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Gain and loss enhancement in active and passive particulate composite materials

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ABSTRACT
Two active dielectric materials may be blended together to realize a homogenized composite material (HCM) which exhibits more gain than either component material. Likewise, two dissipative dielectric materials may be blended together to realize an HCM which exhibits more loss than either component material. Sufficient conditions for such gain/loss enhancement were established using the Bruggeman homogenization formalism. Gain/loss enhancement arises when (i) the imaginary parts of the relative permittivities of both component materials are similar in magnitude and (ii) the real parts of the relative permittivities of both component materials are dissimilar in magnitude.

1. Introduction
Two (or more) particulate materials may be mixed together to realize a homogenized composite material (HCM), provided that the particles making up the component materials are much smaller than the wavelengths involved.\cite{1} To be of practical value, an HCM is generally required to exhibit a desirable blend of certain properties of its component materials. Metamaterials are HCMs whose performances exceed those of their component materials.\cite{2,3} Within the electromagnetic realm, many instances of such HCMs can be found. For examples: through the process of homogenization, the phenomenon of weak nonlinearity may be enhanced,\cite{4–6} and the group speed may be enhanced beyond the maximum group speed in the component materials \cite{7,8} or weakened below the minimum group speed in the component materials.\cite{9}

In this short article, the prospect of enhancing gain by means of homogenization is explored for HCMs arising from active component materials. The dual process of loss enhancement in HCMs arising from dissipative component materials is also considered. The well-established Bruggeman homogenization formalism \cite{10–12} is employed, all component materials being thereby treated on the same footing. Accordingly, this formalism is applicable for all values of the volume fractions of the component materials.

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2. Homogenization via the Bruggeman formalism

Let us consider a composite material comprising two distinct materials labeled ‘a’ and ‘b’ that are distributed randomly as electrically small spheres. Both component materials are isotropic dielectric materials with relative permittivities $\varepsilon_a = \varepsilon_{a}^r + i \varepsilon_{a}^i$ and $\varepsilon_b = \varepsilon_{b}^r + i \varepsilon_{b}^i$, respectively, wherein $\varepsilon_{a}^r, \varepsilon_{a}^i, \varepsilon_{b}^r, \varepsilon_{b}^i \in \mathbb{R}$ and $\varepsilon_a \neq \varepsilon_b$. Physical plausibility requires the imposition of the restriction $\varepsilon_{a}^r \varepsilon_{b}^r > 0$ on the Bruggeman formalism.[13]

The Bruggeman estimate $\varepsilon_{Br} = \varepsilon_{Br}^r + i \varepsilon_{Br}^i$ of the HCM relative permittivity is provided implicitly by the quadratic equation

$$2\varepsilon_{Br}^2 + \varepsilon_{Br} \left[ \varepsilon_a \left( 1 - 3f_a \right) + \varepsilon_b \left( 3f_a - 2 \right) \right] - \varepsilon_a \varepsilon_b = 0 ,$$

(1)

with $f_a$ being the volume fraction of component material ‘a’. The limiting conditions $\varepsilon_{Br} \to \varepsilon_b$ as $f_a \to 0$, and $\varepsilon_{Br} \to \varepsilon_a$ as $f_a \to 1$ allow the correct root to be extracted from Equation (1).

When both component materials are active (i.e. $\varepsilon_{a,b}^i < 0$)[14] the phenomenon of gain enhancement is signified by $\varepsilon_{Br}^i < \min \{ \varepsilon_{a,b}^i \}$. When both component materials are dissipative (i.e. $\varepsilon_{a,b}^i > 0$), the phenomenon of loss enhancement is signified by $\varepsilon_{Br}^i > \max \{ \varepsilon_{a,b}^i \}$.

To illustrate the phenomenon of gain enhancement, let us consider a specific example. Suppose that the component materials are active ones, specified by $\varepsilon_a = 2 - 0.05i$ and $\varepsilon_b = 5 - 0.04i$. The real and imaginary parts of the Bruggeman estimate of the HCM relative permittivity are plotted against volume fraction in Figure 1. Also plotted in this figure are two well-established bounds on the HCM relative permittivity, namely the Wiener bounds[15]

$$W_\alpha = f_a \varepsilon_a + f_b \varepsilon_b$$

$$W_\beta = \left( \frac{f_a}{\varepsilon_a} + \frac{f_b}{\varepsilon_b} \right)^{-1}$$

(2)

and the Hashin–Shtrikman bounds[16]

$$HS_\alpha = \varepsilon_b + \frac{3f_a \varepsilon_b (\varepsilon_a - \varepsilon_b)}{\varepsilon_a + 2 \varepsilon_b - f_a (\varepsilon_a - \varepsilon_b)}$$

$$HS_\beta = \varepsilon_a + \frac{3f_b \varepsilon_a (\varepsilon_b - \varepsilon_a)}{\varepsilon_b + 2 \varepsilon_a - f_b (\varepsilon_b - \varepsilon_a)}$$

(3)

Herein, $f_b = 1 - f_a$ is the volume fraction of component material ‘b’. Originally, the Wiener bounds and the Hashin–Shtrikman bounds were derived for HCMs characterized by wholly real-valued constitutive parameters, but generalizations to complex-valued constitutive parameters later emerged.[17]

The Hashin–Shtrikman bound $HS_\alpha$ is equivalent to the Maxwell Garnett estimate of the HCM relative permittivity, based on the homogenization of a random dispersal of spheres of component material ‘a’ embedded in the host component material ‘b’, valid for $f_a \lesssim 0.3$.[18] Similarly, $HS_\beta$ is equivalent to the Maxwell Garnett estimate of the HCM relative permittivity, based on the homogenization of a random dispersal of spheres of component material ‘b’ embedded in the host component material ‘a’, valid for $f_b \lesssim 0.3$. 

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Figure 1. The real and imaginary parts of the HCM relative permittivity $\varepsilon_{Br}$ estimated by the Bruggeman formalism (red, solid curves) plotted against volume fraction $f_a$, when $\varepsilon_a = 2 - 0.05i$ and $\varepsilon_b = 5 - 0.04i$. Also plotted are the Hashin–Shtrikman bounds: HS$_\alpha$ (thin, green, broken dashed curves) and HS$_\beta$ (thick, green, broken dashed curves); and the Wiener bounds: W$_\alpha$ (thin, blue, dashed curves) and W$_\beta$ (thick, blue, dashed curves).

The real part of $\varepsilon_{Br}$ is seen in Figure 1 to decrease uniformly from $\varepsilon_a^{\prime}$ to $\varepsilon_a^{\prime}$ as $f_a$ increases from 0 to 1. Furthermore, $\varepsilon_{Br}^{\prime}$ is tightly bounded by HS$_\alpha$ and HS$_\beta$, and less tightly bounded by W$_\alpha$ and W$_\beta$. The imaginary part of $\varepsilon_{Br}$ follows a more interesting trajectory as $f_a$ increases: $\varepsilon_{Br}^{\prime}$ decreases from $\varepsilon_b^{\prime}$ at $f_a = 0$, reaches a minimum value at $f_a \approx 0.8$, and then increases to reach $\varepsilon_{a}^{\prime}$ at $f_a = 1$. Thus, according to the Bruggeman formalism, gain enhancement arises in the vicinity of $f_a \approx 0.8$, with the minimum value of $\varepsilon_{Br}^{\prime}$ ($\approx -0.0515$) being approximately 3% smaller than $\min \{ \varepsilon_a^{\prime}, \varepsilon_b^{\prime} \}$. Furthermore, $\text{Im} \left( \text{HS}_\beta \right) < \min \{ \varepsilon_a^{\prime}, \varepsilon_b^{\prime} \}$ when $0.7 \leq f_a < 1$. Thus, gain enhancement is also predicted by the Maxwell Garnett formalism.

Loss enhancement mirrors gain enhancement. To support this assertion, let us consider the dissipative counterpart of the active HCM considered in Figure 1. In Figure 2, plots are
Figure 2. As Figure 1 except that both component materials are dissipative, having the relative permittivities $\varepsilon_a = 2 + 0.05i$ and $\varepsilon_a = 5 + 0.04i$.

presented which are equivalent to those presented in Figure 1 but now the component materials are dissipative ones, specified by $\varepsilon_a = 2 + 0.05i$ and $\varepsilon_b = 5 + 0.04i$. As in Figure 1, $\varepsilon_{Br}^i$ in Figure 2 decreases uniformly from $\varepsilon_b^i$ to $\varepsilon_a^i$ as $f_a$ increases from 0 to 1; moreover, $\varepsilon_{Br}^i$ is tightly bounded by $H_{\alpha}$ and $H_{\beta}$, and less tightly bounded by $W_{\alpha}$ and $W_{\beta}$. The plot of $\varepsilon_{Br}^i$ in Figure 2 displays loss enhancement with the maximum value of $\varepsilon_{Br}^i$ ($\approx 0.0515$) being approximately 3% larger than $\max\{\varepsilon_a^i, \varepsilon_b^i\}$. In addition, $\Im(H_{\beta}) > \max\{\varepsilon_a^i, \varepsilon_b^i\}$ when $0.7 \approx f_a < 1$. Thus, loss enhancement is predicted by both the Bruggeman formalism and the Maxwell Garnett formalism.

Since the active and dissipative scenarios effectively represent two different sides of the same coin, henceforth in this section, we focus on gain enhancement. Let us now turn to the gain-enhancement index

$$\rho = \frac{\varepsilon_{Br}^i}{\min\{\varepsilon_a^i, \varepsilon_b^i\}}$$

(4)
estimated using the Bruggeman formalism. Gain enhancement is signified by $\rho > 1$. For $\varepsilon'_a = 2$, $\varepsilon'_b = 5$, and $\varepsilon^l_b = -0.04$, $\rho$ is plotted against volume fraction $f_a$ and the ratio $\varepsilon'_a/\varepsilon^l_b$ in Figure 3. Gain enhancement is evident for mid-range values of $f_a$ when $\varepsilon'_a/\varepsilon^l_b \approx 1$. Specifically for this particular example,

(a) $\rho$ is as high as about 1.05, with its maximum value occurring for $f_a \approx 0.6$ and $\varepsilon'_a/\varepsilon^l_b = 1$; and

(b) there is no gain enhancement for $\varepsilon'_a/\varepsilon^l_b \lesssim 0.95$ and for $\varepsilon'_a/\varepsilon^l_b \gtrsim 1.07$, regardless of the value of $f_a$.

The dependency of $\rho$ upon $\varepsilon'_a$ and $\varepsilon'_b$ is delineated in Figure 4, wherein $\rho$ is plotted against $f_a$ and $\varepsilon'_a/\varepsilon'_b$ for $\varepsilon'_a = -0.05$, $\varepsilon^l_b = -0.04$, and $\varepsilon'_b = 5$. As in Figure 3, $\rho$ is high for mid-range values of $f_a$ when the ratio $\varepsilon'_a/\varepsilon'_b$ deviates most from unity in Figure 4. Specifically for this particular example,

(a) $\rho$ is as high as about 1.4, with its maximum value occurring for $f_a \approx 0.7$ and $\varepsilon'_a/\varepsilon'_b = 0.1$;

(b) $\rho$ is as high as about 1.2, with its maximum value occurring for $f_a \approx 0.5$ and $\varepsilon'_a/\varepsilon'_b = 10$; and

(c) there is no gain enhancement for $\varepsilon'_a/\varepsilon'_b \approx 1$, regardless of the value of $f_a$.

3. Conditions for gain/loss enhancement

The foregoing and similar calculations led us to conclude that gain enhancement should be expected when

(i) $\varepsilon'_a < 0$ and $\varepsilon'_b < 0$,

(ii) the ratio $\varepsilon'_a/\varepsilon'_b$ is close to unity, and

(iii) the ratio $\varepsilon'_a/\varepsilon'_b$ is either very small or very large.
Loss enhancement should be expected when \( \varepsilon_i^a > 0, \varepsilon_i^b > 0 \), and the conditions (ii) and (iii) are satisfied. In order to formally establish this understanding soundly, we used the Bruggeman equation (1) to obtain the gradient

\[
\frac{d\varepsilon_{Br}}{df_a} = \frac{3 \varepsilon_{Br} (\varepsilon_a - \varepsilon_b)}{4 \varepsilon_{Br} + \varepsilon_a (1 - 3f_a) + \varepsilon_b (3f_a - 2)}.
\]

This expression underlies further analysis.
### 3.1. Gain enhancement

Suppose that both component materials are active, i.e. \( \varepsilon'_a < 0 \) and \( \varepsilon'_b < 0 \). If \( \varepsilon'_a \geq \varepsilon'_b \), then a sufficient condition for gain enhancement is that the gradient

\[
\lim_{f_a \to 0} \frac{d\varepsilon'_b}{d f_a} < 0.
\]

Given that

\[
\lim_{f_a \to 0} \varepsilon'_b = \varepsilon'_b,
\]

Equation (5) yields

\[
\lim_{f_a \to 0} \frac{d\varepsilon'_b}{d f_a} = \frac{3\varepsilon'_b (\varepsilon'_a - \varepsilon'_b)}{2\varepsilon'_b + \varepsilon'_a},
\]

and hence

\[
\lim_{f_a \to 0} \frac{d\varepsilon'_b}{d f_a} = \frac{3 [\varepsilon'_b (\varepsilon'_a - \varepsilon'_b) + \varepsilon'_b (\varepsilon'_b - \varepsilon'_b)] (2\varepsilon'_b + \varepsilon'_a) - [\varepsilon'_b (\varepsilon'_a - \varepsilon'_b) - \varepsilon'_b (\varepsilon'_b - \varepsilon'_a)] (2\varepsilon'_b + \varepsilon'_a)]}{(2\varepsilon'_b + \varepsilon'_a)^2 + (2\varepsilon'_b + \varepsilon'_a)^2}.
\]

The sufficient condition (6) for gain enhancement is therefore logically equivalent to

\[
\left[\varepsilon'_b (\varepsilon'_a - \varepsilon'_b) + \varepsilon'_b (\varepsilon'_b - \varepsilon'_b)\right] (2\varepsilon'_b + \varepsilon'_a) < \left[\varepsilon'_b (\varepsilon'_a - \varepsilon'_b) - \varepsilon'_b (\varepsilon'_b - \varepsilon'_a)\right] (2\varepsilon'_b + \varepsilon'_a).
\]

If \( \varepsilon'_b \geq \varepsilon'_a \), then a sufficient condition for gain enhancement is that the gradient

\[
\lim_{f_a \to 1} \frac{d\varepsilon'_b}{d f_a} > 0.
\]

Following the same argument as used to derive condition (10), we found that the sufficient condition (11) for gain enhancement is logically equivalent to

\[
\left[\varepsilon'_a (\varepsilon'_a - \varepsilon'_b) + \varepsilon'_a (\varepsilon'_a - \varepsilon'_b)\right] (2\varepsilon'_a + \varepsilon'_b) > \left[\varepsilon'_a (\varepsilon'_a - \varepsilon'_b) - \varepsilon'_a (\varepsilon'_a - \varepsilon'_b)\right] (2\varepsilon'_a + \varepsilon'_b).
\]

The special case \( \varepsilon'_a = \varepsilon'_b \) is noteworthy. Both the conditions (10) and (12) then reduce to

\[
(\varepsilon'_a - \varepsilon'_b)^2 > 0.
\]

Since condition (13) is always satisfied because \( \varepsilon'_a \neq \varepsilon'_b \), gain enhancement is guaranteed for all values of \( \varepsilon'_a \) and \( \varepsilon'_b \), provided that \( \varepsilon'_a = \varepsilon'_b \).
Figure 5. Top: Gain-enhancement subspace in the \((-\varepsilon_a^i, -\varepsilon_b^i)\) space, when \(\varepsilon_a^f = 2\) and \(\varepsilon_b^f = 5\). Bottom: Gain-enhancement subspaces in the \((\varepsilon_a^f, \varepsilon_b^f)\) space, when \(\varepsilon_a^i = -0.05\) and \(\varepsilon_b^i = -0.04\).
3.2. Loss enhancement

Suppose both component materials are dissipative, i.e. \( \varepsilon_a' > 0 \) and \( \varepsilon_b' > 0 \). If \( \varepsilon_a' \leq \varepsilon_b' \), then a sufficient condition for loss enhancement is that the gradient

\[
\lim_{f_a \to 0} \frac{d\varepsilon_l}{df_a} > 0, \tag{14}
\]

which, in the manner described in Section 3.1, is logically equivalent to the condition

\[
\left[ \varepsilon_b' (\varepsilon_a' - \varepsilon_b') + \varepsilon_b' (\varepsilon_a' - \varepsilon_b') \right] (2\varepsilon_b' + \varepsilon_a') > \left[ \varepsilon_b' (\varepsilon_a' - \varepsilon_b') - \varepsilon_b' (\varepsilon_a' - \varepsilon_b') \right] (2\varepsilon_b' + \varepsilon_a'). \tag{15}
\]

If \( \varepsilon_b' \leq \varepsilon_a' \), then a sufficient condition for loss enhancement is that the gradient

\[
\lim_{f_a \to 1} \frac{d\varepsilon_l}{df_a} < 0, \tag{16}
\]

which is logically equivalent to the condition

\[
\left[ \varepsilon_a' (\varepsilon_a' - \varepsilon_b') + \varepsilon_a' (\varepsilon_a' - \varepsilon_b') \right] (2\varepsilon_a' + \varepsilon_b') < \left[ \varepsilon_a' (\varepsilon_a' - \varepsilon_b') - \varepsilon_a' (\varepsilon_a' - \varepsilon_b') \right] (2\varepsilon_a' + \varepsilon_b'). \tag{17}
\]

As in Section 3.1, both conditions (15) and (17) reduce to condition (13) for the special case \( \varepsilon_a' = \varepsilon_b' \). Therefore, loss enhancement is guaranteed for all values of \( \varepsilon_a' \) and \( \varepsilon_b' \) when \( \varepsilon_a' = \varepsilon_b' \).

3.3. Numerical illustration

The conditions (10) and (12) provide a convenient means of exploring the parameter space of the relative permittivities of the component materials that support gain enhancement, and conditions (15) and (17) play the same role for loss enhancement. Let us illustrate this assertion with a numerical example.

In Figure 5, the parameter spaces that support gain enhancement are mapped for: (i) \((\varepsilon_a', -\varepsilon_b') \in (0, 1) \times (0, 1)\) with \(\varepsilon_a' = 2\) and \(\varepsilon_b' = 5\); and (ii) \((\varepsilon_a', \varepsilon_b') \in (0.5, 10) \times (0.5, 10)\) with \(\varepsilon_a' = -0.05\) and \(\varepsilon_b' = -0.04\). For \(\varepsilon_a' = 2\) and \(\varepsilon_b' = 5\), the gain-enhancement subspace in the \((\varepsilon_a', -\varepsilon_b')\) space is a window that contains \(\varepsilon_a' = \varepsilon_b'\) and becomes narrower as the magnitudes of \(\varepsilon_a'\) and \(\varepsilon_b'\) are decreased. For \(\varepsilon_a' = -0.05\) and \(\varepsilon_b' = -0.04\), two gain-enhancement subspaces in the \((\varepsilon_a', \varepsilon_b')\) space exist where \(\varepsilon_a'\) and \(\varepsilon_b'\) are dissimilar in magnitude with greater scope for gain enhancement arising when the magnitudes of \(\varepsilon_a'\) and \(\varepsilon_b'\) are increased. These trends gleaned from Figure 5 are wholly consistent with those evident in Figures 3 and 4.

3.4. Non-dissipative and non-active component materials

In passing, let us remark on the special case when both component materials are neither dissipative nor active, i.e. \(\varepsilon_a' = \varepsilon_b' = 0\). Provided that the possibility \(\varepsilon_b r = 0\) is excluded from
consideration (which is not physically plausible for the situation $\varepsilon_a \varepsilon_b > 0$ considered here), we infer from Equation (5) that $d\varepsilon_{Br}/df_a \neq 0$. Therefore, $\varepsilon_{Br}$ is either a uniformly increasing or a uniformly decreasing function of $f_a$. Hence, $\varepsilon_{Br}$ must lie between $\varepsilon_a$ and $\varepsilon_b$ for all values of $f_a \in [0,1]$.

4. Closing remarks

Using the Bruggeman formalism, we have established in the foregoing sections that an HCM comprising two active (resp. dissipative) component materials may exhibit more gain (resp. loss) than either of its component materials. For the range of $\varepsilon_a$ and $\varepsilon_b$ values explored in numerical examples here, gain enhancements of up to 40% were found. Furthermore, sufficient conditions for such gain enhancement and loss enhancement have been established under conditions (10) and (12), and (15) and (17), respectively. These enhancements arise when (i) the imaginary parts of the relative permittivities of both component materials are similar in magnitude and (ii) the real parts of the relative permittivities of both component materials are dissimilar in magnitude. Similar gain/loss enhancements also emerge from the Maxwell Garnett formalism for dilute composite materials.

The reported phenomena of gain enhancement and loss enhancement are likely to be exacerbated by directional effects in anisotropic HCMs, as has been established for nonlinearity enhancement [19,20] and group-speed enhancement.[21]

The Bruggeman homogenization formalism is rigorously established, since it represents the lowest-order formulation of the strong-permittivity-fluctuation theory.[22] But the formalism does not shed light on the physical mechanisms that give rise to the reported gain and loss enhancements. Appropriate experimental studies may provide physical insight. In this regard, we remark that the microstructure (or nanostructure) of the composite materials considered in the foregoing study, i.e. a random dispersal of electrically small spheres, is a relatively simple one that is amenable to experimental realization. This random microstructure is in contrast to the rather complex microstructures that are often associated with metamaterials.[3] Moreover, the permittivity values chosen for the component materials in the numerical illustrations of gain and loss enhancements provided in Figures 1–5 are practicable ones for the most part.

Disclosure statement

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