Improved bounds for the overall properties of a nonlinear composite dielectric

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Abstract

The construction of generalized Hashin-Shtrikman bounds for nonlinear composite problems relies on the introduction of a comparison material. In recent work a nonlinear comparison medium has been used; however this requires detailed knowledge of the properties of the trial fields that are employed. The fields used have the property of ‘bounded mean oscillation’ and this enables the size of the penalty exacted by using a nonlinear comparison material to be controlled. Some recent results concerning Riesz transforms and the Beurling operator are used here to reduce the effect of the penalty and hence to generate improved bounds. In addition, an exact solution is established for a particular class of composites.

1 Introduction

The composites considered here consist of a mixture of two dielectric phases. Each phase is characterized by a convex energy function, which, to be definite, is assumed to grow at least as fast as quadratically at infinity. One approach that has been used to bound the overall energy function of such materials was developed by Talbot & Willis (1985) and Willis (1986) and is the extension to
nonlinear problems of the variational principles of Hashin & Shtrikman (1962; 1963). The methodology relies on the introduction of a comparison medium and, for the composite considered here, a lower bound can be obtained by choosing a linear comparison medium. However, if the energy of one of the phases grows faster than quadratically a finite upper bound cannot be obtained by such a choice. In Talbot & Willis (1994; 1995) this difficulty was avoided by using a nonlinear comparison medium which was linear up to some value of the field and nonlinear thereafter. The introduction of nonlinearity exacts a penalty which is a function of the size of the set where the trial field used is large. The trial fields used by Talbot & Willis (1994; 1995) have the property of bounded mean oscillation (John & Nirenberg, 1961) and this property was used to bound the size of this set. This enabled a new bound to be constructed which showed a small improvement on bounds obtained by elementary methods.

In Talbot & Willis (1994; 1995) a uniform comparison medium was used. An alternative is to use a comparison material which is itself a composite. Linear comparison composites were first used by Ponte Castañeda (1991) to obtain bounds for nonlinear materials which involved a bound for the linear composite. More recently, upper bounds have been obtained by Talbot & Willis (1996, 1997, 1998) using a comparison composite whose behaviour in each phase is linear up to some magnitude of the field and nonlinear thereafter. The resulting bound is sensitive to the three-point statistics of the microstructure through parameters introduced by Milton (1981), who also demonstrated that they must lie in certain ranges. Taking the extreme values generates bounds which are valid for all three-point statistics and which, for linear dielectric materials, coincide with the Hashin-Shtrikman bounds. However for nonlinear materials upper bounds obtained in this way improve on bounds obtained using a uniform comparison material. Lower bounds for composites of the type considered here which involve three-point statistics were obtained by Ponte Castañeda (1992).

When using a nonlinear comparison composite there is still a penalty associated with the nonlinearity. In this paper some recent results of Bañuelos & Wang (1995) for Riesz transforms and Astala (1994) for the Beurling operator are used to obtain an improved bound on the measure of the set over which the trial field is large. In addition, for a composite consisting of one linear and one nonlinear phase, an exact solution is obtained for a class of microstructures.
In §2 the bounding procedure using a comparison composite is briefly outlined. Section 3 develops the new bounds for the penalty term. In §4 some results are presented for a particular composite and an exact solution is identified.

2 Formulation

The composite to be considered is a two-phase dielectric occupying a ball $B$ of unit radius in $d$ dimensions. Here $d$ is 2 or 3 and the ball is centred at the origin. The scale of the composite is taken to be so fine that the shape of $B$ has no influence and hence there is no loss of generality in considering a ball. The microgeometry is also assumed to be statistically isotropic. The constitutive behaviour is described by an energy function

$$W(E, x) = \sum_{r=1}^{2} W_r(E) \chi_r(x),$$

(2.1)

where $E$ is the electric field, $W_r$ is the energy function of material of type $r$ and $\chi_r$ is the characteristic function of the region occupied by material of this type. The functions $W_r$ are assumed to be convex and functions of $|E|$ only. The problem is to bound the mean energy, $\bar{W}$, of the composite, defined by

$$\bar{W}(\bar{E}) = \frac{1}{|B|} \inf_{E \in K} \int_B W(E, x) dx,$$

(2.2)

where

$$K = \left\{ E : E = -\nabla \phi, \quad |B|^{-1} \int_B E(x) dx = \bar{E} \right\}$$

(2.3)

and $|B|$ denotes the volume of $B$. An upper bound is constructed by introducing a comparison composite with energy function $\tilde{W}(E, x)$, having the same microgeometry as the nonlinear composite and defining

$$(W - \tilde{W})_*(P, x) = \inf_E \left\{ P \cdot E - W(E, x) + \tilde{W}(E, x) \right\}.$$

(2.4)

Substituting the upper bound implied by (2.4) into (2.2) now gives

$$|B| \bar{W}(\bar{E}) \leq \int_B \left[ P \cdot E + \tilde{W}(E, x) - (W - \tilde{W})_*(P, x) \right] dx$$

(2.5)
for any field $\mathbf{P}$ and any $\mathbf{E} \in K$. Each $W_r$ is now assumed to grow at least quadratically.

Let

$$
\hat{W}(\mathbf{E}, x) = \sum_{r=1}^{2} \hat{W}_r(\mathbf{E}) \chi_r(x) = \sum_{r=1}^{2} \frac{1}{2} \varepsilon_{0r} \mathbf{E} \cdot \mathbf{E} \chi_r(x) + N(\mathbf{E}) H(|\mathbf{E}| - \lambda) \quad (2.6)
$$

where the constants $\lambda$, $\varepsilon_{0r}$ remain to be chosen and $H$ denotes the Heaviside step function. The function $N(\mathbf{E})$ depends on the application and grows at least as fast as any of the $W_r$ as $|\mathbf{E}| \to \infty$. It follows that

$$
|B| \hat{W}(\mathbf{E}) \leq \int_B \left[ \sum_{r=1}^{2} \frac{1}{2} \varepsilon_{0r} \mathbf{E} \cdot \mathbf{E} \chi_r(x) + \mathbf{P} \cdot \mathbf{E} - (W - \hat{W})(\mathbf{P}, x) \right] \, dx \\
+ \int_{S_\lambda} N(\mathbf{E}) \, dx, \quad (2.7)
$$

where $S_\lambda = \{x \in B : |\mathbf{E}(x)| > \lambda\}$. The last term in (2.7) is the penalty for taking $\hat{W}$ to be non-quadratic. The bound of Talbot & Willis (1994) is recovered by setting $\varepsilon_{01} = \varepsilon_{02} = \varepsilon_0$, say, and choosing $N(\mathbf{E})$ appropriately.

The polarization $\mathbf{P}$ is now chosen to have the piecewise constant form

$$
\mathbf{P} = \mathbf{P}_1 \chi_1 + \mathbf{P}_2 \chi_2 \quad (2.8)
$$

and the trial field $\mathbf{E}$ is taken as

$$
\mathbf{E} = \overline{\mathbf{E}} - \Gamma(f_1 \eta)(x), \quad (2.9)
$$

where $\eta$ is a constant vector and

$$
f_1(x) = \begin{cases} 
\chi_1(x) - c_1, & x \in B \\
0, & x \notin B,
\end{cases} \quad (2.10)
$$

with $c_1$ the volume fraction of phase 1. The kernel of the operator $\Gamma$ is $\frac{\partial^2 G}{\partial x_1 \partial x_2}$, where $G$ is the infinite body Green’s function for a medium with dielectric constant equal to unity. It is easy to check that the integral of $\Gamma$ over $B$ is a constant. It follows that the mean value of the right side of (2.9) is $\overline{\mathbf{E}}$ as $f_1$ has zero mean value. Substituting the trial field (2.9) into (2.7) leads to an expression which involves the one, two and three-point statistics of the composite. The terms involving three points can be expressed in terms of
the parameters $\zeta_1$ and $\zeta_2 = 1 - \zeta_1$ of Milton (1981) and the terms involving two points are dealt with by using the results

$$
\int_B |\Gamma(f_1\eta)|^2 \, dx \leq \int_{R^d} |\Gamma(f_1\eta)|^2 \, dx = \int_B f_1(x)\eta \cdot \Gamma(f_1\eta)(x) \, dx = \frac{c_1c_2}{d} I, \quad (2.11)
$$

where $c_2 = 1 - c_1$. The result after minimizing with respect to $P_1$ and $P_2$ and using the fact that $(W - \hat{W})_{**} = -(W - W)^{**}$ is

$$
|B| \hat{W}(E) \leq c_1 \left\{ \frac{1}{\varepsilon_0} \left[ |E - \frac{c_2}{d}\eta|^2 + \frac{(d-1)}{d^2} c_2 \zeta_1 |\eta|^2 \right] - (\hat{W}_1 - W_1)^{**}(|E - \frac{c_2}{d}\eta|) \right\} + c_2 \left\{ \frac{1}{\varepsilon_0} \left[ |E + \frac{c_1}{d}\eta|^2 + \frac{(d-1)}{d^2} c_1 \zeta_2 |\eta|^2 \right] - (\hat{W}_2 - W_2)^{**}(|E + \frac{c_1}{d}\eta|) \right\} + \int_{S_{\lambda}} N(E) \, dx. \quad (2.12)
$$

Further details can be found in Talbot & Willis (1996). The best bound follows by minimizing the right side of (2.12) with respect to $\varepsilon_0$, $\varepsilon_0$, $\lambda$ and $\eta$.

3 The nonlinear penalty term

First let $N(E) = N_1(E)\chi_1(x) + N_2(E)\chi_2(x)$. Then, with

$$
\nu(s) = \max_{r} \{N_r(s)\}, \quad (3.1)
$$

the penalty term is bounded by

$$
\int_{\lambda}^{\infty} \mu(s) \, d\nu(s) \quad (3.2)
$$

where

$$
\mu(s) = |\{x \in B : |E| > s\}|. \quad (3.3)
$$

The composite is isotropic and hence it suffices to take $E = (E, 0, 0)$ and $\eta = (\eta, 0, 0)E$. In this case the components of the trial field are given by

$$
E_i = E(\delta_{i1} - \Gamma_{i1}(f_i)\eta). \quad (3.4)
$$
Let \( g \) be the vector with components \( g_i = \Gamma_{ij}(f_1) \). Then
\[
\mathbf{E} - \mathbf{E} - g \eta \mathbf{E}
\]
and if \( |E|^2 > s^2 \) at least one of the \( |E_i|^2 \) is greater than \( s^2/d \). Hence
\[
\{ x \in B : |E| > s \} \subseteq \bigcup_{i=1}^{d} \{ x \in B : |E_i| > s/\sqrt{d} \}
\]
and it follows that
\[
\mu(s) \leq \sum_{i=1}^{d} \left| \{ x \in B : |E_i| > s/\sqrt{d} \} \right|.
\]  \hspace{1cm} (3.6)

Next, if \( |E_1| > s/\sqrt{d} \), then \( |g_1| = \eta |\mathbf{E}| > s/\sqrt{d} - \mathbf{E} \). Hence
\[
\left| \{ x \in B : |E_1| > s/\sqrt{d} \} \right| \leq \left| \left\{ x \in B : |g_1| > \frac{s - \sqrt{d |\mathbf{E}|}}{\sqrt{d |\eta| |\mathbf{E}|}} \right\} \right|
\]
with similar expressions for the remaining components of \( \mathbf{E} \). It follows that
\[
\mu(s) \leq \sum_{i=1}^{d} \left| \left\{ x \in B : |g_i| > \frac{s - \sqrt{d |\mathbf{E}|}}{\sqrt{d |\eta| |\mathbf{E}|}} \right\} \right|
\]  \hspace{1cm} (3.7)

and the problem now is to bound \( \left| \{ x \in B : |g_i| > s \} \right| \).

In Talbot & and Willis (1994) a bound on \( \mu(s) \) was established by using the fact that the operator \( \Gamma \) that was used has the property of bounded mean (square) oscillation. In this work the estimates are improved by exploiting the relationship between \( \Gamma_{ij} \) and the Riesz transforms. Now, \( \Gamma_{ij} \) has Fourier multiplier \( \tilde{\Gamma}_{ij}(\xi) = \xi_i \xi_j / |\xi|^2 \) and since the Fourier multiplier of the Riesz transform \( R_i \) is \( i \xi_i / |\xi| \) (see Stein (1979)), it follows that \( \Gamma_{ij} = -R_i R_j \). Theorem 4 of Bañuelos and Wang (1995) can now be used to get
\[
\| \Gamma_{ij}(f_1) \|_p = \| R_i R_j(f_1) \|_p \leq (p-1) \| f_1 \|_p, \quad p \geq 2.
\]  \hspace{1cm} (3.8)

It follows that
\[
\left| \{ x \in \mathbb{R}^d : |g_i| > s \} \right| = \left| \{ x \in \mathbb{R}^d : |\Gamma_{ij}(f_1)| > s \} \right| \leq \left( \frac{p-1}{s} \right)^p \| f_1 \|_p^p, \quad p \geq 2.
\]  \hspace{1cm} (3.9)
The best bound follows by minimizing the right side of (3.9) with respect to \( p \) for any given \( s \). Some numerical experiments were performed and it was found that a sufficiently accurate approximation to the minimum, which is itself a strict bound, can be obtained by using 
\[
(1-c_m)e^{-1} \exp \left( -\frac{s}{c_m e} \right) \left( 1 + \exp \left( \ln \left( \frac{1-c_m}{c_m} \right) \frac{s}{c_m e} \right) \right).
\]

This bound is valid for any number of space dimensions. However when \( d = 2 \), a better bound can be obtained by using the results of Astala (1994) for the Beurling operator \( S \). This is a complex operator with Fourier multiplier \( \xi/\bar{\xi} \), so that in terms of Riesz transforms
\[
S = R_2^2 - R_1^2 - 2iR_1R_2.
\]
Now \( R_2^2 + R_1^2 = -I \), where \( I \) is the identity operator, so that
\[
S = -I - 2R_1^2 - 2iR_1R_2.
\]
It follows that
\[
g_1 = -\frac{1}{2}Re(S + I)(f_1), \quad g_2 = -\frac{1}{2}ImS(f_1). \tag{3.13}
\]
Next, Corollary 1.7 of Astala (1994) states that, for any measurable set \( E \subset B \),
\[
\int_B |S(\chi_E)| \, dm \leq |E| \log \left( \frac{\alpha}{|E|} \right).
\]
where, due to work of Eremenko & Hamilton (1995), the constant \( \alpha \) has been identified as \( \epsilon \pi \). Following Astala (1994), let \( E_+ = \{ z \in B : ReS(f_1) > t \} \). Then, as \( S \) has a symmetric kernel,
\[
t |E_+| \leq Re \int_{E_+} S(f_1) \, dm = Re \int_{B} f_1S(\chi_{E_+}) \, dm \leq c_m \int_B |S(\chi_{E_+})| \, dm
\leq c_m |E_+| \log \left( \frac{\alpha}{|E_+|} \right). \tag{3.15}
\]
Hence \( |E_+| \leq \alpha \exp(-t/c_m) \). The same argument can be used to bound \( |E_-| = |\{ z \in B : ReS(f_1) < -t \}| \) and the result
\[
|\{ z \in B : |ReS(f_1)| > t \}| < 2\alpha \exp \left( -\frac{t}{c_m} \right). \tag{3.16}
\]
follows. Similar reasoning can be used to obtain the same bound on $|\{z \in B : \text{Im} S(f_1)\}| >$

It follows that

$$|\{z \in B : |g_1| > s\}| \leq 2\pi \exp \left( -\frac{2s}{c_m} + c_m \right),$$

$$|\{z \in B : |g_2| > s\}| \leq 2\pi \exp \left( -\frac{2s}{c_m} \right).$$

**(3.17)**

Bounds on $\mu(s)$ can now be obtained by using (3.10) in (3.7) in three dimensions and (3.17) in (3.7) in two dimensions. This immediately induces a bound on the penalty term. One clear difference between the two cases is that, in two dimensions the information in (3.14) used to obtain (3.17) relates only to the values of $S(\chi E)$ in $B$, whereas in using (3.8) to obtain (3.10), the effect of the value of $R_tR_t(f_1)$ outside $B$ is included as well.

## 4 A special composite

In this section the special case of a composite comprising one linear phase and one nonlinear phase is considered. Let

$$W_1(E) = \frac{1}{2} r_1 |F|^2$$

**(4.1)**

and

$$N(E) = (W_2(E) - W_2(\lambda)) H(|E| - \lambda) \chi(\mathbf{x}),$$

**(4.2)**

so that $\nu(s) = W_2(s) - W_2(\lambda)$. Then, in (2.12), the minimum over $\varepsilon_{01}$ is attained when $\varepsilon_{01} = \varepsilon_1$. On using (3.2) and the forms for $\mathbf{E}$ and $\eta$ given after (3.3), (2.12) becomes

$$|E| \langle \bar{W}(E) \rangle \leq c_1 \left\{ \frac{1}{2} \varepsilon_{01} \bar{E}^2 \left[ 1 + \frac{c_2}{d} \eta \right] ^2 + \frac{(d-1)}{d^2} c_2 \zeta_i \eta^2 \right\} + c_2 \left\{ \frac{1}{2} \varepsilon_{02} \bar{E}^2 \left[ 1 + \frac{c_1}{d} \eta \right] ^2 + \frac{(d-1)}{d^2} c_2 \zeta_i \eta^2 \right\} - (\bar{W}_2 - W_2) \langle (1 + \frac{c_2}{d} \eta, \bar{E}) \rangle + \int_\lambda^\infty \mu(s) d\nu(s).$$

**(4.3)**

When $\zeta_1 = 1$, it is shown in Appendix A that the bound (4.3) becomes

8
\[ |B| \bar{W}(E) \leq \min_\omega \left\{ \frac{1}{2} \varepsilon_1 c_1 E^2 \left[ (1 - c_2 \omega)^2 + (d - 1)c_2 \omega^2 \right] + c_2 W_2(1 + c_1 \omega \mid E) \right\}, \]

(4.4)

This is exactly the lower bound when \( \zeta_1 = 1 \) given by Ponte Castañeda (1992), equation (3.23). Hence the right side of (4.4) is the overall energy of the composite when \( \zeta_1 = 1 \). An analogous result was obtained as the result of a computation by Talbot & Willis (1998) in the context of bounding the overall response of an elastoplastic composite.

For \( \zeta_1 \neq 1 \) further knowledge of \( W_2 \) is required. Willis (1986) and Talbot & Willis (1994) considered the example

\[ W_2(E) = \frac{1}{2} \varepsilon_2 |E|^2 + \frac{1}{4} \gamma |E|^4, \quad (4.5) \]

where \( \varepsilon_2 \) and \( \gamma \) are constants. Taking \( N(E) = \frac{1}{4} \gamma (|E|^4 - \lambda^4)H(|E| - \lambda)\chi_2(x) \), it follows that \((\bar{W} - W_2)''\) has the same form as a function considered by Talbot & Willis (1996). The bound can now be written

\[ |B| \bar{W}(E) \leq \frac{1}{4} \varepsilon_1 c_1 E^2 \left[ \left( 1 - \frac{c_2}{d} \eta \right)^2 + \frac{(d - 1)}{d^2} c_2 \zeta_1 \eta^2 \right] \]
\[ + \frac{1}{4} c_2 \varepsilon_2 E^2 \left[ \left( 1 + \frac{c_1}{d} \eta \right)^2 + \frac{(d - 1)}{d^2} c_1 \zeta_2 \eta^2 \right] \]
\[ + c_2 \gamma \bar{E}^4 \left( 1 + \frac{c_1}{d} \eta \right)^4 \Psi(\lambda_2, \hat{X}_2) + \gamma \int_\lambda^\infty s^3 \mu(s) ds, \quad (4.6) \]

where

\[ \hat{X}_2 = \frac{1}{2} + \frac{(d - 1)}{d^2} c_1 \zeta_2 \eta^2 \left( 1 + \frac{c_1}{d} \eta \right)^2, \quad \lambda_2 = \lambda / \left( 1 + c_1 \varepsilon / d \eta \right) \quad (4.7) \]

and \( \Psi \) is the function described in Appendix B of Talbot & Willis (1996). Manipulation of formulae given there leads to

\[ \Psi(x, y) = \frac{1}{2} y x^2 - \frac{1}{4} \left( \frac{x - 2y}{x - 1} \right)^2 \left( \frac{x^2 - 2y}{x} \right) \left( \frac{x^2 - 2y}{x} \right), \quad \frac{1}{2} < y < \frac{1}{2} x, \]
\[ = \frac{1}{2} y x^2, \quad \frac{1}{2} x < y < \frac{1}{2} x^2, \]
\[ = \frac{1}{4} x^4, \quad \frac{1}{2} x^2 < y. \]

(4.8)

Results have been obtained for the parameter values \( \varepsilon_2 / \varepsilon_1 = 8 \) and \( c_1 = \frac{1}{2} \). Figures 1 and 2 show sets of bounds when \( d = 2 \) and \( d = 3 \), respectively. The
outer two curves are the simple classical bounds obtained by substituting constant trial fields into (2.2) and the principle dual to (2.2). The curves labelled (a) and (b) are upper bounds when \( \zeta_1 = 0 \). Curve (b) was obtained by estimating the penalty using the results of §3, while for curve (a) the results of Talbot & Willis (1994) were used. The remaining curves are (c), the lower bound when \( \zeta_1 = 0 \), calculated from formulae given by Ponte Castañeda (1992), and (d), the exact result when \( \zeta_1 = 1 \). It can be seen that the results of §3 give an improvement over previous upper bounds which is more significant when \( d = 2 \). When no information is available concerning the three-point statistics of the medium, the best bounds now available are curves (b) and (d).

Although it may be possible to improve further the estimate (3.10) (S. Montgomery-Smith, private communication), the effect on the upper bound is likely to be small. For the energy functions considered in this section, Talbot (1999) used linear bounds for a particular microstructure and was able to obtain tighter bounds for a nonlinear matrix–inclusion composite.

### Appendix A. The upper bound when \( \zeta_1 = 1 \).

When \( \zeta_1 = 1, \zeta_2 = 0 \) and in (4.3) it is necessary to find

\[
\min_{\varepsilon_{02}} \left\{ \frac{1}{2} \varepsilon_{02} E^2 \left[ 1 + \frac{c_1}{d} \eta \right]^2 - (\hat{W}_2 - W_2)^{**}(1 + \frac{c_1}{d} \eta, E) \right\}. \tag{A.1}
\]

First choose \( \lambda > 1 + \frac{c_1}{d} \eta, E \). Now

\[
(\hat{W} - W_2)(s) = \frac{1}{2} \varepsilon_{02} s^2 + (W_2(s) - W_2(\lambda))H(s - \lambda) - W_2(s) \tag{A.2}
\]

and this is convex for \( s > \lambda \). With the assumption that \( W \) is twice differentiable it is clearly possible to choose \( \varepsilon_{02} \) so that \( (\hat{W}_2 - W_2)' > 0 \) for \( s < \lambda \). It follows that \( \hat{W}_2 - W_2 \) is a convex function for all \( s \) and particularly for \( s = 1 + \frac{c_1}{d} \eta, E \). Hence

\[
(\hat{W}_2 - W_2)^{**}(1 + \frac{c_1}{d} \eta, E) = (\hat{W}_2 - W_2)(1 + \frac{c_1}{d} \eta, E)
\]

and (A.1) is bounded by \( W_2(1 + \frac{c_1}{d} \eta, E) \). The only term in (4.3) that still depends on \( \lambda \) is the penalty, so that, in the limit \( \lambda \to \infty \), the bound is given by (4.4), where the change of variables \( \omega = \eta/d \) has been used.
References


Figure 1: A set of bounds when $d = 2$. 
Figure 2: A set of bounds when \( d = 3 \).