be randomly dispersed. The combined volumetric fraction of the particles introduced into the HCM is \( f \ll 1 \), with \( f_{fa} \) and \( f_{fb} \) being the respective volumetric fractions of the two component materials in the particles. Hence,

\[ \alpha_{i/h} = f_a \alpha_{a/HCM} + f_b \alpha_{b/HCM} \tag{12} \]
is the polarizability density of a \textit{material–averaged} particle embedded in a material with $\epsilon_h = \epsilon_{HCM}$. Equation $\hat{\text{exP}}$ then yields

$$P = \epsilon_0 \left[ \frac{f_a \alpha_a/HCM + f_b \alpha_b/HCM}{1 - f_a \alpha_a/HCM + f_b \alpha_b/HCM}/3\epsilon_{HCM} \right] E, \quad (13)$$

for the excess polarization.

But the introduction of the material–averaged particles must not change the HCM’s constitutive properties, as the relative proportion of the component materials remains unchanged; accordingly, the excess polarization of $\hat{\text{exPBr}}$ is null–valued, and the solution of the equation

$$0 = f_a \alpha_a/HCM + f_b \alpha_b/HCM \quad (14)$$
yields an estimate of $\epsilon_{HCM}$. Thus the standard interpretation of the Bruggeman approach leading to $\hat{\text{eq3}}$ is as the \textit{average polarizability density} approach.

### 4.3 Shanker’s Interpretation of the Bruggeman approach: Eq. $\hat{\text{eq4}}$

Once again, suppose that the composite material has been homogenized into a HCM with relative permittivity $\epsilon_{HCM}$. Suppose, next, that particles of materials $a$ and $b$ are randomly dispersed the HCM and that their respective volumetric fractions in the new composite material are $f_a$ and $f_b$. Following Shanker Shan, we find that the excess polarizations due to the two types of particles add up to

$$P = \epsilon_0 \left[ \frac{f_a \alpha_a/HCM}{1 - f_a \alpha_a/HCM}/3\epsilon_{HCM} + \frac{f_b \alpha_b/HCM}{1 - f_b \alpha_b/HCM}/3\epsilon_{HCM} \right] E, \quad (15)$$

by virtue of $\hat{\text{exP}}$.

The introduction of the particles into the HCM amounts simply to the complete replacement of the HCM by itself; hence, basic leads to

$$\epsilon_{HCM} = \epsilon_{HCM} + \frac{f_a \alpha_a/HCM}{1 - f_a \alpha_a/HCM}/3\epsilon_{HCM} + \frac{f_b \alpha_b/HCM}{1 - f_b \alpha_b/HCM}/3\epsilon_{HCM}, \quad (16)$$

which yields the formula

$$0 = \frac{f_a \alpha_a/HCM}{1 - f_a \alpha_a/HCM}/3\epsilon_{HCM} + \frac{f_b \alpha_b/HCM}{1 - f_b \alpha_b/HCM}/3\epsilon_{HCM} \quad (17)$$

for an estimate of $\epsilon_{HCM}$. Thus Shanker’s interpretation of the Bruggeman approach leading to $\hat{\text{eq4}}$ is as the \textit{null excess polarization} approach.
4.4 Comparison of the Two Interpretations

Equation \( \hat{\alpha}_{\text{NEP}} \) differs from \( \hat{\alpha}_{\text{exPBr}} \) in a very significant way: Whereas particles of the two component materials were amalgamated into material–averaged particles whose polarizability density was used to estimate the excess polarization as per \( \hat{\alpha}_{\text{exPBr}} \), material–averaging was not done for \( \hat{\alpha}_{\text{NEP}} \); instead, particles of both materials were kept apart and two separate contributions were made to the estimate \( \hat{\alpha}_{\text{NEP}} \) of the excess polarization.

This difference can be understood also in terms of the different treatments of the local field. For \( \hat{\alpha}_{\text{exPBr}} \), the local field pertains to material–averaged particles, which is quite reasonable. In contrast, \( \hat{\alpha}_{\text{NEP}} \) contains two different local fields. The first local field pertains only to particles of material \( a \) embedded in the HCM, and leads to the first term in the sum on the right side of \( \hat{\alpha}_{\text{NEP}} \); while the second local field pertains only to particles of material \( b \) embedded in the HCM, and leads to the second term in the sum on the right side of \( \hat{\alpha}_{\text{NEP}} \). Accordingly, \( \hat{\alpha}_{\text{NEP}} \) lacks rigor in comparison to \( \hat{\alpha}_{\text{exPBr}} \), and the former can be considered simply as an empirical formula.

In closing, if \( P_a \) and \( P_b \) could somehow be separately estimated in Shanker’s interpretation with the same local field, the two interpretations could very possibly yield identical estimates of the constitutive parameters of the HCM.

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Figure 1: Estimated values of $\epsilon_{HCM}$, $\mu_{HCM}$ and $\beta_{HCM}/\beta_a$ as functions of $f_b$, when $\epsilon_a = 1$, $\mu_a = 1$, $\beta_a = 10^{-3}$ m, $\epsilon_b = 2$, $\mu_b = 1.5$, $\beta_b = 2.37 \times 10^{-3}$ m, and $k_0 R \to 0$. Solid lines represent data computed using eq3, while dashed lines join datapoints obtained using eq4.
Figure 2: Same as Figure 1, but for $k_oR = 0.2$. 