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Bounds for the effective conductivity of a composite with an imperfect interface

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The problem of bounding the effective conductivity of a two-phase composite with an imperfect interface is considered. The interface can be either highly conducting or resistive, and both the material properties and geometric arrangement of the phases can be anisotropic. The problem is formulated variationally and by choosing appropriate trial fields, new bounds are obtained in terms of upper and lower bounds on the effective conductivity of a composite with the same microgeometry in which the phases are perfectly bonded. The methodology also applies to composites with a nonlinear interface, and a particular example is described.

Keywords: composites; imperfect interface; bounds; variational methods

1. Introduction

The transmission conditions at the interface separating materials in multi-phase composites can have a significant effect on the overall properties. This is seen in the experimental work of Garret & Rosenberg (1974) and Hasselman & Donaldson (1992). Electrical contact resistance often appears, due to the presence of a thin highly resistive layer or 'interphase' between two conducting phases. The effects of a thin layer can be modelled by an interface with appropriate discontinuous transmission conditions. Here, the electric potential jumps across the interface, while the normal component of current is continuous across it. On the interface, the jump in the electric potential is proportional to the normal component of the current.

On the other hand, the effective properties of mixtures can be strongly influenced by surface conduction of current in thin layers between bulk phases. The conduction layer between phases is often modelled by an interface across which the normal component of current suffers a jump, whereas the electric potential is continuous across the interface. Here, the jump in the normal current is proportional to the surface Laplacian of the electric potential at the interface. Both of these transmission conditions are distinct from the standard 'perfectly bonded' interface conditions, where both the electric potential and current are continuous across material interfaces. We refer to the former interface transmission condition as a resistive interface and to the latter as a highly conducting interface.

In this paper we derive new bounds on the overall conductivity tensor for these two cases. In addition, we consider nonlinear interfacial transmission conditions that model the dielectric breakdown of a thin interphase between two materials. New bounds are obtained for this case. The methods we develop are new and variational in nature.

The variational principles for the description of the effective conductivity for multiphase composites with resistive interface follow from the work of Hashin (1992). These principles are the analogues of the Dirichlet and Thompson variational principles for perfectly bonded composites. Variational principles analogous to those of Dirichlet and Thompson for the effective conductivity of composites with highly conducting interface are developed in Lipton (1997*a*). New variational principles of comparison type for resistive interfaces are introduced in the work by Lipton & Vernescu (1995, 1996). The variational principles of Lipton & Vernescu (1996) recover those of Hashin & Shtrikman (1962) in the limit of perfectly bonded interface conditions. For the highly conducting interface case, variational principles of comparison type were introduced in Lipton (1997*a*). These principles also recover those of Hashin & Shtrikman (1962) in the limit of perfect bonding.

Lipton & Vernescu (1996) obtained upper and lower bounds on the effective conductivity when the interface is resistive, using variational principles of comparison type. These bounds were applied to predict new size effects for particle reinforced suspensions. For a composite consisting of a distribution of particles embedded in a matrix, the lower bound on the effective conductivity is given in terms of the effective conductivity of a composite having the same microgeometry, with non-conducting inclusions. The lower bound also contains additional geometric information, including the particle size distribution and the particle volume fraction. It is shown in Lipton & Vernescu (1996) that for fixed particle volume fraction, this bound is optimal in the limit when the size of the inclusions tends to zero. The associated upper bound on the effective conductivity is given in terms of the Hashin–Shtrikman upper bound for the effective conductivity of a perfectly bonded composite with the same geometry, as well as information on the particle size distribution and particle volume fraction. We show here that this bound can be improved. To fix ideas, we consider an isotropic monodisperse suspension of spheres of radius a. For a matrix of conductivity σ_2 containing an isotropic monodisperse suspension of spherical particles of conductivity σ_1, we denote the effective conductivity by σ^{e}. The volume fraction of the particles and matrix are given by c_1 and c_2, respectively. The interfacial contact resistance is characterized by β, with units of conductivity per unit length. We suppose that the particles are better conductors than the matrix, i.e. $\sigma_1 > \sigma_2$. We introduce the function $C(x)$ defined by

$$C(x) = x - \frac{\sigma_1(c_1 + \gamma(x - \bar{\sigma}))^2}{\sigma_1\gamma^2(x - \bar{\sigma}) + c_1(1 + (a\beta)/\sigma_1)}, \tag{1.1}$$

where $\gamma = (\sigma_1 - \sigma_2)^{-1}$ and $\bar{\sigma} = c_1\sigma_1 + c_2\sigma_2$. One easily checks that this function is monotone increasing with x. With this function in hand, we can write the upper bound on σ^{e}, given by (3.62) of Lipton & Vernescu (1996) as

$$\sigma^{\mathrm{e}} \leqslant C(HS^+), \tag{1.2}$$

where HS^+ is the Hashin–Shtrikman upper bound on the effective conductivity for perfectly bonded composites $\tilde{\sigma}$ given by

$$\tilde{\sigma} \leqslant HS^+ = \sigma_1 - \frac{c_2}{\gamma - c_1/(3\sigma_1)}. \tag{1.3}$$

This provides the motivation to search for a tighter upper bound that is obtained by replacing HS^+ with $\tilde{\sigma}$. The methods developed in this paper establish the desired upper bound

$$\sigma^{\mathrm{e}} \leqslant C(\tilde{\sigma}), \tag{1.4}$$

where $\tilde{\sigma}$ is the effective property of a composite with the same microstructure, but with perfectly bonded inclusions (see theorem 4.2).

When the interface is highly conducting, upper and lower bounds on the effective conductivity are obtained in Lipton (1997*a*). These bounds were also applied to predict new size effects for particle reinforced suspensions. When the interface is highly conducting, the comparison variational principles developed in Lipton (1997*a*) are used to obtain an upper bound expressed in terms of the effective conductivity of a matrix containing a distribution of perfectly conducting particles. The upper bound is also given in terms of the particle size distribution and the particle volume fraction. It is shown in Lipton (1997*b*) that for fixed particle volume fraction, this bound is optimal in the limit when the size of the inclusions tends to zero. The associated lower bound is given in terms of the Hashin–Shtrikman lower bound for the effective conductivity of a perfectly bonded composite with the same geometry, together with information on the particle size distribution and particle volume fraction. We show here that this bound can be improved. The highly conducting interface is characterized by α, with units of conductivity multiplied by unit length. To fix ideas, we restrict our attention again to mono-disperse suspensions of spheres of common radius a and introduce the function $H(x)$ defined by

$$H(x) = x - \frac{(c_1 + \gamma(x - \bar{\sigma}))^2}{\gamma^2(x - \bar{\sigma}) - c_1(a/(2\alpha))}. \tag{1.5}$$

One easily checks that this function is monotone increasing with x. With this function in hand, we can write the lower bound on σ^{e} given by (6.19) of Lipton (1997*a*) as

$$\sigma^{\mathrm{e}} \geqslant H(HS_-), \tag{1.6}$$

where HS_- is the lower Hashin–Shtrikman bound on the effective conductivity $\tilde{\sigma}$ of a perfectly bonded composite given by

$$\tilde{\sigma} \geqslant HS_- = \sigma_1 - \frac{c_2}{\gamma - c_1/(3\sigma_1)}. \tag{1.7}$$

Again, one seeks to establish a tighter lower bound obtained by replacing HS_- with $\tilde{\sigma}$. We are able to establish the desired lower bound given by

$$\sigma^{\mathrm{e}} \geqslant H(\tilde{\sigma}), \tag{1.8}$$

where $\tilde{\sigma}$ is the effective property of a composite with the same microstructure but with perfectly bonded inclusions (see theorem 3.2).

In order to obtain the tighter bounds, we start with variational principles of Dirichlet type for both the resistive and highly conducting interface cases. Our approach is to partly follow the derivation of the comparison principles of Lipton & Vernescu (1996) and Lipton (1997*a*), in that we consider the Fenchel dual of the surface energies associated with the interfacial transmission conditions. Once we have dualized the surface energies, we obtain a variational structure for the bulk energy analogous to the problem of finding the effective properties of a linear thermoelastic composite. This latter problem was considered by Talbot & Willis (1992) in the context of finding bounds for nonlinear composites. The solution was given by Talbot & Willis (1992), who used formulae of Levin (1967) and Laws (1973), and involves the effective properties of the purely mechanical problem. In the present context, this allows the bounds to be expressed in terms of the effective conductivity tensor of the perfectly bonded composite with the same microgeometry.

We apply this new method to also recover the lower bounds of Lipton & Vernescu (1996) for the resistive interface case and the upper bound of Lipton (1997*a*) for the highly conducting interface case. We go on to provide new upper and lower bounds on the effective properties when both conductors are anisotropic. The methodology provided here can be adapted to nonlinear interfacial transmission conditions. To fix ideas, we consider an interface condition that models dielectric breakdown at an interface between two materials. Here, the dielectric potential is allowed to be discontinuous across the interface separating the particle and matrix phases. The jump in the potential ϕ across the interface is written as $\phi_{\mathrm{p}} - \phi_{\mathrm{m}} = [\phi]$, where the subscripts indicate the side of the interface on which the potential is evaluated. On the part of the interface where $-\phi_0 < -[\phi] < \phi_0$, no current passes across the interface. At points on the interface where $-[\phi]$ reaches $-\phi_0$, breakdown occurs and the normal current flows through the interface into the particle. On the portion of the interface where $-[\phi]$ reaches ϕ_0, breakdown also occurs and the normal current flows into the matrix. We apply the bounds to obtain new size effects for particle reinforced composites in § 6.

The paper is organized as follows. In § 2 the basic variational structure is outlined, in §§ 3 and 4 the new bounds for a composite with a highly conducting interface and a resistive interface, respectively, are derived and some results for random composites are presented in § 5. We obtain new bounds for composites with nonlinear interface conditions in § 6.

2. Formulation

The medium considered here is constructed by selecting a cube Q of composite and replicating it to form an infinite periodic medium. For convenience, Q is assumed to have unit volume and to contain the origin. The composite consists of a distribution of N particles with conductivity tensor $\boldsymbol{\sigma}_1$, embedded in a matrix with conductivity tensor $\boldsymbol{\sigma}_2$. The region occupied by particle r is denoted Ω_r and has boundary Γ_r. The conductivity tensor $\boldsymbol{\sigma}(\boldsymbol{x})$ of the medium is given by

$$\boldsymbol{\sigma}(\boldsymbol{x}) = \boldsymbol{\sigma}_1 f_1(\boldsymbol{x}) + \boldsymbol{\sigma}_2 f_2(\boldsymbol{x}), \tag{2.1}$$

where $f_r(\boldsymbol{x})$ is the characteristic function of the region occupied by phase r, taking the value 1 in that phase and zero outside. In what follows, the particles and matrix are designated phases 1 and 2, respectively. If the electric potential is decomposed into a periodic fluctuation $\tilde{\phi}$ and a linear part $\bar{\boldsymbol{E}} \cdot \boldsymbol{x}$, the average electric field as measured by an outside observer is

$$\bar{\boldsymbol{E}} = \int_{\partial Q} (\tilde{\phi} + \bar{\boldsymbol{E}} \cdot \boldsymbol{x}) \boldsymbol{n} \, \mathrm{d}s, \tag{2.2}$$

where ∂Q is the boundary of Q and $\boldsymbol{n}$ is the outward normal to ∂Q. In the sequel, Γ will denote the internal boundary between the phases and a jump in a quantity q across Γ will be represented by $[q] = q_1 - q_2$. First, if the interface is highly conducting, $\tilde{\phi}$ is continuous across Γ and satisfies

$$\left.\begin{aligned} \Delta\tilde{\phi} &= 0, && \text{in each phase,} \\ [\boldsymbol{\sigma}(\nabla\tilde{\phi} + \bar{\boldsymbol{E}})] \cdot \boldsymbol{n} &= \alpha\Delta_s(\tilde{\phi} + \bar{\boldsymbol{E}} \cdot \boldsymbol{x}), && \text{on } \Gamma. \end{aligned}\right\} \tag{2.3}$$

Here, $\boldsymbol{n}$ is the unit normal pointing into phase 2 and Δ_s is the Laplace–Beltrami operator on Γ, defined by

$$\Delta_s(\tilde{\phi} + \bar{\boldsymbol{E}} \cdot \boldsymbol{x}) = \delta_i\delta_i(\tilde{\phi} + \bar{\boldsymbol{E}} \cdot \boldsymbol{x}), \tag{2.4}$$

where δ_i is the tangential gradient on Γ,

$$\delta_i\psi = \psi_{,i} - (\boldsymbol{n} \cdot \nabla\psi)n_i. \tag{2.5}$$

The limiting cases $\alpha = 0$ and $\alpha = \infty$ of the tangential conductivity correspond to a perfectly bonded composite and a matrix containing perfectly conducting inclusions respectively. The variational principle for the effective conductivity tensor $\boldsymbol{\sigma}^{\mathrm{e}}$ is

$$\tfrac{1}{2}\bar{\boldsymbol{E}}\cdot\boldsymbol{\sigma}^{\mathrm{e}}\bar{\boldsymbol{E}} = \inf_{\phi\in V}\left\{\int_Q \tfrac{1}{2}(\nabla\phi+\bar{\boldsymbol{E}})\cdot\boldsymbol{\sigma}(\boldsymbol{x})(\nabla\phi+\bar{\boldsymbol{E}})\,\mathrm{d}\boldsymbol{x} + \tfrac{1}{2}\alpha\int_\Gamma |\delta(\phi+\bar{\boldsymbol{E}}\cdot\boldsymbol{x})|^2\,\mathrm{d}s\right\}, \tag{2.6}$$

where

$$V = \{\phi \in W^{1,2}(Q), \phi \text{ is } Q \text{ periodic}\}. \tag{2.7}$$

In the second problem considered, the interface is resistive and $\tilde{\phi}$ satisfies

$$\left.\begin{aligned} \operatorname{div}(\boldsymbol{\sigma}(\boldsymbol{x})(\nabla\tilde{\phi} + \bar{\boldsymbol{E}})) &= 0 && \text{in phases 1 and 2,} \\ [\boldsymbol{\sigma}(\boldsymbol{x})(\nabla\tilde{\phi} + \bar{\boldsymbol{E}})] \cdot \boldsymbol{n} &= 0 && \text{on } \Gamma, \\ \boldsymbol{\sigma}_2(\nabla\tilde{\phi} + \bar{\boldsymbol{E}}) \cdot \boldsymbol{n} &= -\beta[\phi] && \text{on } \Gamma. \end{aligned}\right\} \tag{2.8}$$

The parameter β^{-1} represents the barrier resistance and the two cases $\beta = \infty$ and $\beta = 0$ correspond to perfect contact and a perfectly insulating interface, respectively. Here, the variational principle is

$$\tfrac{1}{2}\bar{\boldsymbol{E}} \cdot \boldsymbol{\sigma}^{\mathrm{e}}\bar{\boldsymbol{E}} = \inf_{\phi\in W}\left\{\int_Q \tfrac{1}{2}(\nabla\phi + \bar{\boldsymbol{E}}) \cdot \boldsymbol{\sigma}(\boldsymbol{x})(\nabla\phi + \bar{\boldsymbol{E}})\,\mathrm{d}\boldsymbol{x} + \tfrac{1}{2}\beta\int_\Gamma ([\phi])^2\,\mathrm{d}s\right\}, \tag{2.9}$$

where W is the space of all Q-periodic square integrable functions such that $\nabla\phi$ is square integrable in each phase. For the nonlinear interface condition, $\tilde{\phi}$ satisfies

$$\left.\begin{aligned} \operatorname{div}(\boldsymbol{\sigma}(\boldsymbol{x})(\nabla\tilde{\phi} + \bar{\boldsymbol{E}})) &= 0 && \text{in phases 1 and 2,} \\ [\boldsymbol{\sigma}(\boldsymbol{x})(\nabla\tilde{\phi} + \bar{\boldsymbol{E}})] \cdot \boldsymbol{n} &= 0 && \text{on } \Gamma \end{aligned}\right\} \tag{2.10}$$

and

$$\left.\begin{aligned} \boldsymbol{\sigma}_2(\boldsymbol{x})(\nabla\tilde{\phi} + \bar{\boldsymbol{E}}) \cdot \boldsymbol{n} &= 0 && \text{on } \Gamma, && \text{where } -\phi_0 < -[\tilde{\phi}] < \phi_0, \\ \boldsymbol{\sigma}_2(\boldsymbol{x})(\nabla\tilde{\phi} + \bar{\boldsymbol{E}}) \cdot \boldsymbol{n} &\leqslant 0 && \text{on } \Gamma, && \text{where } -[\tilde{\phi}] = \phi_0, \\ \boldsymbol{\sigma}_2(\boldsymbol{x})(\nabla\tilde{\phi} + \bar{\boldsymbol{E}}) \cdot \boldsymbol{n} &\geqslant 0 && \text{on } \Gamma, && \text{where } -[\tilde{\phi}] = -\phi_0. \end{aligned}\right\} \tag{2.11}$$

Here, $\boldsymbol{\sigma}_2(\boldsymbol{x})(\nabla\tilde{\phi} + \bar{\boldsymbol{E}}) \cdot \boldsymbol{n}$ denotes the normal current on the matrix side of the interface. For this case, we have that the overall energy is the energy density given by

$$\tilde{W}(\bar{\boldsymbol{E}}) = \inf_{\phi \in W} \left\{ \int_Q \tfrac{1}{2}(\nabla\phi + \bar{\boldsymbol{E}}) \cdot \boldsymbol{\sigma}(\boldsymbol{x})(\nabla\phi + \bar{\boldsymbol{E}})\,\mathrm{d}\boldsymbol{x} + \int_\Gamma W_s(-[\phi])\,\mathrm{d}s \right\}, \tag{2.12}$$

where

$$W_s(-[\phi]) = \begin{cases} 0, & -\phi_0 < -[\tilde{\phi}] < \phi_0. \\ \infty, & -[\tilde{\phi}] \geqslant \phi_0, \\ \infty, & -[\tilde{\phi}] \leqslant -\phi_0. \end{cases} \tag{2.13}$$

Here, the potential $\tilde{\phi}$ is the minimizer. This is established in the appendix.

3. Bounds for a composite with a highly conducting interface

In this section upper and lower bounds on the energy are obtained starting from the variational principle (2.6). The first result that we present is a lower bound on the effective conductivity tensor for an anisotropic suspension of anisotropically conducting particles in an anisotropically conducting matrix. We introduce the 'scale' tensor $\mathcal{U}$ with components

$$\mathcal{U}_{ij} = \int_\Gamma \delta_k(\phi^i)\delta_k(\phi^j)\,\mathrm{d}s, \tag{3.1}$$

where the functions ϕ^j are the solutions of

$$\left.\begin{aligned} \Delta_s\phi^j &= -n_j, & \boldsymbol{x} &\in \Gamma_r, \\ \Delta\phi_j &= 0, & \boldsymbol{x} &\in \Omega_r, \end{aligned}\right\} \tag{3.2}$$

and n_j is the jth component of the outward normal $\boldsymbol{n}$. The 'scale' tensor $\mathcal{U}$ was introduced in Lipton (1999). Next we introduce the tensors $\boldsymbol{S}$ and $\boldsymbol{T}$ given by

$$\left.\begin{aligned} \boldsymbol{S} &= c_1\boldsymbol{I} - (\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_2)^{-1}(\bar{\boldsymbol{\sigma}} - \tilde{\boldsymbol{\sigma}}), \\ \boldsymbol{T} &= (\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_2)^{-1}(\bar{\boldsymbol{\sigma}} - \tilde{\boldsymbol{\sigma}})(\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_2)^{-1} + \alpha^{-1}\mathcal{U}. \end{aligned}\right\} \tag{3.3}$$

Here, $\tilde{\boldsymbol{\sigma}}$ is the effective conductivity of a perfectly bonded composite with the same microgeometry and $\bar{\boldsymbol{\sigma}} = c_1\boldsymbol{\sigma}_1 + c_2\boldsymbol{\sigma}_2$. The lower bound on the effective conductivity is given by the following result.

Theorem 3.1 (lower bound on the effective conductivity).

$$\bar{\boldsymbol{E}} \cdot \boldsymbol{\sigma}^{\mathrm{e}}\bar{\boldsymbol{E}} \geqslant \bar{\boldsymbol{E}} \cdot \{\tilde{\boldsymbol{\sigma}} + \boldsymbol{S}\boldsymbol{T}^{-1}\boldsymbol{S}\}\bar{\boldsymbol{E}}. \tag{3.4}$$

When the particles are spheres of possibly different radii $a_1, a_2, \dots, a_n$, we consider the 'volume-averaged radius' $\langle a \rangle$ given by

$$\langle a \rangle = \sum_{i=1}^{n} V(a_i)a_i,$$

where $c_1 \times V(a_i)$ is the volume fraction occupied by spheres of radius a_i. For this case, the scale tensor is $\mathcal{U} = c_1\frac{1}{2}\langle a \rangle\boldsymbol{I}$ (see Lipton 1999). When the conductivities of the particles and the matrix are isotropic and the particles form an isotropic monodisperse suspension of spheres of radius a, the scale tensor is $\mathcal{U} = c_1\frac{1}{2}a\boldsymbol{I}$ and the lower bound becomes as follows.

Theorem 3.2 (lower bound on the effective conductivity for isotropic suspension of spheres—highly conducting interface).

$$\sigma^{\mathrm{e}} \geqslant H(\tilde{\sigma}). \tag{3.5}$$

We now establish the lower bound. First note that, for any vector $\boldsymbol{v}$,

$$\tfrac{1}{2}\alpha^{-1}\boldsymbol{v}^2 = \sup_{\boldsymbol{w}}\{\boldsymbol{v}\cdot\boldsymbol{w} - \tfrac{1}{2}\alpha\boldsymbol{w}^2\}, \tag{3.6}$$

and so

$$\tfrac{1}{2}\alpha|\boldsymbol{w}|^2 \geqslant \boldsymbol{v}\cdot\boldsymbol{w} - \tfrac{1}{2}\alpha^{-1}|\boldsymbol{v}|^2. \tag{3.7}$$

We apply this estimate to bound the integrand of the surface integral in (2.6) to obtain

$$\tfrac{1}{2}\bar{\boldsymbol{E}}\cdot\boldsymbol{\sigma}^{\mathrm{e}}\bar{\boldsymbol{E}} \geqslant \inf_{\phi\in V}\left\{\int_Q \tfrac{1}{2}(\nabla\phi+\bar{\boldsymbol{E}})\cdot\boldsymbol{\sigma}(\boldsymbol{x})(\nabla\phi+\bar{\boldsymbol{E}})\,\mathrm{d}\boldsymbol{x} + \int_\Gamma[\boldsymbol{v}\cdot\delta(\phi+\bar{\boldsymbol{E}}\cdot\boldsymbol{x}) - \tfrac{1}{2}\alpha^{-1}|\boldsymbol{v}|^2]\,\mathrm{d}s\right\}. \tag{3.8}$$

Next, choose $\boldsymbol{v} = \delta(\phi^j\eta_j)$ on the surface of each particle, where η_j are the components of a constant vector $\boldsymbol{\eta}$. Then

$$\begin{aligned}\int_{\Gamma_r}\boldsymbol{v}\cdot\delta(\phi+\bar{\boldsymbol{E}}\cdot\boldsymbol{x})\,\mathrm{d}s &= \int_{\Gamma_r}\delta(\phi^j\eta_j)\cdot\delta(\phi+\bar{\boldsymbol{E}}\cdot\boldsymbol{x})\,\mathrm{d}s\\ &= -\int_{\Gamma_r}(\phi+\bar{\boldsymbol{E}}\cdot\boldsymbol{x})\Delta_s(\phi^j\eta_j)\,\mathrm{d}s\\ &= \int_{\Omega_r}\boldsymbol{\eta}\cdot(\nabla\phi+\bar{\boldsymbol{E}})\,\mathrm{d}\boldsymbol{x},\end{aligned} \tag{3.9}$$

after using (3.2) and the divergence theorem. It now follows that

$$\tfrac{1}{2}\bar{\boldsymbol{E}}\cdot\boldsymbol{\sigma}^{\mathrm{e}}\bar{\boldsymbol{E}} \geqslant \inf_{\phi\in V}\int_Q\{\tfrac{1}{2}(\nabla\phi+\bar{\boldsymbol{E}})\cdot\boldsymbol{\sigma}(\boldsymbol{x})(\nabla\phi+\bar{\boldsymbol{E}}) + f_1(\boldsymbol{x})\boldsymbol{\eta}\cdot(\nabla\phi+\bar{\boldsymbol{E}})\}\,\mathrm{d}\boldsymbol{x} - \tfrac{1}{2}\alpha^{-1}\boldsymbol{\eta}\cdot\mathcal{U}\boldsymbol{\eta}. \tag{3.10}$$

The problem of finding the infimum in (3.10) is analogous to finding the energy of a linear thermoelastic composite. The solution given by Talbot & Willis (1992) is given, for completeness, in the appendix and leads to the bound

$$\begin{aligned}\tfrac{1}{2}\bar{\boldsymbol{E}}\cdot\boldsymbol{\sigma}^{\mathrm{e}}\bar{\boldsymbol{E}} \geqslant{}& \tfrac{1}{2}\bar{\boldsymbol{E}}\cdot\bar{\boldsymbol{\sigma}}\bar{\boldsymbol{E}} + c_1\boldsymbol{\eta}\cdot\bar{\boldsymbol{E}}\\ &- \tfrac{1}{2}[\bar{\boldsymbol{E}} + (\boldsymbol{\sigma}_1-\boldsymbol{\sigma}_2)^{-1}\boldsymbol{\eta}]\cdot(\bar{\boldsymbol{\sigma}}-\tilde{\boldsymbol{\sigma}})[\bar{\boldsymbol{E}} + (\boldsymbol{\sigma}_1-\boldsymbol{\sigma}_2)^{-1}\boldsymbol{\eta}] - \tfrac{1}{2}\alpha^{-1}\boldsymbol{\eta}\cdot\mathcal{U}\boldsymbol{\eta}.\end{aligned} \tag{3.11}$$

Maximizing the right-hand side of (3.11) with respect to $\boldsymbol{\eta}$ gives (3.4), and the lower bound is established.

Next we present a new upper bound on the effective conductivity tensor for anisotropic suspensions of particles with anisotropic conductivity in a matrix with anisotropic conductivity. We introduce the surface energy tensor $\mathcal{G}$ defined by

$$\mathcal{G} = -\int_\Gamma \mathcal{H}\boldsymbol{n}\otimes\boldsymbol{x}\,\mathrm{d}s, \tag{3.12}$$

where $\mathcal{H} = -\operatorname{div}\boldsymbol{n}$ is the mean curvature. The effective conductivity of a composite with perfectly conducting inclusions, with the same microgeometry, is given by $\boldsymbol{\sigma}^{\infty}$. The upper bound is given by the following result.

Theorem 3.3 (upper bound on the effective conductivity).

$$\bar{\boldsymbol{E}} \cdot \boldsymbol{\sigma}^{\mathrm{e}} \bar{\boldsymbol{E}} \leqslant \bar{\boldsymbol{E}} \cdot \boldsymbol{U} \bar{\boldsymbol{E}}, \tag{3.13}$$

where

$$\boldsymbol{U} = \{\boldsymbol{\sigma}^{\infty} - (\boldsymbol{\sigma}^{\infty} - \boldsymbol{\sigma}_2)[\boldsymbol{\sigma}^{\infty} - \boldsymbol{\sigma}_2 + c_1(\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_2) + \alpha\mathcal{G}]^{-1}(\boldsymbol{\sigma}^{\infty} - \boldsymbol{\sigma}_2)\}. \tag{3.14}$$

One readily checks that the tensor

$$[\boldsymbol{\sigma}^{\infty} - \boldsymbol{\sigma}_2 + c_1(\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_2) + \alpha\mathcal{G}]^{-1}$$

is positive definite. For isotropic composites with isotropically conducting inclusions and matrix, equation (3.13) recovers the upper bound given by (5.10) in Lipton (1997*a*). To obtain the upper bound, we relax the infimum in (2.6) and allow for the substitution of any admissible trial field into the integrals. In the rth particle, choose

$$\phi + (\bar{\boldsymbol{E}} - \boldsymbol{\eta}) \cdot \boldsymbol{x} = C_r, \tag{3.15}$$

where C_r is a constant and $\boldsymbol{\eta}$ is a constant vector. Then, as $[\phi] = 0$ on $\varGamma$, the trial field in the matrix satisfies

$$\phi + (\bar{\boldsymbol{E}} - \boldsymbol{\eta}) \cdot \boldsymbol{x} = C_r \tag{3.16}$$

on the surface of the rth particle. Hence ϕ in the matrix is the fluctuating part of the field in a matrix containing a population of perfectly conducting particles, subject to an applied field $\bar{\boldsymbol{E}} - \boldsymbol{\eta}$. Now, by writing $\nabla\phi + \bar{\boldsymbol{E}} = \nabla\phi + (\bar{\boldsymbol{E}} - \boldsymbol{\eta}) + \boldsymbol{\eta}$, it is easy to show that

$$\begin{aligned}\int_Q f_2(\boldsymbol{x})(\nabla\phi + \bar{\boldsymbol{E}}) \cdot \boldsymbol{\sigma}_2(\nabla\phi + \bar{\boldsymbol{E}})\,\mathrm{d}\boldsymbol{x} \\ = \int_Q f_2(\boldsymbol{x})(\nabla\phi + \bar{\boldsymbol{E}} - \boldsymbol{\eta}) \cdot \boldsymbol{\sigma}_2(\nabla\phi + \bar{\boldsymbol{E}} - \boldsymbol{\eta})\,\mathrm{d}\boldsymbol{x} + 2\boldsymbol{\eta} \cdot \boldsymbol{\sigma}_2(\bar{\boldsymbol{E}} - \boldsymbol{\eta}) + c_2\boldsymbol{\eta} \cdot \boldsymbol{\sigma}_2\boldsymbol{\eta}.\end{aligned} \tag{3.17}$$

The energy in the particles is easily calculated and the surface integral is dealt with by noting that, on $\varGamma$, $\Delta_s(\boldsymbol{\eta} \cdot \boldsymbol{x}) = \mathcal{H}\boldsymbol{\eta} \cdot \boldsymbol{n}$. Then, using (3.15),

$$\begin{aligned}\int_{\varGamma_r} |\delta(\phi + \bar{\boldsymbol{E}} \cdot \boldsymbol{x})|^2\,\mathrm{d}s &= -\int_{\varGamma_r} \Delta_s(\phi + \bar{\boldsymbol{E}} \cdot \boldsymbol{x})(\phi + \bar{\boldsymbol{E}} \cdot \boldsymbol{x})\,\mathrm{d}s \\ &= -\int_{\varGamma_r} \mathcal{H}(\boldsymbol{\eta} \cdot \boldsymbol{n})(\boldsymbol{\eta} \cdot \boldsymbol{x})\,\mathrm{d}s.\end{aligned} \tag{3.18}$$

Using (3.17) and taking the infimum over fields satisfying (3.15) gives the bound

$$\begin{aligned}\tfrac{1}{2}\bar{\boldsymbol{E}} \cdot \boldsymbol{\sigma}^{\mathrm{e}}\bar{\boldsymbol{E}} \leqslant \inf_{\phi} \int_Q \tfrac{1}{2} f_2(\boldsymbol{x})(\nabla\phi + \bar{\boldsymbol{E}} - \boldsymbol{\eta}) \cdot \boldsymbol{\sigma}_2(\nabla\phi + \bar{\boldsymbol{E}} - \boldsymbol{\eta})\,\mathrm{d}\boldsymbol{x} \\ + \boldsymbol{\eta} \cdot \boldsymbol{\sigma}_2(\bar{\boldsymbol{E}} - \boldsymbol{\eta}) + \tfrac{1}{2}\boldsymbol{\eta} \cdot \bar{\boldsymbol{\sigma}}\boldsymbol{\eta} + \tfrac{1}{2}\alpha\boldsymbol{\eta} \cdot \mathcal{G}\boldsymbol{\eta}.\end{aligned} \tag{3.19}$$

The first term in (3.19) is the energy of a matrix containing a distribution of perfectly conducting inclusions and, after minimizing with respect to $\boldsymbol{\eta}$, the bound is finally obtained.

4. Bounds for a composite with a resistive interface

We start by presenting a new upper bound for the effective conductivity for particle reinforced conductors with resistive interface. We introduce the surface moment of inertia tensor defined by

$$\mathcal{M} = \sum_{r=1}^{N} \int_{\Gamma_r} (\boldsymbol{x} - \boldsymbol{x}^r) \otimes (\boldsymbol{x} - \boldsymbol{x}^r)\, \mathrm{d}s, \tag{4.1}$$

where $\boldsymbol{x}^r$ is a reference point inside the rth particle. We introduce the tensors $\boldsymbol{N}$ and $\boldsymbol{M}$ given by

$$\left.\begin{aligned} \boldsymbol{N} &= c_1\boldsymbol{\sigma}_1 - (\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_2)^{-1}\boldsymbol{\sigma}_1(\bar{\boldsymbol{\sigma}} - \tilde{\boldsymbol{\sigma}}), \\ \boldsymbol{M} &= -\boldsymbol{\sigma}_1(\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_2)^{-1}(\bar{\boldsymbol{\sigma}} - \tilde{\boldsymbol{\sigma}})(\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_2)^{-1}\boldsymbol{\sigma}_1 + (c_1\boldsymbol{\sigma}_1 + \beta\mathcal{M}). \end{aligned}\right\} \tag{4.2}$$

The upper bound is given by the following result.

Theorem 4.1 (upper bound for particulate composites with resistive interface).

$$\bar{\boldsymbol{E}} \cdot \boldsymbol{\sigma}^{\mathrm{e}} \bar{\boldsymbol{E}} \leqslant \bar{\boldsymbol{E}} \cdot \{\tilde{\boldsymbol{\sigma}} - \boldsymbol{N}\boldsymbol{M}^{-1}\boldsymbol{N}\}\bar{\boldsymbol{E}}. \tag{4.3}$$

One easily checks that the tensor $\boldsymbol{M}$ is positive definite. For a mono-disperse suspension of spheres of radius 'a', calculation gives $\mathcal{M} = ac_1\boldsymbol{I}$. When the composite is an isotropic suspension of isotropically conducting spheres in an isotropic matrix, the upper bound (4.3) becomes as follows.

Theorem 4.2 (upper bound on the effective conductivity for isotropic suspension of spheres—resistive interface).

$$\sigma^{\mathrm{e}} \leqslant C(\tilde{\sigma}). \tag{4.4}$$

The upper bound is found by choosing the trial field

$$\phi = \phi_{\mathrm{p}} + \phi', \tag{4.5}$$

where $\phi_{\mathrm{p}} \in V$ and

$$\phi' = \begin{cases} \boldsymbol{\eta} \cdot (\boldsymbol{x} - \boldsymbol{x}^r) & \text{in } r\text{th particle}, \\ 0 & \text{in the matrix}, \end{cases} \tag{4.6}$$

where $\boldsymbol{\eta}$ is a constant vector. Substituting this into the integrals in (2.9) leads to

$$\tfrac{1}{2}\bar{\boldsymbol{E}} \cdot \boldsymbol{\sigma}^{\mathrm{e}} \bar{\boldsymbol{E}} \leqslant \int_Q \{\tfrac{1}{2}(\nabla\phi_{\mathrm{p}} + \bar{\boldsymbol{E}}) \cdot \boldsymbol{\sigma}(\boldsymbol{x})(\nabla\phi_{\mathrm{p}} + \bar{\boldsymbol{E}}) + f_1(\boldsymbol{x})\boldsymbol{\eta} \cdot \boldsymbol{\sigma}_1(\nabla\phi_{\mathrm{p}} + \bar{\boldsymbol{E}})\}\, \mathrm{d}\boldsymbol{x} + \tfrac{1}{2}\boldsymbol{\eta} \cdot (c_1\boldsymbol{\sigma}_1 + \beta\mathcal{M})\boldsymbol{\eta}. \tag{4.7}$$

Next, take the infimum over ϕ_{p} in (4.7) and use the result in the appendix to get

$$\begin{aligned} \tfrac{1}{2}\bar{\boldsymbol{E}} \cdot \boldsymbol{\sigma}^{\mathrm{e}} \bar{\boldsymbol{E}} \leqslant{}& \tfrac{1}{2}\bar{\boldsymbol{E}} \cdot \bar{\boldsymbol{\sigma}}\bar{\boldsymbol{E}} + c_1\boldsymbol{\eta} \cdot \boldsymbol{\sigma}_1\bar{\boldsymbol{E}} \\ &- \tfrac{1}{2}(\bar{\boldsymbol{E}} + (\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_2)^{-1}\boldsymbol{\sigma}_1\boldsymbol{\eta}) \cdot (\bar{\boldsymbol{\sigma}} - \tilde{\boldsymbol{\sigma}})(\bar{\boldsymbol{E}} + (\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_2)^{-1}\boldsymbol{\sigma}_1\boldsymbol{\eta}) \\ &+ \tfrac{1}{2}\boldsymbol{\eta} \cdot (c_1\boldsymbol{\sigma}_1 + \beta\mathcal{M})\boldsymbol{\eta}, \end{aligned} \tag{4.8}$$

where $\tilde{\boldsymbol{\sigma}}$ is the effective conductivity tensor of the perfectly bonded composite. Finally, the right-hand side of (4.8) is minimized with respect to $\boldsymbol{\eta}$ to obtain the upper bound.

We present a lower bound for particle reinforced composites with resistive interface. We introduce the tensor $\mathcal{B}$ defined by

$$\mathcal{B} = \int_{\Gamma} \boldsymbol{n} \otimes \boldsymbol{n} \, \mathrm{d}s, \tag{4.9}$$

and the tensors $\boldsymbol{R}$ and $\boldsymbol{Q}$ given by

$$\left.\begin{aligned} \boldsymbol{R} &= (\boldsymbol{\sigma}_2 - \boldsymbol{\sigma}^0)\boldsymbol{\sigma}_2^{-1}, \\ \boldsymbol{Q} &= \boldsymbol{\sigma}_2^{-1}(c_2\boldsymbol{\sigma}_2 - \boldsymbol{\sigma}^0)\boldsymbol{\sigma}_2^{-1} + c_1\boldsymbol{\sigma}_1^{-1} + \beta^{-1}\mathcal{B}. \end{aligned}\right\} \tag{4.10}$$

The lower bound is given by the following result.

Theorem 4.3 (lower bound for particulate composites with resistive interface).

$$\bar{\boldsymbol{E}} \cdot \boldsymbol{\sigma}^{\mathrm{e}} \bar{\boldsymbol{E}} \geqslant \bar{\boldsymbol{E}} \cdot \{\boldsymbol{\sigma}^0 + \boldsymbol{R}\boldsymbol{Q}^{-1}\boldsymbol{R}\}\bar{\boldsymbol{E}}. \tag{4.11}$$

A straightforward calculation shows that the tensor

$$(c_2\boldsymbol{\sigma}_2 - \boldsymbol{\sigma}^0)$$

is positive definite. For isotropic composites with isotropically conducting inclusions and matrix, equation (4.11) is precisely the lower bound given by (3.13) in Lipton & Vernescu (1996).

To obtain a lower bound, we follow Lipton & Vernescu (1996) and complete the square to find that

$$\tfrac{1}{2}\beta \int_{\Gamma} ([\phi])^2 \, \mathrm{d}s \geqslant \int_{\Gamma} v[\phi] \, \mathrm{d}s - \frac{1}{2\beta} \int_{\Gamma} v^2 \, \mathrm{d}s, \tag{4.12}$$

for any scalar v. Now introduce a comparison composite with conductivity tensor

$$\hat{\boldsymbol{\sigma}}(\boldsymbol{x}) = f_1(\boldsymbol{x})\boldsymbol{\sigma}_0 + f_2(\boldsymbol{x})\boldsymbol{\sigma}_2, \tag{4.13}$$

where $\boldsymbol{\sigma}_0$ is chosen so that $\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_0$ is positive definite. Then

$$\tfrac{1}{2}\boldsymbol{P} \cdot (\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_0)^{-1}\boldsymbol{P} = \sup_{\boldsymbol{E}} \{\boldsymbol{P} \cdot \boldsymbol{E} - \tfrac{1}{2}\boldsymbol{E} \cdot (\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_0)\boldsymbol{E}\}, \tag{4.14}$$

for any vector $\boldsymbol{P}$, and so

$$\tfrac{1}{2}\boldsymbol{E} \cdot \boldsymbol{\sigma}_1\boldsymbol{E} \geqslant \tfrac{1}{2}\boldsymbol{E} \cdot \boldsymbol{\sigma}_0\boldsymbol{E} + \boldsymbol{P} \cdot \boldsymbol{E} - \tfrac{1}{2}\boldsymbol{P} \cdot (\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_0)^{-1}\boldsymbol{P}, \tag{4.15}$$

for any vectors $\boldsymbol{P}$ and $\boldsymbol{E}$. Using (4.12) and (4.15) in (2.9) gives

$$\begin{aligned} \tfrac{1}{2}\bar{\boldsymbol{E}} \cdot \boldsymbol{\sigma}^{\mathrm{e}}\bar{\boldsymbol{E}} \geqslant \inf_{\phi \in W} \bigg\{ &\int_Q \{\tfrac{1}{2}(\nabla\phi + \bar{\boldsymbol{E}}) \cdot \hat{\boldsymbol{\sigma}}(\boldsymbol{x})(\nabla\phi + \bar{\boldsymbol{E}}) + f_1(\boldsymbol{x})\boldsymbol{P} \cdot (\nabla\phi + \bar{\boldsymbol{E}})\} \, \mathrm{d}\boldsymbol{x} \\ &- \int_Q f_1(\boldsymbol{x})\tfrac{1}{2}\boldsymbol{P} \cdot (\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_0)^{-1}\boldsymbol{P} \, \mathrm{d}\boldsymbol{x} + \int_{\Gamma} v[\phi] \, \mathrm{d}s - \frac{1}{2\beta}\int_{\Gamma} v^2 \, \mathrm{d}s \bigg\}, \end{aligned} \tag{4.16}$$

for any vector $\boldsymbol{P}$ and scalar v. Next, let ϕ^* be the minimizer of

$$\inf_{\phi\in V}\int_Q\{\tfrac{1}{2}(\nabla\phi+\bar{\boldsymbol{E}})\cdot\hat{\boldsymbol{\sigma}}(\boldsymbol{x})(\nabla\phi+\bar{\boldsymbol{E}})+f_1(\boldsymbol{x})\boldsymbol{P}\cdot(\nabla\phi+\bar{\boldsymbol{E}})\}\,\mathrm{d}\boldsymbol{x}, \tag{4.17}$$

so that ϕ^* is continuous. It follows that ϕ^* satisfies

$$\operatorname{div}[\hat{\boldsymbol{\sigma}}(\boldsymbol{x})(\nabla\phi^*+f_1(\boldsymbol{x})\boldsymbol{P})]=0,\quad \boldsymbol{x}\in Q, \tag{4.18}$$

and

$$\boldsymbol{\sigma}_2(\nabla\phi^*+\bar{\boldsymbol{E}})\cdot\boldsymbol{n}=(\boldsymbol{\sigma}_0(\nabla\phi^*+\bar{\boldsymbol{E}})+\boldsymbol{P})\cdot\boldsymbol{n},\quad \boldsymbol{x}\in\Gamma. \tag{4.19}$$

For any $\phi\in W$, let

$$\phi=\phi^*+\phi', \tag{4.20}$$

where $\phi'=\phi-\phi^*$. Then

$$\begin{aligned}\int_Q\{\tfrac{1}{2}(\nabla\phi+\bar{\boldsymbol{E}})\cdot\hat{\boldsymbol{\sigma}}(\boldsymbol{x})(\nabla\phi+\bar{\boldsymbol{E}})+f_1(\boldsymbol{x})\boldsymbol{P}\cdot(\nabla\phi+\bar{\boldsymbol{E}})\}\,\mathrm{d}\boldsymbol{x}&\\ \geqslant\int_Q\{\tfrac{1}{2}(\nabla\phi^*+\bar{\boldsymbol{E}})\cdot\hat{\boldsymbol{\sigma}}(\boldsymbol{x})(\nabla\phi^*+\bar{\boldsymbol{E}})+f_1(\boldsymbol{x})\boldsymbol{P}\cdot(\nabla\phi^*+\bar{\boldsymbol{E}})\}\,\mathrm{d}\boldsymbol{x}&\\ +\int_\Gamma[\phi']\{\boldsymbol{\sigma}_0(\nabla\phi^*+\bar{\boldsymbol{E}})+\boldsymbol{P}\}\cdot\boldsymbol{n}\,\mathrm{d}s,&\end{aligned} \tag{4.21}$$

on using the divergence theorem, equations (4.18), (4.19) and omitting the term involving $\nabla\phi'\cdot\hat{\boldsymbol{\sigma}}(\boldsymbol{x})\nabla\phi'$. It now follows that, for any $\phi\in W$,

$$\begin{aligned}\int_Q\{\tfrac{1}{2}(\nabla\phi+\bar{\boldsymbol{E}})\cdot\hat{\boldsymbol{\sigma}}(\boldsymbol{x})(\nabla\phi+\bar{\boldsymbol{E}})+f_1(\boldsymbol{x})\boldsymbol{P}\cdot(\nabla\phi+\bar{\boldsymbol{E}})\}\,\mathrm{d}\boldsymbol{x}+\int_\Gamma v[\phi]\,\mathrm{d}s&\\ \geqslant\int_Q\{\tfrac{1}{2}(\nabla\phi^*+\bar{\boldsymbol{E}})\cdot\hat{\boldsymbol{\sigma}}(\boldsymbol{x})(\nabla\phi^*+\bar{\boldsymbol{E}})+f_1(\boldsymbol{x})\boldsymbol{P}\cdot(\nabla\phi^*+\bar{\boldsymbol{E}})\}\,\mathrm{d}\boldsymbol{x}&\\ +\int_\Gamma[\phi']\{\boldsymbol{\sigma}_0(\nabla\phi^*+\bar{\boldsymbol{E}})\cdot\boldsymbol{n}+\boldsymbol{P}\cdot\boldsymbol{n}+v\}\,\mathrm{d}s.&\end{aligned} \tag{4.22}$$

If $\boldsymbol{\sigma}_0$ and v are now chosen to be zero and $-\boldsymbol{P}\cdot\boldsymbol{n}$, respectively, the right-hand side of (4.22) is independent of ϕ' and, from (4.16), we obtain the bound

$$\begin{aligned}\tfrac{1}{2}\bar{\boldsymbol{E}}\cdot\boldsymbol{\sigma}^{\mathrm{e}}\bar{\boldsymbol{E}}\geqslant\inf_{\phi\in V}\int_Q\{\tfrac{1}{2}(\nabla\phi+\bar{\boldsymbol{E}})\cdot\hat{\boldsymbol{\sigma}}(\boldsymbol{x})(\nabla\phi+\bar{\boldsymbol{E}})+f_1(\boldsymbol{x})\boldsymbol{P}\cdot(\nabla\phi+\bar{\boldsymbol{E}})\}\,\mathrm{d}\boldsymbol{x}&\\ -\frac{1}{2\beta}\boldsymbol{P}\cdot\mathcal{B}\boldsymbol{P}-\tfrac{1}{2}c_1\boldsymbol{P}\cdot\boldsymbol{\sigma}_1^{-1}\boldsymbol{P},&\end{aligned} \tag{4.23}$$

in which $\boldsymbol{P}$ has now been chosen to be a constant vector. Using the result in the appendix, the first term on the right-hand side of (4.23) can be expressed in terms of $\boldsymbol{\sigma}^0$, the effective conductivity tensor of a matrix containing a distribution of perfectly insulating particles. The result is

$$\begin{aligned}\tfrac{1}{2}\bar{\boldsymbol{E}}\cdot\boldsymbol{\sigma}^{\mathrm{e}}\bar{\boldsymbol{E}}\geqslant\tfrac{1}{2}c_2\bar{\boldsymbol{E}}\cdot\boldsymbol{\sigma}_2\bar{\boldsymbol{E}}+c_1\boldsymbol{P}\cdot\bar{\boldsymbol{E}}-\tfrac{1}{2}(\bar{\boldsymbol{E}}-\boldsymbol{\sigma}_2^{-1}\boldsymbol{P})\cdot(c_2\boldsymbol{\sigma}_2-\boldsymbol{\sigma}^0)(\bar{\boldsymbol{E}}-\boldsymbol{\sigma}_2^{-1}\boldsymbol{P})&\\ -\tfrac{1}{2}c_1\boldsymbol{P}\cdot\boldsymbol{\sigma}_1^{-1}\boldsymbol{P}-\frac{1}{2\beta}\boldsymbol{P}\cdot\mathcal{B}\boldsymbol{P}&\end{aligned} \tag{4.24}$$

The bound follows after optimizing this with respect to $\boldsymbol{P}$.

5. Results and discussion

Results have been obtained for an isotropic distribution of spherical inclusions and for a distribution of aligned spheroids. The first composite for which results have been obtained consists of spheres of inclusion phase, with radius a, coated with a corona of matrix phase. These coated spheres are packed in Q to occupy 60% of its volume and any gaps are filled with matrix phase. The spheres are assumed to be distributed isotropically and both phases are isotropic with conductivities σ_1 and σ_2.

When the interface is highly conducting, it is easy to check that the upper bound (3.13) coincides with that of Lipton (1997*a*). The tensor $\mathcal{G}$ was given by Lipton (1997*a*) and is $(2/a)c_1\boldsymbol{I}$. The simplest bound is found using the trivial upper bound given by ∞ for $\boldsymbol{\sigma}^\infty$, while an improved bound can be found using the bound of Bruno (1991). The lower bound (3.5) relies on knowledge of any lower bound for $\tilde{\boldsymbol{\sigma}} = \tilde{\sigma}\boldsymbol{I}$, where $\tilde{\sigma}$ is the effective conductivity of the perfectly bonded composite. The simplest choice of lower bound is

$$\tilde{\sigma} \geqslant \left(\frac{c_1}{\sigma_1} + \frac{c_2}{\sigma_2}\right)^{-1}. \tag{5.1}$$

An improved bound can be obtained by using the Hashin–Shtrikman lower bound. In this case, the lower bound of Lipton (1997*a*) given by (1.6) is recovered. The bound (3.5) does allow for the use of more sophisticated lower bounds and, for the composite considered here, can be further improved by using the lower bound of Bruno (1991) for $\tilde{\sigma}$. Bounds for the effective conductivity are displayed in figure 1 when $\sigma_1/\sigma_2 = 0.2$ for the volume fractions $c_1 = 0.1296$ and $c_2 = 0.4826$. It can be seen that use of the lower bound of Bruno (1991) improves on the bound of Lipton (1997*a*), given by (1.6) using the Hashin–Shtrikman bound. It should be noted, however, that the latter bound and the simplest upper and lower bounds are valid for all isotropic composites, whereas the bounds of Bruno incorporate details of the microstructure.

When the interface is resistive, it is easy to check that the lower bound (4.11) is the same as that given by Lipton & Vernescu (1996) (see (3.13) of that paper). A simple lower bound can be found by using the trivial lower bound on $\boldsymbol{\sigma}^0$ given by 0, while an improved bound is found by using the lower bound of Bruno (1991) for $\boldsymbol{\sigma}^0$. The tensor $\mathcal{B}$ in (4.9) was given by Lipton & Vernescu (1996) and is $(c_1/a)\boldsymbol{I}$. The upper bound (4.4) requires an upper bound on $\tilde{\boldsymbol{\sigma}}$. The simplest is found by using

$$\tilde{\sigma} \leqslant c_1\sigma_1 + c_2\sigma_2, \tag{5.2}$$

while improved bounds can be found by either using the Hashin–Shtrikman or Bruno upper bound. Here, the bound obtained using the Hashin–Shtrikman bound coincides with those obtained by Lipton & Vernescu (1996), as given by (1.2). By choosing the reference point $\boldsymbol{x}^r$ in (4.1) to be the centre of the rth sphere, the tensor $\mathcal{M}$ is found to be $c_1 a\boldsymbol{I}$. Results are displayed in figure 2 when $\sigma_1/\sigma_2 = 5$, and it can be seen that using the bound of Bruno (1991) improves on the upper bound of Lipton & Vernescu (1996), obtained here using the Hashin–Shtrikman bound. Bounds have also been obtained for a distribution of aligned isotropic spheroids with semiaxes $(1/a, 1/a, \varepsilon/a)$ with a highly conducting interface when $\varepsilon = \frac{1}{2}$. The tensor $\mathcal{G}$ is evaluated numerically, while the tensor $\mathcal{U}$ is estimated. For a particle of any shape, one has the bound on the scale tensor $\mathcal{U}$ derived in Lipton (1999). The bound is

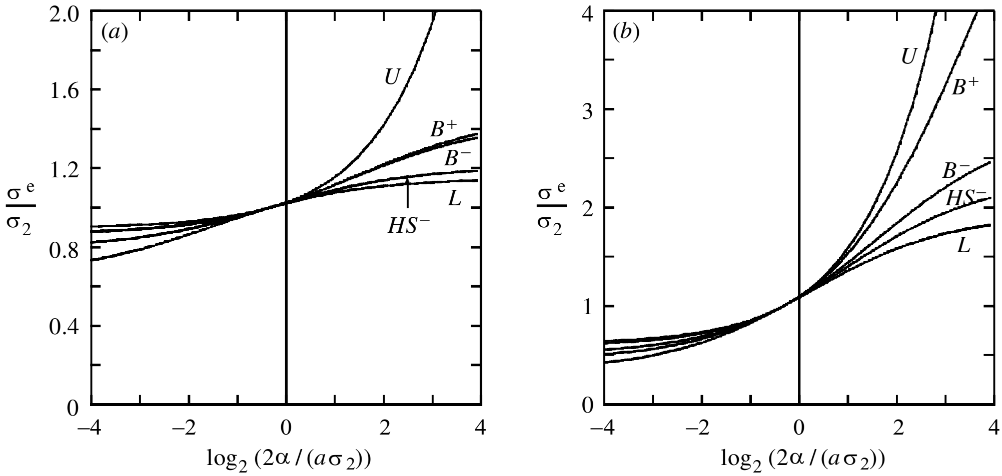

Figure 1. Bounds for a composite with a highly conducting interface: (*a*) $c_1 = 0.1296$; (*b*) $c_1 = 0.4826$. The curves are labelled as follows. U and L are the simple upper and lower bounds obtained by using infinity for σ^{∞} and (5.1). B^+ and B^- are the upper and lower bounds obtained by using the bounds of Bruno (1991) and HS^- is the lower bound obtained by using the Hashin–Shtrikman lower bound.

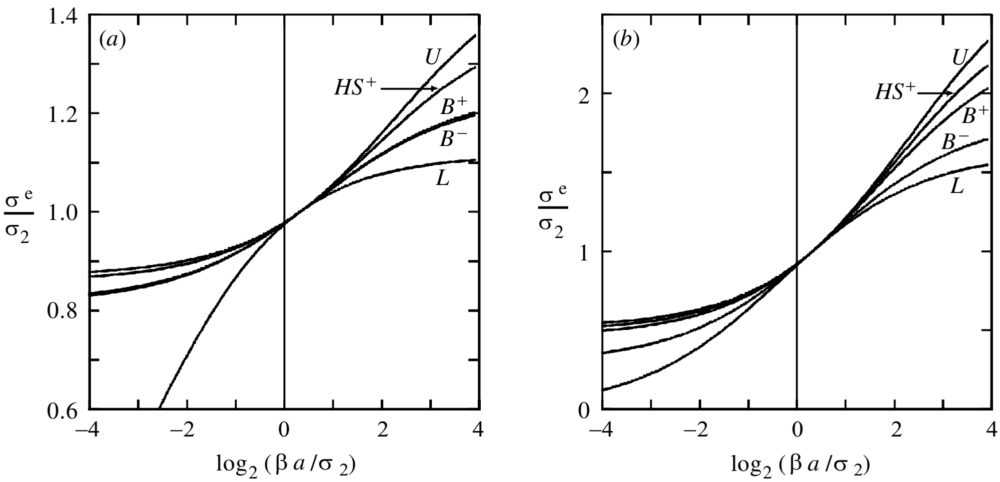

Figure 2. Bounds for a composite with a resistive interface: (*a*) $c_1 = 0.1296$; (*b*) $c_1 = 0.4826$. The curves are labelled as follows. U and L are the simple upper and lower bounds obtained by using (5.2) and zero for σ^0. B^+ and B^- are the upper and lower bounds obtained by using the bounds of Bruno (1991) and HS^+ is the upper bound obtained by using the Hashin–Shtrikman upper bound.

given by

$$\boldsymbol{r}\mathcal{U}\boldsymbol{r} \leqslant c_1(\sigma_1\beta)^{-1}|\boldsymbol{r}|^2, \tag{5.3}$$

where $\boldsymbol{r}$ is any vector and β is the surface to volume dissipation introduced by Lipton (1998). Equality is seen to hold for spheres. For spheroidal inclusions, a lower bound for $\sigma_1\beta$ can be calculated numerically from formulae given by Lipton (1998) for star-like particles. Results are shown in figure 3, when $\sigma_1/\sigma_2 = 0.2$ and $c_1 = 0.3$. The

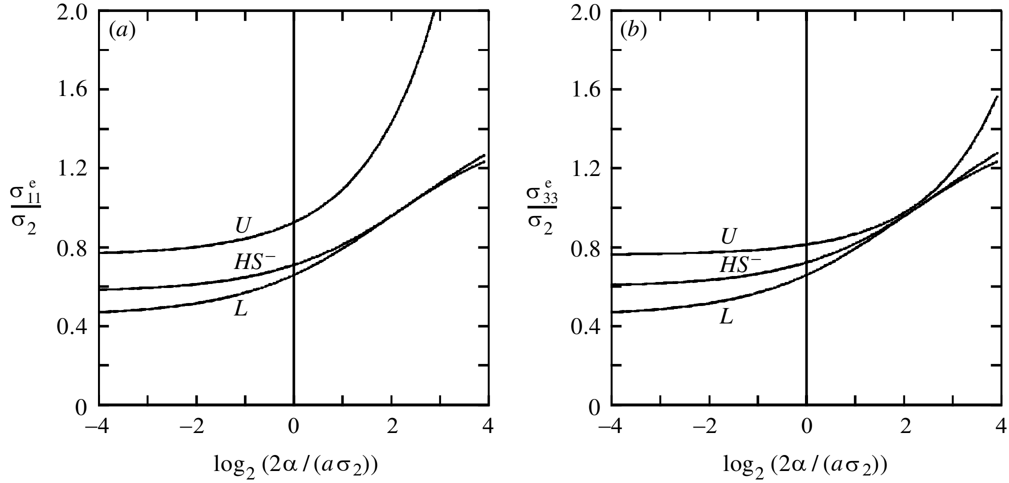

Figure 3. Bounds for a distribution of aligned ellipsoids with highly conducting interface. The curves are labelled as follows. U and L are the simple upper and lower bounds obtained by using infinity for σ^∞ and (5.1). HS^- is the lower bound obtained by using the Hashin–Shtrikman lower bound.

upper bound was found by substituting infinity for $\boldsymbol{\sigma}^\infty$ in (3.13), the simple lower bound was generated using the lower bound (5.1) and the Hashin–Shtrikman lower bound was found by using formulae given by Willis (1977). Some improvement is obtained by using the new lower bound (3.4).

6. A composite with a nonlinear interface

The problem defined by (2.12) is now considered. First, to obtain a lower bound, note that, for any x and y,

$$W_s(x) \geqslant xy - W_s^*(y), \tag{6.1}$$

where W_s^* is the convex dual of W_s, given by

$$W_s^*(y) = \phi_0|y|. \tag{6.2}$$

The same reasoning that led to (4.23) can now be used to obtain

$$\tilde{W}(\bar{\boldsymbol{E}}) \geqslant \inf_{\phi\in V}\int_Q \{\tfrac{1}{2}(\nabla\phi+\bar{\boldsymbol{E}})\cdot\hat{\boldsymbol{\sigma}}(\boldsymbol{x})(\nabla\phi+\bar{\boldsymbol{E}}) + f_1(\boldsymbol{x})\boldsymbol{P}\cdot(\nabla\phi+\bar{\boldsymbol{E}})\}\,\mathrm{d}\boldsymbol{x} \\ - \tfrac{1}{2}c_1\boldsymbol{P}\cdot\boldsymbol{\sigma}_1^{-1}\boldsymbol{P} - \phi_0\int_\Gamma |\boldsymbol{P}\cdot\boldsymbol{n}|\,\mathrm{d}s. \tag{6.3}$$

The infimum can be evaluated using the result in the appendix. For an isotropic distribution of isotropic spherical inclusions, the result, after maximizing with respect to $\boldsymbol{P}$, is

$$\frac{\tilde{W}(\bar{\boldsymbol{E}})}{W_2(\bar{\boldsymbol{E}})} \geqslant \begin{cases} \dfrac{\sigma^0}{\sigma_2} + \dfrac{(1-\sigma^0/\sigma_2-\frac{1}{2}\gamma)^2}{c_1\sigma_2/\sigma_1 + c_2 - \sigma^0/\sigma_2}, & \gamma < 2(1-\sigma^0/\sigma_2), \\ \dfrac{\sigma^0}{\sigma_2}, & \gamma \geqslant 2(1-\sigma^0/\sigma_2), \end{cases} \tag{6.4}$$

where $W_2(\boldsymbol{E}) = \frac{1}{2}\sigma_2|\boldsymbol{E}|^2$ and

$$\gamma = \frac{3c_1\phi_0}{a|\bar{\boldsymbol{E}}|}. \tag{6.5}$$

An upper bound is found by substituting the trial field given by (4.5) and (4.6) directly into the integral on the right-hand side of (2.12) and using the reasoning leading to (4.8) to get

$$\begin{aligned}\tilde{W}(\bar{\boldsymbol{E}}) \leqslant \tfrac{1}{2}\bar{\boldsymbol{E}}\cdot\bar{\boldsymbol{\sigma}}\bar{\boldsymbol{E}} + c_1\boldsymbol{\eta}\cdot\boldsymbol{\sigma}_1\bar{\boldsymbol{E}} + \tfrac{1}{2}c_1\boldsymbol{\eta}\cdot\boldsymbol{\sigma}_1\boldsymbol{\eta} \\ - \tfrac{1}{2}(\bar{\boldsymbol{E}} + (\boldsymbol{\sigma}_1-\boldsymbol{\sigma}_2)^{-1}\boldsymbol{\sigma}_1\boldsymbol{\eta})\cdot(\bar{\boldsymbol{\sigma}}-\tilde{\boldsymbol{\sigma}})(\bar{\boldsymbol{E}} + (\boldsymbol{\sigma}_1-\boldsymbol{\sigma}_2)^{-1}\boldsymbol{\sigma}_1\boldsymbol{\eta}),\end{aligned} \tag{6.6}$$

where $\boldsymbol{\eta}$ is subject to the restriction $|\boldsymbol{\eta}\cdot(\boldsymbol{x}-\boldsymbol{x}^r)| < \phi_0$ in the rth particle. After minimizing with respect to $\boldsymbol{\eta}$, the bound for an isotropic distribution of isotropic spheres is

$$\frac{\tilde{W}(\bar{\boldsymbol{E}})}{W_2(\bar{\boldsymbol{E}})} \leqslant \begin{cases} \dfrac{\tilde{\sigma}}{\sigma_2} + \dfrac{\sigma_1}{\sigma_2}\dfrac{c_1(\sigma_1-\sigma_2)^2 - \sigma_1(\bar{\sigma}-\tilde{\sigma})}{(\sigma_1-\sigma_2)^2}\dfrac{\gamma^2}{9c_1^2} - 2\dfrac{\sigma_1}{\sigma_2}\dfrac{\tilde{\sigma}-\sigma_2}{\sigma_1-\sigma_2}\dfrac{\gamma}{3c_1}, & \gamma < \gamma_0, \\ \dfrac{\tilde{\sigma}}{\sigma_2} - \dfrac{\sigma_1}{\sigma_2}\dfrac{(\tilde{\sigma}-\sigma_2)^2}{c_1(\sigma_1-\sigma_2)^2 - \sigma_1(\bar{\sigma}-\tilde{\sigma})}, & \gamma \geqslant \gamma_0, \end{cases} \tag{6.7}$$

where

$$\gamma_0 = \frac{3c_1(\sigma_1-\sigma_2)(\tilde{\sigma}-\sigma_2)}{c_1(\sigma_1-\sigma_2)^2 - \sigma_1(\bar{\sigma}-\tilde{\sigma})}. \tag{6.8}$$

We note that, as $\phi_0 \to \infty$, equation (6.4) implies that the lower bound tends to the effective conductivity of a matrix containing a distribution of perfect insulators, whereas, if $\phi_0 \to 0$, equation (6.7) implies that the upper bound tends to the effective conductivity of the perfectly bonded composite. It is also worth noting that the method used here would also apply for more general forms of W_s and can also be applied, in principle, to a highly conducting interface exhibiting nonlinear behaviour.

The simplest upper and lower bounds are found by using $\tilde{\sigma} = \bar{\sigma}$ and $\sigma^0 = 0$, respectively, in (6.7) and (6.4). Improved upper bounds are found by using either the Hashin–Shtrikman bound for $\tilde{\sigma}$ or the bound of Bruno (1991). The lower bound can be improved by using the lower bound of Bruno (1991) for σ^0. Bounds for $\tilde{W}/W_2$ are shown in figure 4 when $\sigma_1/\sigma_2 = 0.2$ for volume fractions of spheres of 0.1296 and 0.4826.

Appendix A.

In the present context, the result of Talbot & Willis (1992) is that, if

$$\tilde{W}_T(\bar{\boldsymbol{E}};\boldsymbol{P}) = \inf_{\phi\in V}\int_Q [\tfrac{1}{2}(\nabla\phi + \bar{\boldsymbol{E}})\cdot\boldsymbol{\sigma}(\boldsymbol{x})(\nabla\phi + \bar{\boldsymbol{E}}) + \boldsymbol{P}\cdot(\nabla\phi + \bar{\boldsymbol{E}})]\,\mathrm{d}\boldsymbol{x}, \tag{A 1}$$

where $\boldsymbol{P}$ is a piecewise constant vector, then, for two-phase materials,

$$\tilde{W}_T(\bar{\boldsymbol{E}};\boldsymbol{P}) = \tfrac{1}{2}\bar{\boldsymbol{E}}\cdot\bar{\boldsymbol{\sigma}}\bar{\boldsymbol{E}} + \bar{\boldsymbol{E}}\cdot\overline{\boldsymbol{P}} - S,$$

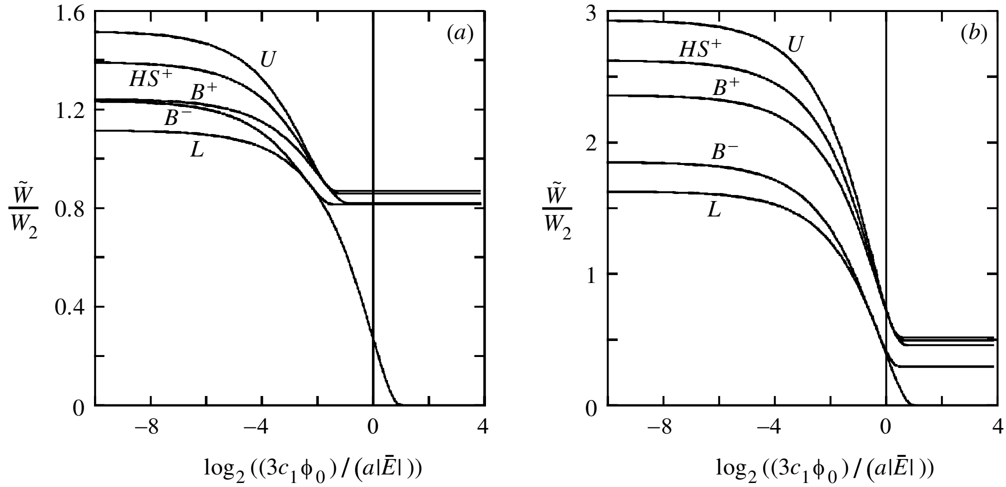

Figure 4. Bounds for a composite with a nonlinear interface: (*a*) $c_1 = 0.1296$; (*b*) $c_1 = 0.4826$. The curves are labelled as follows. U and L are the simple upper and lower bounds obtained by using (5.2) and zero for σ^0 in (6.7) and (6.4), respectively. B^+ and B^- are the upper and lower bounds obtained by using the bounds of Bruno (1991) and HS^+ is the upper bound obtained by using the Hashin–Shtrikman upper bound.

where

$$S = \tfrac{1}{2}[\bar{\boldsymbol{E}} + (\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_2)^{-1}(\boldsymbol{P}_1 - \boldsymbol{P}_2)] \cdot (\bar{\boldsymbol{\sigma}} - \tilde{\boldsymbol{\sigma}})[\bar{\boldsymbol{E}} + (\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_2)^{-1}(\boldsymbol{P}_1 - \boldsymbol{P}_2)], \tag{A 2}$$

where $\bar{\boldsymbol{P}} = c_1\boldsymbol{P}_1 + c_2\boldsymbol{P}_2$ and $\tilde{\boldsymbol{\sigma}}$ is the effective conductivity tensor of the composite.

Finally, we establish that the solution of (2.11) is the minimizer of (2.12). The overall energy is easily seen to be equivalent to

$$\tilde{W}(\bar{\boldsymbol{E}}) = \inf_{\phi \in K} \left\{ \int_Q \tfrac{1}{2}(\nabla\phi + \bar{\boldsymbol{E}}) \cdot \boldsymbol{\sigma}(\boldsymbol{x})(\nabla\phi + \bar{\boldsymbol{E}})\,\mathrm{d}\boldsymbol{x} \right\}, \tag{A 3}$$

where K is the closed convex subset of the Hilbert space defined by W in § 2 given by

$$K = \{\phi \text{ in } W \mid -\phi_0 \leqslant -[\phi] \leqslant \phi_0\}. \tag{A 4}$$

We set

$$F(\phi) = \int_Q \tfrac{1}{2}(\nabla\phi + \bar{\boldsymbol{E}}) \cdot \boldsymbol{\sigma}(\boldsymbol{x})(\nabla\phi + \bar{\boldsymbol{E}})\,\mathrm{d}\boldsymbol{x},$$

and well-known methods (see Duvaut & Lions 1976) show that the minimizer is characterized by the variational inequality

$$\int_Q F'(\phi) \cdot (\nabla\phi - \nabla v)\,\mathrm{d}x \geqslant 0, \tag{A 5}$$

for all v in K. This is easily seen to be equivalent to (2.11).

References

Bruno, O. P. 1991 The effective conductivity of strongly heterogeneous composites. *Proc. R. Soc. Lond.* A **433**, 353–381.

Duvaut, G. & Lions, J. L. 1976 *Inequalities in mechanics and physics*. Springer.

Garret, K. W. & Rosenberg, H. M. 1974 The thermal conductivity of epoxy-resin powder composite materials. *J. Phys.* D **7**, 1247–1258.

Hashin, Z. 1992 Extremum principles for elastic heterogeneous media with imperfect interfaces and their application to bounding of effective moduli. *J. Mech. Phys. Solids* **40**, 767–781.

Hashin, Z. & Shtrikman, S. 1962 A variational approach to the theory of the elastic behaviour of polycrystals. *J. Mech. Phys. Solids* **10**, 343–352.

Hasselman, D. P. H. & Donaldson, K. Y. 1992 Effect of reinforcement particle size on the thermal conductivity of a particulate-silicon carbide-reinforced aluminum matrix composite. *J. Am. Ceram. Soc.* **75**, 3137–3140.

Laws, N. 1973 On the thermostatics of composite materials. *J. Mech. Phys. Solids* **21**, 9–17.

Levin, V. M. 1967 Thermal expansion coefficients of heterogeneous materials. *Mekh. Tverd. Tela.* **2**, 88–94.

Lipton, R. 1997*a* Variational methods bounds and size effects for composites with highly conducting interface. *J. Mech. Phys. Solids* **45**, 361–384.

Lipton, R. 1997*b* Reciprocal relations bounds and size effects for composites with highly conducting interface. *SIAM J. Appl. Math.* **37**, 347–363.

Lipton, R. 1998 Influence of interfacial surface conduction on the DC electrical conductivity of particle reinforced composites. *Proc. R. Soc. Lond.* A **454**, 1371–1382.

Lipton, R. 1999 Variational methods, bounds and size effects for two-phase composites with coupled heat and mass transport processes at the two-phase interface. *J. Mech. Phys. Solids* **47**, 1699–1736.

Lipton, R. & Vernescu, B. 1995 Variational methods, size effects and extremal microgeometries for elastic composites with imperfect interface. *Math. Models Methods Appl. Sci.* **8**, 1139–1173.

Lipton, R. & Vernescu, B. 1996 Composites with imperfect interface. *Proc. R. Soc. Lond.* A **452**, 329–358.

Talbot, D. R. S. & Willis, J. R. 1992 Some simple explicit bounds for the overall behaviour of nonlinear composites. *Int. J. Solids Struct.* **29**, 1981–1987.

Willis, J. R. 1977 Bounds and self-consistent estimates for the overall properties of anisotropic composites. *J. Mech. Phys. Solids* **25**, 185–202.