The effect of the interface on the dc transport properties of nonlinear composite materials

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The effects of the interface separating two strongly nonlinear electric conductors is investigated. The interface may either be highly conducting or exhibit an electric contact resistance. Our analysis and results are based upon new variational principles for nonlinear composites with surface energies. For monodisperse suspensions of spheres separated from the matrix by a highly conducting interface, a critical direct-current (dc) applied field strength is found for which the electric potential inside the sample is the same as for a sample containing no spheres. For this field strength the overall electric current passing through the sample is the same as for a sample containing pure matrix conductor. When there is a contact resistance between the matrix and sphere phase, a critical dc applied current density is found for which the current density inside the sample is the same as for a sample containing no spheres. These results are shown to be independent of the location of the spheres within the sample. Moreover, this effect is independent of the concentration of spheres in the sample even beyond the onset of interface percolation.
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I. INTRODUCTION

Nonlinear direct-current (dc) electric conductivity is a property intrinsic to many ceramic materials.[1] More generally, nonlinear inhomogeneous materials are pervasive, appearing in applications ranging from transient voltage protection to the selective absorption of solar energy.[2] This article investigates the effect of the interface on the overall dc electric properties of nonlinear composite conductors. A composite made from a monodisperse suspension of spheres embedded in a matrix is considered. It is supposed that the interface separating the spheres and matrix is highly conducting. A critical applied voltage is found for which the electric potential inside the sample is the same as for a sample of identical shape containing no spheres whatsoever. At the critical voltage, the overall electric current passing through the sample is the same as in a homogeneous conductor. On the other hand, in the presence of an electric contact resistance at the interface, we show that there is a critical applied dc current density for which the current density inside the sample is the same as for a sample containing no spheres. The overall electric field in the sample corresponds to that associated with a homogeneous conductor made from matrix material. These effects are shown to be independent of the location of the spheres within the sample. Moreover, these effects are independent of the concentration of spheres in the sample even beyond the onset of interface percolation.

It is demonstrated from first principles that these phenomena occur for a wide range of nonlinear constitutive behavior. Indeed, it is shown here that cloaking phenomena can occur when the potential energy density of each phase is a convex function of the magnitude of the electric field. This requirement naturally includes local constitutive relations associated with nonlinear behavior of the form

$$\mathbf{j}=\gamma|\mathbf{E}|^{t}\mathbf{E},$$

where $\mathbf{j}$ is the local current density and $\mathbf{E}$ is the local electric field. Here the nonlinear susceptibility γ takes different values inside each phase.

For the case of a highly conducting interface, the electric conduction on the interfacial surface is linear and is described by the scalar α, with dimensions of conductivity $\times$length. Surface conduction along the two-phase interface has been shown to strongly influence the overall properties of random mixtures of ionic insulators, see Refs. 3–5. It naturally facilitates the transport of ions across a fluid saturated porous medium and has a strong effect on the overall properties.[6] Surface conduction is also shown to exert a significant influence on the overall dc electric conductivity of mortar, see Refs. 7 and 8.

The transmission of current across the interface is characterized by a jump in the current normal to the interface. The jump in the normal current produces an interfacial charge density. The associated electric potential is continuous across the interface and is coupled to the charge density through a Poisson equation supported on the interface [see Eq. (1)]. The interface transmission condition can be thought of as the limiting case of electric transport across bulk phases separated by a thin highly conducting, linear, interphase layer.[9]

On the other hand, contact resistance often appears due to the presence of a thin highly resistive layer or ‘‘interphase'' between two conducting phases. The effects of the thin layer can be modeled by a discontinuous potential that suffers a jump across the interface. The associated current density flowing into the interface is continuous across it and is proportional to the jump in electric potential. Here, the constant of proportionality is denoted by β and has dimensions given by conductivity per unit length. Both of these interface transmission conditions are distinct from the standard ''perfectly bonded'' interface conditions where both the electric potential and normal current density are continuous across the interface. We remark that contact resistance is not limited to electrostatic problems and can appear in the mathematically analogous context of heat conductivity. Here contact resistance can arise due to surface roughness[10] or from an acoustic mismatch between phases at liquid helium temperatures, see Ref. 11.

II. EQUILIBRIUM EQUATIONS AND VARIATIONAL PRINCIPLES FOR OVERALL PROPERTIES

We start by formulating the equations of equilibrium and associated variational principles for the case of a highly conducting interface between phases. The potential functions of the matrix and particle phases are written as, $W_m(\mathbf{E})$ and $W_p(\mathbf{E})$, respectively. On the two-phase interface the potential function is defined by $W_s(\mathbf{E}_{\tan}) \triangleq (\alpha/2)|\mathbf{E}_{\tan}|^2$. Here, $\mathbf{E}_{\tan}$ is the projection of the electric field onto the interfacial surface. We subject the composite to linear boundary conditions, i.e., given a constant vector $\overline{\mathbf{E}}$ the electric potential ϕ satisfies $\phi = -\overline{\mathbf{E}}\cdot\mathbf{x}$ on the boundary of the composite sample. The electric potential is continuous throughout the composite and the electric field is given by $\mathbf{E} = -\nabla\phi$. We denote the gradient of the potential W_m with respect to its argument by DW_m, and the constitutive relation in the matrix phase is written $\mathbf{j} = DW_m(\mathbf{E})$. Similarly the constitutive relation in the particle phase is given by $\mathbf{j} = DW_p(\mathbf{E})$. On the two-phase interface

$$\mathbf{E}_{\tan} = -(I - \mathbf{n}\otimes\mathbf{n})\nabla\phi = -\nabla_s\phi,$$

where ∇_s is the gradient operator on the interfacial surface and $\mathbf{n}$ is the normal vector on the interface directed into the matrix phase. The constitutive relation on the interface is given by $\mathbf{j}_s = \alpha\mathbf{E}_{\tan}$. The equilibrium equations are

$$\begin{aligned} &-\mathrm{div}[DW_m(\mathbf{E})] = 0, \quad \text{in the matrix,} \\ &-\mathrm{div}[DW_p(\mathbf{E})] = 0, \quad \text{in each particle,} \end{aligned} \tag{1}$$

and

$$-\alpha\Delta_s\phi = \mathbf{j}_p\cdot\mathbf{n} - \mathbf{j}_m\cdot\mathbf{n}, \quad \text{on the two phase interface.}$$

Here Δ_s denotes the surface Laplacian on the two phase interface and the subscripts indicate the side of the interface that the normal current is evaluated on. The potential energy function in the composite is written

$$W[\mathbf{E},\mathbf{x}] = \begin{cases} W_m(\mathbf{E}), & \mathbf{x} \text{ in the matrix,} \\ W_p(\mathbf{E}), & \mathbf{x} \text{ in the particles.} \end{cases} \tag{2}$$

The macroscopic (or overall) energy is defined by

$$\tilde{W}(\overline{\mathbf{E}}) = \inf_{\mathbf{E}\in K}|Q|^{-1} \times\left(\int_Q W[\mathbf{E}(\mathbf{x}),\mathbf{x}]dx + \int_\Gamma W_s(\mathbf{E}_{\tan})ds\right), \tag{3}$$

where Q denotes the sample domain and Γ represents the union of all two-phase interfaces. The admissible set of trial fields K is given by

$$K = \begin{cases} \mathbf{E}:\mathbf{E} = -\nabla\varphi, \\ \varphi = -\overline{\mathbf{E}}\cdot\mathbf{x}, \quad \text{on the sample boundary.} \end{cases} \tag{4}$$

Taking the first variation in Eq. (3) shows that the minimizer is a solution of the equilibrium equations given by Eq. (1). The overall current passing through the composite sample as measured by an outside observer is given by

$$\overline{\mathbf{j}} = \frac{1}{|Q|}\int_{\partial Q}\mathbf{j}\cdot\mathbf{n}\mathbf{x}\,ds. \tag{5}$$

The overall constitutive relation for the composite sample is given by

$$\overline{\mathbf{j}} = D\tilde{W}(\overline{\mathbf{E}}). \tag{6}$$

When there is a contact resistance between phases, the associated surface potential function is defined by $W_s([\phi]) \triangleq (\beta/2)([\phi])^2$. Here, $[\phi]$ is the jump in the potential across the interface. We denote the unit normal pointing out of the composite domain by $\mathbf{n}$ and inject an electric current into the composite, i.e., given a constant vector $\overline{\mathbf{j}}$ the electric current density $\mathbf{j}$ satisfies $\mathbf{j}\cdot\mathbf{n} = -\overline{\mathbf{j}}\cdot\mathbf{n}$ on the boundary of the composite sample. On the two-phase interface $[\mathbf{j}\cdot\mathbf{n}] = 0$, where $\mathbf{n}$ is the normal vector on the interface directed into the matrix phase. The equilibrium equations are

$$\begin{aligned} &-\mathrm{div}[DW_m(\mathbf{E})] = 0, \quad \text{in the matrix,} \\ &-\mathrm{div}[DW_p(\mathbf{E})] = 0, \quad \text{in each particle,} \end{aligned} \tag{7}$$

and

$$\mathbf{j}\cdot\mathbf{n} = -\beta[\phi], \quad \text{on the two phase interface.}$$

Here $[\phi] = \phi_m - \phi_p$, where the subscripts denote the side of the interface that the potential is evaluated on.

The overall electric field across the composite sample as measured by an outside observer is given by

$$\overline{\mathbf{E}} = \frac{-1}{|Q|}\int_{\partial Q}\phi\mathbf{n}\,ds. \tag{8}$$

The macroscopic (or overall) energy[12] is defined by

$$\tilde{C}(\overline{\mathbf{E}}) = \inf_{\varphi\in K_c}|Q|^{-1} \times\left(\int_Q W[\mathbf{E}(\mathbf{x}),\mathbf{x}]dx + \int_\Gamma W_s([\varphi])ds\right), \tag{9}$$

where the admissible set of trial fields K_c is given by

$$K_c = \begin{cases} \varphi:\varphi, \text{ is differentiable in the particles and in the matrix,} \\ \overline{\mathbf{E}} = \dfrac{-1}{|Q|}\displaystyle\int_{\partial Q}\varphi\mathbf{n}\,ds. \end{cases} \tag{10}$$

Taking the first variation in Eq. (9) shows that the minimizer is a solution of the equilibrium equations given by Eq. (7). A straight forward computation shows that the average current density is related to the overall electric field through the relation $\overline{\mathbf{j}}=D\tilde{C}(\overline{\mathbf{E}})$. Denoting the convex dual of the macroscopic energy by $\tilde{C}^*$ one has the equivalent overall constitutive relation for the composite sample given by

$$\overline{\mathbf{E}}=D\tilde{C}^*(\overline{\mathbf{j}}). \tag{11}$$

III. CLOAKING PHENOMENA

In this section we show that the effect of the interface on the macroscopic properties is dependent on the particle size and field intensity. We provide several results illustrating the influence of these two factors.

Our conclusions will apply to all nonlinear constitutive laws given by potential energy densities of the form,

$$W_m(\mathbf{E})=H_m(|\mathbf{E}|),$$

and

$$W_p(\mathbf{E})=H_p(|\mathbf{E}|),$$

where H_m and H_p are strictly convex functions. For this case we have

$$DW_m(\mathbf{E})=H_m'(|\mathbf{E}|)\frac{\mathbf{E}}{|\mathbf{E}|},$$

and

$$DW_p(\mathbf{E})=H_p'(|\mathbf{E}|)\frac{\mathbf{E}}{|\mathbf{E}|},$$

and the macroscopic energies $\tilde{W}(\overline{\mathbf{E}})$ and $\tilde{C}(\overline{\mathbf{E}})$ are convex.[13] Strict convexity insures that the derivataves H_m' and H_p' are invertible. We denote the inverse functions by:

$$(H_m')^{-1}(|\mathbf{j}|),$$

and

$$(H_p')^{-1}(|\mathbf{j}|).$$

We begin by demonstrating the effect of interfacial surface conduction on the macroscopic properties.

A. Cloaking phenomenon for a monodisperse suspension of spheres with highly conducting interface

Given a monodisperse suspension of spheres of radius a, with interface conductivity α, if the electric field intensity $|\overline{\mathbf{E}}|$ across the composite satisfies, $H_m'(|\overline{\mathbf{E}}|)-H_p'(|\overline{\mathbf{E}}|)>0$, and

$$\frac{a}{\alpha}=\frac{2|\overline{\mathbf{E}}|}{(H_m'(|\overline{\mathbf{E}}|)-H_p'(|\overline{\mathbf{E}}|))}, \tag{12}$$

then the electric field $\mathbf{E}$ is precisely $\overline{\mathbf{E}}$, everywhere inside the composite sample and the overall current passing through the sample is given by

$$\overline{\mathbf{j}}=DW_m(\overline{\mathbf{E}}). \tag{13}$$

This result is independent of the shape of the domain Q occupied by the composite and the configuration of the spheres inside the composite domain.

Conversely, this result shows that for prescribed values of the common sphere radius and electric field intensity $|\overline{\mathbf{E}}|$ satisfying $H_m'(|\overline{\mathbf{E}}|)-H_p'(|\overline{\mathbf{E}}|)>0$, that the interfacial conductivity α can be chosen so as to render the particles undetectable.

To fix ideas we present the cloaking phenomenon for the three following cases:

Case 1: Nonlinear particles in a linear matrix.

$$W_m(\mathbf{E})=\left(\frac{\sigma_m}{2}|\mathbf{E}|^2\right)$$

and

$$W_p(\mathbf{E})=\left(\frac{\sigma_p}{2}|\mathbf{E}|^2+\frac{\gamma_p}{t}|\mathbf{E}|^t\right), \tag{14}$$

where $\sigma_m>\sigma_p>0\,\gamma_p>0$, and $t>2$. For a given value of the ratio $\frac{a}{\alpha}$ and a composite with bulk energy density given by Eq. (14), if $|\overline{\mathbf{E}}|$ satisfies

$$\frac{(\sigma_m-\sigma_p)}{\gamma_p}\geqslant|\overline{\mathbf{E}}|^{t-2}$$

and

$$\frac{a}{\alpha}=\frac{2}{(\sigma_m-\sigma_p)-\gamma_p|\overline{\mathbf{E}}|^{t-2}}, \tag{15}$$

then, the electric field inside the composite is $\overline{\mathbf{E}}$ everywhere and $\overline{\mathbf{j}}=DW_m(\overline{\mathbf{E}})$. It is evident that for small fields the particle conductivity lies below that of the matrix. The effect interfacial conduction is seen to compensate for the reduced particle conductivity at low field intensities.

Case 2: Linear particles in a nonlinear matrix.

$$W_m(\mathbf{E})=\left(\frac{\sigma_m}{2}|\mathbf{E}|^2+\frac{\gamma_m}{t}|\mathbf{E}|^t\right)$$

and

$$W_p(\mathbf{E})=\left(\frac{\sigma_p}{2}|\mathbf{E}|^2\right), \tag{16}$$

where $\sigma_m>0$, $\sigma_p>0$, $\gamma_m>0$, and $t>2$. For a given value of the ratio $\frac{a}{\alpha}$ and a composite with bulk energy density given by Eq. (16); if $|\overline{\mathbf{E}}|$ satisfies

$$|\overline{\mathbf{E}}|^{t-2}\geqslant\max\left\{\frac{(\sigma_p-\sigma_m)}{\gamma_m},0\right\},$$

and

$$\frac{a}{\alpha}=\frac{2}{(\sigma_m-\sigma_p)+\gamma_m|\overline{\mathbf{E}}|^{t-2}}, \tag{17}$$

then, the electric field inside the composite is $\overline{\mathbf{E}}$ everywhere and $\overline{\mathbf{j}}=DW_m(\overline{\mathbf{E}})$. For sufficiently large electric fields, the particle conductivity lies below that of the matrix. In this context, interfacial conduction is seen to compensate for the reduced particle conductivity at high field intensities.

Case 3: Nonlinear particles in a nonlinear matrix.

$$W_m(\mathbf{E}) = \left(\frac{\gamma_m}{t+1} |\mathbf{E}|^{t+1} \right)$$

and (18)

$$W_p(\mathbf{E}) = \left(\frac{\gamma_p}{t+1} |\mathbf{E}|^{t+1} \right),$$

where $\gamma_m > \gamma_p > 0$ and $t > 1$. For a given value of the ratio $\frac{a}{\alpha}$ and a composite with bulk energy density given by Eq. (18); if

$$\frac{a}{\alpha} = \frac{2}{(\gamma_m - \gamma_p)|\overline{\mathbf{E}}|^{t-1}}, \tag{19}$$

then, the electric field inside the composite is $\overline{\mathbf{E}}$ everywhere and $\overline{\mathbf{j}} = DW_m(\overline{\mathbf{E}})$.

In the presence of interfacial contact resistance the cloaking effect is expressed in terms of a critical applied current intensity.

B. Cloaking phenomenon for a monodisperse suspension of spheres in the presence of interfacial contact resistance

Given a monodisperse suspension of spheres of radius a, with interface conductivity β, if the applied current intensity $|\overline{\mathbf{j}}|$ satisfies,

$$(H_m')^{-1}(|\overline{\mathbf{j}}|) - (H_p')^{-1}(|\overline{\mathbf{j}}|) > 0, \tag{20}$$

and

$$a \times \beta = \frac{|\overline{\mathbf{j}}|}{((H_m')^{-1}(|\overline{\mathbf{j}}|) - (H_p')^{-1}(|\overline{\mathbf{j}}|))}, \tag{21}$$

then the current density $\mathbf{j}$ is precisely $\overline{\mathbf{j}}$, everywhere inside the composite sample and the overall electric field across the sample is given by

$$\overline{\mathbf{E}} = D\overset{*}{W}_m(\overline{\mathbf{j}}), \tag{22}$$

where $\overset{*}{W}_m$ is the convex dual to the potential energy density of the matrix phase. This result is independent of the shape of the domain Q occupied by the composite and the configuration of the spheres inside the composite domain.

Conversely, this result shows that for prescribed values of the common sphere radius and any field intensity $|\overline{\mathbf{j}}|$. satisfying Eq. (20), that the interfacial conductivity β can be chosen so as to render the particles undetectable.

To fix ideas we present the cloaking phenomenon for the two following cases:

Case 1: Nonlinear particles in a nonlinear matrix.

$$W_m(\mathbf{E}) = \left(\frac{\gamma_m}{t+1} |\mathbf{E}|^{t+1} \right)$$

and (23)

$$W_p(\mathbf{E}) = \left(\frac{\gamma_p}{t+1} |\mathbf{E}|^{t+1} \right),$$

where $\gamma_p > \gamma_m > 0$, $t > 1$ and we set $t' = t/(t-1)$. For a given value of the product $a \times \beta$ and a composite with bulk energy density given by Eq. (23); if

$$a \times \beta = \frac{|\overline{\mathbf{j}}|^{1/t'}}{(\gamma_m)^{-1/t} - (\gamma_p)^{-1/t}}, \tag{24}$$

then, the electric current inside the composite is $\overline{\mathbf{j}}$ everywhere and

$$\overline{\mathbf{E}} = D\overset{*}{W}_m(\overline{\mathbf{j}}) = (\gamma_m)^{-1/t} |\overline{\mathbf{j}}|^{1/t'} \overline{\mathbf{j}}.$$

Case 2: Nonlinear particles in a linear matrix.

$$W_m(\mathbf{E}) = \left(\frac{\sigma_m}{2} |\mathbf{E}|^2 \right)$$

and (25)

$$W_p(\mathbf{E}) = \left(\frac{\sigma_p}{2} |\mathbf{E}|^2 + \frac{\gamma_p}{4} |\mathbf{E}|^4 \right).$$

For a given value of the product $a \times \beta$ and a composite with bulk energy density given by Eq. (25); if $|\overline{\mathbf{j}}|$ satisfies

$$\sigma_m^{-1} |\overline{\mathbf{j}}| - (H_p')^{-1}(|\overline{\mathbf{j}}|) > 0,$$

where

$$(H_p')^{-1}(|\overline{\mathbf{j}}|) = \left(\frac{|\overline{\mathbf{j}}|}{2\gamma_p^2} + \left(\frac{|\overline{\mathbf{j}}|^2}{4\gamma_p^2} + \left(\frac{\sigma_p}{3\gamma_p} \right)^3 \right)^{1/2} \right)^{1/3} + \left(\frac{|\overline{\mathbf{j}}|}{2\gamma_p^2} - \left(\frac{|\overline{\mathbf{j}}|^2}{4\gamma_p^2} + \left(\frac{\sigma_p}{3\gamma_p} \right)^3 \right)^{1/2} \right)^{1/3}, \tag{26}$$

and

$$a \times \beta = \frac{|\overline{\mathbf{j}}|}{\sigma_m^{-1} |\overline{\mathbf{j}}| - (H_p')^{-1}(|\overline{\mathbf{j}}|)}, \tag{27}$$

then, the current density inside the composite is $\overline{\mathbf{j}}$ everywhere and

$$\overline{\mathbf{E}} = D\overset{*}{W}_m(\overline{\mathbf{j}}) = \sigma_m^{-1} \overline{\mathbf{j}}.$$

It is evident that for large fields the particle resistivity lies below that of the matrix. The effect of interfacial contact resistance is seen to compromise the reduced particle resistivity at high current intensities.

It is emphasized that in all cases the cloaking effect applies to anisotropic suspensions of spheres and that the effect is independent of sphere configuration, sample shape, and the concentration of spheres in the sample even beyond the interface percolation threshold. We note that our approach is variational in nature and allows one to treat the problem directly without approximating the field interactions between particles.

In the context of two-phase linear conductors we have, $H_m(|\overline{\mathbf{E}}|) = \sigma_m/2|\overline{\mathbf{E}}|^2$ and $H_p(|\overline{\mathbf{E}}|) = \sigma_p/2|\overline{\mathbf{E}}|^2$ and Eq. (12) becomes

$$a=\mathcal{A}_s=\frac{2\alpha}{(\sigma_m-\sigma_p)}. \tag{28}$$

For $\sigma_m>\sigma_p>0$, $\mathcal{A}_s$ gives a critical radius that is independent of field intensity for which $\mathbf{E}=\overline{\mathbf{E}}$ and $\overline{\mathbf{j}}=\sigma_m\overline{\mathbf{E}}$. These results are independent of field intensity and particle configuration. This result was established in Lipton.[14] For isotropic composites, in the linear context, using different methods Torquato and Rintoul[15] found for $a=\mathcal{A}_s$ that the effective conductivity is precisely that of the matrix phase.

For two phase linear conductors in the presence of an interface contact resistance we have, $H_m(|\overline{\mathbf{E}}|)=\sigma_m/2|\overline{\mathbf{E}}|^2$ and $H_p(|\overline{\mathbf{E}})=\sigma_p/2|\overline{\mathbf{E}}|^2$ and Eq. (21) becomes

$$a\times\beta=\mathcal{A}_c\times\beta=\frac{1}{(\sigma_m^{-1}-\sigma_p^{-1})}. \tag{29}$$

For $\sigma_p>\sigma_m>0$, $\mathcal{A}_c$ gives a critical radius that is independent of field intensity for which $\mathbf{j}=\overline{\mathbf{j}}$ and $\overline{\mathbf{E}}=\sigma_m^{-1}\overline{\mathbf{j}}$. This result is shown to be independent of field intensity and particle configuration. This result was presented in Lipton and Vernescu.[16] For isotropic composites, in the linear context, using different methods Torquato and Rintoul[15] found for $a=\mathcal{A}_c$ that the effective conductivity is precisely that of the matrix phase. The results obtained here translate immediately into the mathematically analogous problems of nonlinear thermal conductivity and antiplane shear.[17]

IV. DUAL VARIATIONAL PRINCIPLES AND ANALYSIS

The cloaking effect for both contact resistance and the highly conducting interface case follows from the variational formulations of the macroscopic energies given by Eqs. (3) and (9), and from new ''dual'' variational formulations described below. We begin by introducing the variational principle describing the convex dual of the macroscopic energy in Eq. (3). We denote the surface of the ith particle by Γ_i and introduce the space of trial currents K' given by

$$K'=\begin{cases}\hat{\mathbf{j}}\colon \dfrac{1}{|Q|}\displaystyle\int_{\partial Q}\hat{\mathbf{j}}\cdot\mathbf{n}\mathbf{x}\;ds=\overline{\mathbf{j}},\\ \displaystyle\int_{\Gamma_i}(\hat{\mathbf{j}}_p-\hat{\mathbf{j}}_m)\cdot\mathbf{n}\;ds=0,\\ \operatorname{div}\hat{\mathbf{j}}=0,\ \text{in}\ Q\backslash\Gamma.\end{cases} \tag{30}$$

The solution, ψ, of the Poisson equation on the interfacial surface given by

$$-\Delta_s\psi=(\hat{\mathbf{j}}_p-\hat{\mathbf{j}}_m)\cdot\mathbf{n}, \tag{31}$$

is written as, $\Delta_s^{-1}\{(\hat{\mathbf{j}}_p-\hat{\mathbf{j}}_m)\cdot\mathbf{n}\}$. We denote the Legendre dual of the matrix and particle potential energy functions as W_m^* and W_p^*. The dual of the potential energy on the interface is simply $W_s^*(\mathbf{D})=1/2\alpha|\mathbf{D}|^2$ for any vector $\mathbf{D}$. We set

$$W^*[\hat{\mathbf{j}},\mathbf{x}]=\begin{cases}W_m^*(\hat{\mathbf{j}}),\ \mathbf{x}\ \text{in the matrix},\\ W_p^*(\hat{\mathbf{j}}),\ \text{x in the particles}.\end{cases} \tag{32}$$

The Legendre dual of the macroscopic energy is written as $\widetilde{W}^*$ and

$$\widetilde{W}^*(\overline{\mathbf{j}})=\inf_{\hat{\mathbf{j}}\in K'}|Q|^{-1}\left\{\int_Q W^*[\hat{\mathbf{j}}(\mathbf{x}),\mathbf{x}]dx+\frac{1}{2\alpha}\int_\Gamma|\nabla_s(\Delta_s^{-1}\{(\hat{\mathbf{j}}_p-\hat{\mathbf{j}}_m)\cdot\mathbf{n}\})|^2ds\right\}. \tag{33}$$

This variational principle is new and is an extension of the variational principle developed in Lipton[18] to the nonlinear case. The dual of the macroscopic energy is found to be convex. For $\overline{\mathbf{j}}=D\widetilde{W}(\overline{\mathbf{E}})$ it is evident that $\widetilde{W}^*(\overline{\mathbf{j}})=\widetilde{W}(\overline{\mathbf{E}})$. These properties together with the variational principle are established in the sequel.

We introduce the variational principle for the dual of the macroscopic energy for the contact resistance case. We denote the surface of the ith particle by Γ_i and introduce the space of trial currents K_c' given by

$$K_c'=\begin{cases}\hat{\mathbf{j}}\colon\hat{\mathbf{j}}\cdot\mathbf{n}=-\overline{\mathbf{j}}\cdot\mathbf{n},\ \text{on the boundary of}\ Q,\\ \operatorname{div}\hat{\mathbf{j}}=0,\ \text{in}\ Q.\end{cases} \tag{34}$$

The dual of the potential energy on the interface is simply $W_s^*(v)=1/2\beta v^2$ for any scalar v. The Legendre dual of the macroscopic energy is given by the following variational principle.

$$\widetilde{C}^*(\overline{\mathbf{j}})=\inf_{\hat{\mathbf{j}}\in K_c'}|Q|^{-1}\left\{\int_Q W^*[\hat{\mathbf{j}}(\mathbf{x}),\mathbf{x}]dx+\frac{1}{2\beta}\int_\Gamma(\hat{\mathbf{j}}\cdot\mathbf{n})^2ds\right\}. \tag{35}$$

This variational principle is new and is an extension of the variational principle developed by Hashin[19] to the nonlinear case. The dual of the macroscopic energy is found to be convex. For $\overline{\mathbf{E}}=D\widetilde{C}^*(\overline{\mathbf{j}})$ it is evident that $\widetilde{C}^*(\overline{\mathbf{j}})=\widetilde{W}(\overline{\mathbf{E}})$. These properties together with the variational principle are established in the sequel.

The cloaking effect is established using simple bounds on the macroscopic energies. We provide the analysis for the highly conducting interface case and we give a brief sketch of the analysis for the contact resistance case as both analyses follow similar lines. The bounds on the macroscopic energy for the highly conducting interface case follow from the substitution of simple trial fields into the new variational principles given by Eqs. (3) and (33). Substitution of the trial potential $\varphi=-\overline{\mathbf{E}}\cdot\mathbf{x}$ into the variational principle of Eq. (3) gives the upper bound

$$U^+(\overline{\mathbf{E}})=\theta_m W_m(\overline{\mathbf{E}})+\theta_p\left(W_p(\overline{\mathbf{E}})+\frac{\alpha}{a}|\overline{\mathbf{E}}|^2\right). \tag{36}$$

Here θ_p and θ_m denote the volume fraction of the matrix and particle phase, respectively.

A lower bound on the macroscopic energy is obtained by finding an upper bound for $\widetilde{W}^*(\overline{\mathbf{j}})$. For any choice of $\overline{\mathbf{j}}$, convex duality implies that

$$\widetilde{W}(\overline{\mathbf{E}})\geqslant\overline{\mathbf{j}}\cdot\overline{\mathbf{E}}-\widetilde{W}^*(\overline{\mathbf{j}}), \tag{37}$$

so for any upper bound $U^{*+}(\overline{\mathbf{j}})$ on the dual energy $\widetilde{W}^*(\overline{\mathbf{j}})$ we have a lower bound on the macroscopic energy given by

$$\widetilde{W}(\overline{\mathbf{E}})\geqslant\overline{\mathbf{j}}\cdot\overline{\mathbf{E}}-U^{*+}(\overline{\mathbf{j}}). \tag{38}$$

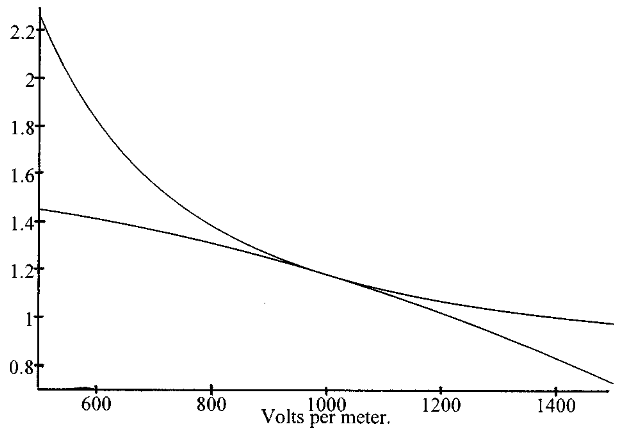

FIG. 1. Tight upper and lower bounds on the macroscopic potential energy for a random, nonlinear, particle reinforced composite with highly conducting interface.

Substitution of the trial current

$$\hat{\mathbf{j}}=\begin{cases}\overline{\mathbf{j}}, & \text{in the matrix,}\\ \overline{\mathbf{j}}+\boldsymbol{\eta}, & \text{in the particles,}\end{cases} \tag{39}$$

gives the upper bound

$$U^{*+}(\overline{\mathbf{j}})=\theta_m W_m^*(\overline{\mathbf{j}})+\theta_p W_p^*(\overline{\mathbf{j}}+\boldsymbol{\eta}) + \frac{1}{|Q|}\int_\Gamma \frac{1}{2\alpha}|\nabla_s\{\Delta_s^{-1}(\boldsymbol{\eta}\cdot\mathbf{n})\}|^2 ds. \tag{40}$$

For a sphere of radius a one finds that $\Delta_s^{-1}(\boldsymbol{\eta}\cdot\mathbf{n}) = -a^2/2\boldsymbol{\eta}\cdot\mathbf{n}$ and calculation gives

$$U^{*+}(\overline{\mathbf{j}})=\theta_m W_m^*(\overline{\mathbf{j}})+\theta_p\left(W_p^*(\overline{\mathbf{j}}+\boldsymbol{\eta})+\frac{a}{4\alpha}|\boldsymbol{\eta}|^2\right). \tag{41}$$

We set $\overline{\mathbf{j}}=DW_m(\overline{\mathbf{E}})$ and $\eta=DW_p(\overline{\mathbf{E}})-DW_m(\overline{\mathbf{E}})$ in Eq. (41); and a straightforward calculation delivers the lower bound $U^-(\overline{\mathbf{E}})$ on the macroscopic energy given by

$$U^-(\overline{\mathbf{E}})=\theta_m W_m(\overline{\mathbf{E}})+\theta_p W_p(\overline{\mathbf{E}})+\theta_p[DW_m(\overline{\mathbf{E}})\cdot\overline{\mathbf{E}}] - \theta_p[DW_p(\overline{\mathbf{E}})\cdot\overline{\mathbf{E}}]-\theta_p\frac{a}{4\alpha}|DW_p(\overline{\mathbf{E}})-DW_m(\overline{\mathbf{E}})|^2. \tag{42}$$

Inspection of Eqs. (36) and (42) shows that the upper and lower bounds touch when

$$0=-4|\overline{\mathbf{E}}|^2+4\frac{a}{\alpha}(H_m'(|\overline{\mathbf{E}}|)-H_p'(|\overline{\mathbf{E}}|))|\overline{\mathbf{E}}| - \left(\frac{a}{\alpha}\right)^2|H_m'(|\overline{\mathbf{E}}|)-H_p'(|\overline{\mathbf{E}}|)|^2. \tag{43}$$

It is evident that Eq. (43) holds when $H_m'(|\overline{\mathbf{E}}|)-H_p'(|\overline{\mathbf{E}}|)>0$ and $\frac{a}{\alpha}$ satisfies Eq. (12). Strict convexity of the energy densities of each phase insure that $\varphi=-\overline{\mathbf{E}}\cdot\mathbf{x}$ is the unique minimizer of the variational principle of Eq. (3) when $\frac{a}{\alpha}$ satisfies Eq. (12). Last, it is easily calculated that the overall current density is given by $DW_m(\overline{\mathbf{E}})$.

Figure 1 shows upper and lower bounds on the macroscopic potential energy for a random, particle reinforced composite. The composite is comprised of two conductors with cubic nonlinear behavior, namely $DW_m(\mathbf{E})=\gamma_m|\mathbf{E}|^2\mathbf{E}$ and $DW_p(\mathbf{E})=\gamma_p|\mathbf{E}|^2\mathbf{E}$. The bounds on the macroscopic potential energy density are normalized with respect to the matrix potential energy density given by $\gamma_m|E|^4/4$. The bounds are plotted for a random dispersion of particles of radius $a=20\,\mu$m occupying a volume fraction of 0.2. The dimensionless ratio γ_p/γ_m is set to 0.1, and the combination $\alpha/a\gamma_m$ is chosen to be 450 000 (v/m)^2. The critical field strength for which the electric potential in the composite is the same as an unreinforced sample is 1000 v/m.

For the contact resistance case we perform a similar analysis. However our analysis now delivers bounds for the dual macroscopic energy $\tilde{C}^*$. An upper bound on $\tilde{C}^*$ follows from substitution of the trial field $\overline{\mathbf{j}}$ into the variational principle of Eq. (35). It is given by

$$\overset{*}{U}{}^+(\overline{\mathbf{j}})=\theta_m \overset{*}{W}_m(\overline{\mathbf{j}})+\theta_p\left(\overset{*}{W}_p(\overline{\mathbf{j}})+\frac{1}{2\beta a}|\overline{\mathbf{j}}|^2\right). \tag{44}$$

A lower bound on $\tilde{C}^*$ is obtained by finding an upper bound for $\tilde{C}(\overline{\mathbf{E}})$. For any upper bound $U^+(\overline{\mathbf{E}})$ on the energy $\tilde{C}(\overline{\mathbf{E}})$ we have the lower bound given by

$$\tilde{C}^*(\overline{\mathbf{j}})\geqslant \overset{*}{U}{}^-(\overline{\mathbf{j}})=\overline{\mathbf{j}}\cdot\overline{\mathbf{E}}-U^+(\overline{\mathbf{E}}). \tag{45}$$

Substitution of the trial field

$$\phi=\begin{cases}[D\overset{*}{W}_m(\overline{\mathbf{j}})]\cdot\mathbf{x}, & \text{in the matrix,}\\ [D\overset{*}{W}_p(\overline{\mathbf{j}})]\cdot\mathbf{x}, & \text{in the particles,}\end{cases} \tag{46}$$

into the variational principle for $\tilde{C}(\overline{\mathbf{E}})$ delivers the lower bound

$$\overset{*}{U}{}^-(\overline{\mathbf{j}})=\theta_m\overset{*}{W}_m(\overline{\mathbf{j}})+\theta_p\overset{*}{W}_p(\overline{\mathbf{j}})+\theta_p[D\overset{*}{W}_m(\overline{\mathbf{j}})\cdot\overline{\mathbf{j}}] - \theta_p[D\overset{*}{W}_p(\overline{\mathbf{j}})\cdot\overline{\mathbf{j}}]-\theta_p\frac{a\beta}{2}|D\overset{*}{W}_p(\overline{\mathbf{j}})-D\overset{*}{W}_m(\overline{\mathbf{j}})|^2. \tag{47}$$

The cloaking effect for the contact resistance case follows immediately from the condition that $\overset{*}{U}{}^-(\overline{\mathbf{j}})=\overset{*}{U}{}^+(\overline{\mathbf{j}})$ and the identities:

$$D\overset{*}{W}_m(\overline{\mathbf{j}})=(H_m')^{-1}(|\overline{\mathbf{j}}|)\frac{\overline{\mathbf{j}}}{|\overline{\mathbf{j}}|},\qquad D\overset{*}{W}_p(\overline{\mathbf{j}})=(H_p')^{-1}(|\overline{\mathbf{j}}|)\frac{\overline{\mathbf{j}}}{|\overline{\mathbf{j}}|}. \tag{48}$$

Figure 2 shows upper and lower bounds on the dual macroscopic potential energy for a random, particle reinforced composite. The composite is comprised of two conductors with cubic nonlinear behavior, namely $DW_m(\mathbf{E})=\gamma_m|\mathbf{E}|^2\mathbf{E}$ and $DW_p(\mathbf{E})=\gamma_p|\mathbf{E}|^2\mathbf{E}$. The bounds on the macroscopic potential energy density are normalized with respect to the dual matrix potential energy density given by $3\gamma_m^{-1/3}|j|^{4/3}/4$. The bounds are plotted for a random dispersion of particles of radius $a=20\,\mu$m occupying a volume fraction of 0.2. The dimensionless ratio γ_p/γ_m is set to 8.0,

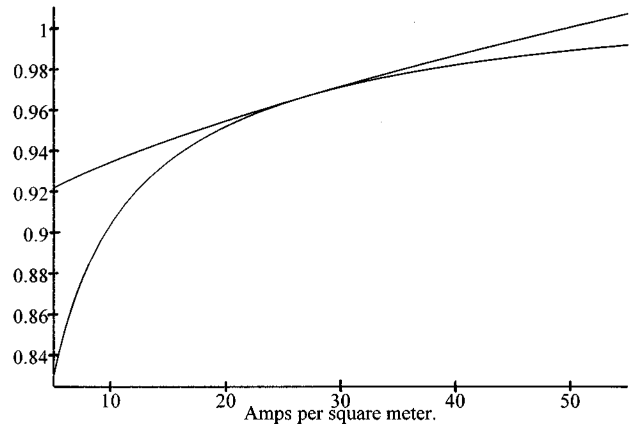

FIG. 2. Tight upper and lower bounds on the dual macroscopic potential energy for a random, nonlinear, particle reinforced composite in the presence of interface contact resistance.

and the combination $a\beta/\gamma_p^{1/3}$ is chosen to be 9 $(\mathrm{A/m^2})^{2/3}$. The critical field strength for which the current density in the composite is the same as an unreinforced sample is 27 $\mathrm{A/m^2}$.

V. VARIATIONAL PRINCIPLES FOR DUAL MACROSCOPIC ENERGIES

Last, the variational principles for the convex duals to the macroscopic energies are established. We provide the analysis for the highly conducting interface case—a similar analysis delivers variational principles for the dual macroscopic energy in the presence of interface contact resistance. Application of convex duality to the bulk and surface energies gives

$$\tilde{W}(\tilde{\mathbf{E}}) \geqslant \inf_{\mathbf{E}\in K}\left\{\frac{1}{|Q|}\int_Q [\hat{\mathbf{j}}(\mathbf{x})\cdot\mathbf{E}(\mathbf{x})] - W^*[\hat{\mathbf{j}}(\mathbf{x}),\mathbf{x}]dx + \frac{1}{|Q|}\int_\Gamma [\mathbf{v}_{\tan}(\mathbf{x})\cdot\mathbf{E}_{\tan}(\mathbf{x})] - W_s^*[\mathbf{v}_{\tan}(\mathbf{x})]ds\right\}. \tag{49}$$

In order that the infimum be greater than $-\infty$, integration by parts shows that the fields $\hat{\mathbf{j}}$ and $\mathbf{v}$ must satisfy,

$$\operatorname{div}\hat{\mathbf{j}}=0, \text{ in } Q/\Gamma,$$

$$\int_{\Gamma_i}(\hat{\mathbf{j}}_p-\hat{\mathbf{j}}_m)\cdot\mathbf{n}ds=0, \tag{50}$$

and

$$(\hat{\mathbf{j}}_p-\hat{\mathbf{j}}_m)\cdot\mathbf{n}=\operatorname{div}_s\mathbf{v}_{\tan}+(\mathbf{v}\cdot\mathbf{n})\mathcal{J}, \text{ on } \Gamma.$$

Here div_s is the surface divergence operator and $\mathcal{J}=-\operatorname{div}_s\mathbf{n}$ is the mean curvature on the interface. Application of these identies gives

$$\tilde{W}(\overline{\mathbf{E}}) \geqslant \overline{\hat{\mathbf{j}}}\cdot\overline{\mathbf{E}} - \left\{\frac{1}{|Q|}\int_Q W^*[\hat{\mathbf{j}}(\mathbf{x}),\mathbf{x}]dx - \frac{1}{|Q|}\int_\Gamma W_s^*[\mathbf{v}_{\tan}(\mathbf{x})]ds\right\}, \tag{51}$$

where $\overline{\hat{\mathbf{j}}}$ is the average of $\hat{\mathbf{j}}$ given by

$$\overline{\hat{\mathbf{j}}} = \frac{1}{|Q|}\int_{\partial Q}\hat{\mathbf{j}}\cdot\mathbf{n}\mathbf{x}\, ds. \tag{52}$$

Among all $\hat{\mathbf{j}}$ and $\mathbf{v}$ satisfying Eq. (50) we consider only trial fields $\mathbf{v}$ of the special form $\mathbf{v}=\nabla_s g$ and set

$$\hat{W}(\overline{\hat{\mathbf{j}}}) = \inf_{(\hat{\mathbf{j}},\mathbf{v}=\nabla_s g)}\frac{1}{|Q|}\left\{\int_Q W^*[\hat{\mathbf{j}}(\mathbf{x}),\mathbf{x}]dx + \int_\Gamma W_s^*(\nabla_s g)ds\right\}. \tag{53}$$

It is evident that for this choice $(\hat{\mathbf{j}}_p-\hat{\mathbf{j}}_m)\cdot\mathbf{n}=\Delta_s g$. It follows from Eq. (51) that

$$\tilde{W}(\overline{\mathbf{E}}) \geqslant \overline{\hat{\mathbf{j}}}\cdot\overline{\mathbf{E}} - \hat{W}(\overline{\hat{\mathbf{j}}}). \tag{54}$$

The convexity of $\hat{W}$ follows from standard arguments. The inequality given by Eq. (54) implies $\tilde{W}(\overline{\mathbf{E}}) \geqslant \hat{W}^*(\overline{\mathbf{E}})$. On the other hand one easily checks that $\tilde{W}(\overline{\mathbf{E}}) = \hat{W}^*(\overline{\mathbf{E}})$ for the choice $\hat{\mathbf{j}}=DW_m(-\nabla\phi)$, in the matrix, $\hat{\mathbf{j}}=DW_p(-\nabla\phi)$, in the particles and $\mathbf{v}=-\nabla_s\phi$, where ϕ is the electric potential in the composite. We conclude from the convexity of $\hat{W}$ that $\tilde{W}^*(\overline{\hat{\mathbf{j}}}) = \hat{W}(\overline{\hat{\mathbf{j}}})$ and the variational principle follows.

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