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Duality Relations for Non-Ohmic Composites, with Applications to Behavior near Percolation

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Keller, Dykhne, and others have exploited duality to derive exact results for the effective behavior of two-dimensional Ohmic composites. This paper addresses similar issues in the non-Ohmic context. We focus primarily on three different types of nonlinearity: (a) the weakly nonlinear regime; (b) power-law behavior; and (c) dielectric breakdown. We first make the consequences of duality explicit in each setting. Then we draw conclusions concerning the critical exponents and scaling functions of "dual pairs" of random non-Ohmic composites near a percolation threshold. These results generalize, unify, and simplify relations previously derived for nonlinear resistor networks. We also discuss some self-dual nonlinear composites. Our treatment is elementary and self-contained; however, we also link it with the more abstract mathematical discussions of duality by Jikov and Kozlov.

KEY WORDS: Nonlinear Composites; duality; effective properties; percolation.

1. INTRODUCTION

Convex duality is important in many areas of physics. In the present context —inhomogeneous conducting materials—it gives the relationship between two well-known variational principles: one for the electric field, the other for the current field. Other examples of duality include the complementary variational principles of mechanics, and duality in linear programming.

In most settings the primal and dual variational principles involve unknowns of different types. In 3D conductivity, for example, the primal involves curl-free fields while the dual involves divergence-free fields. The situation is different, however, for systems which are effectively two-dimensional—for example when the microgeometry has cylindrical symmetry, or

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when a thin film has thickness much smaller than all other microgeometric length scales. Then the primal and dual variational principles involve unknowns which are fundamentally similar, since divergence-free and curl-free fields are equivalent under rotation by 90 degrees.

For 2D composites, duality can be viewed differently. Instead of describing each composite by a pair of dual variational principles, we can associate with each composite a physically different *dual composite*, whose structure and properties are related to those of the starting composite in a simple, algebraic way. We shall explain this in detail in Section 2. Viewed this way, duality becomes a scheme for relating the effective behavior of certain pairs of systems.

The consequences of duality have been studied quite extensively for linearly conducting composites—i.e. those made from Ohmic materials, whose local conductivity is independent of the local electric field. The first application was by Keller, who derived a "phase interchange relation" for two-component composites made from isotropic Ohmic conductors.[1,2] Subsequently but independently, Dykhne derived an exact expression for the conductivity of an isotropic polycrystalline film made of a single anisotropic material.[3] These results were unified by Mendelson, who extended them to systems with general anisotropic conductivity tensors.[4,5] Alternative proofs of Mendelson's relations have been given by Kohler and Papanicolaou[6] and Nevard and Keller.[7] Related results have also been obtained for other two-dimensional problems, see e.g. Milton,[8] Helsing, Milton, and Movchan[9] and the references therein.

In the non-Ohmic case the existence of dual variational principles is well-known. The literature on nonlinear conductivity in composites is relatively sparse, but a huge amount of work has been done on mechanical properties of nonlinear composites. In polycrystal plasticity, for example, Hill,[10] Hutchinson,[11] and many others have made use of the dual variational principles for stress and strain-rate. A rigorous mathematical treatment of dual variational principles for nonlinear composites can be found in the book by Jikov, Kozlov, and Oleinik.[12]

For 2D non-Ohmic composites, duality can once again be viewed differently—as a means of relating each system to a physically different dual. Curiously, however, the consequences of duality for 2D composites have been almost entirely ignored in the non-Ohmic context. The only investigations we know are the short article of Kozlov[13] and a short discussion in the book of Jikov, Kozlov, and Oleinik.[12] This work clearly communicates the *idea* that duality should have consequences, but it considers in detail only a few specific applications. There is also some work addressing analogous issues for *networks* of non-Ohmic resistors, see e.g. Straley and Kenkel.[14]

The goal of this paper is a systematic and concrete treatment of the consequences of duality for 2D non-Ohmic composites. We start, in Section 2, with a self-contained treatment of duality in the nonlinear setting. Then we specialize, in Section 3, to three different cases: (a) the weakly nonlinear regime; (b) strongly nonlinear composites with power-law behavior; and (c) dielectric breakdown. We concentrate in each case on the consequences of duality for behavior near a percolation threshold. In particular we show: (a) equality of the critical exponents describing the weakly nonlinear behavior of certain composites above and below a percolation threshold (see Eq. (44)); (b) a simple relation between the critical exponents of power law composites (Eq. (59)); and (c) equality of the critical exponents describing the threshold field of a system undergoing dielectric breakdown and the critical current of a inhomogeneous perfect conductor (Eq. (70)).

Our conclusions, based on duality, are among the few exact relations that exist concerning the effective behavior of nonlinear dielectric composites. Such materials have attracted increasing theoretical and experimental attention during the past 10 years, in part because of their many potential applications in optical and electronic devices, see e.g. Bergman and Stroud[15] and the references cited there. For mathematical treatments see Miksis,[16] Dell'Antonio[17] and Jikov et al.[12] It is generally not practical to solve for the bulk effective properties of such composites analytically or numerically. Therefore it is necessary to rely upon limiting cases, special geometries, bounds, exact relations, and self-consistent estimates. Some specific results include: (1) an equivalence between the effective nonlinearity of a weakly nonlinear composite and the flicker noise amplitude of a linear composite with the same microgeometry;[18] (2) an evaluation of the effective dielectric constant of a strongly nonlinear composite to second order in the local dielectric constant fluctuations;[19] (3) a calculation of the bulk effective conductivity of layered microgeometries;[20] (4) various extensions of the Hashin–Shtrikman bounds to strongly nonlinear composites;[21–24] and (5) an extension of the Maxwell Garnett theory to dilute mixtures of nonlinear inclusions in a linear host.[25, 26] The duality relation explored in this paper provides an additional exact result.

2. DUALITY OF 2D NONLINEAR COMPOSITES

There are two alternative approaches: one based on differential equations, the other on variational principles. We shall discuss them both, in Sections 2.1 and 2.2 respectively, after establishing some basic notation. Section 2.3 discusses a little-known subtlety that arises when the component materials have power law behavior with different exponents.

Our composites are heterogeneous bodies in the x, y plane whose local conductivity $\sigma(x, y, E)$ depends on the spatial coordinates x, y and also on the local electric field E. The local electric current J is thus a nonlinear function of the electric field E:

$$J = J(x, y, E(x, y)) = \sigma(x, y, E(x, y))\, E(x, y) \tag{1}$$

We will usually drop the explicit dependence on x, y to achieve greater compactness of notation. The electric field comes from a potential, $E = -\nabla\Phi$, and the current field is divergence free, $\nabla \cdot J = 0$, so the electric potential satisfies the partial differential equation

$$\nabla \cdot [\sigma(x, y, -\nabla\Phi)\, \nabla\Phi] = 0$$

and a suitable boundary condition.

When discussing the bulk effective conductivity it is natural to impose one of the "constant applied field" boundary conditions:

$$\Phi = -E_0 \cdot (x, y) \tag{2}$$

or

$$J \cdot \hat{n} = J_0 \cdot \hat{n} \tag{3}$$

where $\hat{n}$ is a unit vector normal to the boundary. Abusing notation conveniently, we shall say "$E = E_0$ at the boundary" when we mean (2), and "$J = J_0$ at the boundary" when we mean (3). Writing

$$\langle E \rangle = \frac{1}{V} \int E(x, y)\, dV, \qquad \langle J \rangle = \frac{1}{V} \int J(x, y)\, dV \tag{4}$$

for the volume-averaged electric and current fields, we may define the bulk effective conductivity σ^* to be the ratio of the two:

$$\langle J \rangle = \sigma^* \langle E \rangle \tag{5}$$

Notice that if $E = E_0$ at the boundary then $\langle E \rangle = E_0$ but $\langle J \rangle$ must be calculated by solving the differential equation. Similarly, if $J = J_0$ at the boundary $\langle J \rangle = J_0$ but $\langle E \rangle$ must be calculated by solving the differential equation.

The effective conductivity depends on the composition of the body, i.e. the form of the function $x, y, E \to \sigma(x, y, E)$. If the local behavior is non-Ohmic then so is the bulk behavior: σ^* is a function of $\langle E \rangle$ (or, equivalently, of $\langle J \rangle$) as we vary the applied field. In general σ^* depends on the

form of the boundary condition, i.e. the choice of (2) vs. (3). However this dependence disappears as length scale of the microstructure tends to zero—the limit in which the composite behaves as a homogeneous body with conductivity σ^*.

Our definition (5) is standard, but it is not the only approach. Some authors prefer to define the effective behavior of a nonlinear composite through a variational principle for its "effective energy," see e.g. ref. 12. The consistency of these two definitions is well-known; we will review it in Section 2.2.

A word is in order concerning restrictions on the local conductivity $\sigma(x, y, E)$. We shall always assume that the dependence of J on E is derivable from a convex potential.[2] This means

$$J_i = \frac{\partial U}{\partial E_i} \tag{6}$$

for some potential energy $U(x, y, E)$, assumed convex in its dependence on E with x and y held fixed. This hypothesis guarantees the uniqueness of the local fields. It also assures invertibility of the relation $J = J(E)$, in other words it lets us solve for E in terms of J. The existence of a potential energy is equivalent to $\partial J_i/\partial E_j = \partial J_j/\partial E_i$. This is easily expressed in terms of σ using (1):

$$\sigma_{ij} + \sum_{k=1}^{2} \frac{\partial \sigma_{ik}}{\partial E_j} E_k = \sigma_{ji} + \sum_{k=1}^{2} \frac{\partial \sigma_{jk}}{\partial E_i} E_k \tag{7}$$

which reduces in the Ohmic case to the familiar symmetry condition $\sigma_{ij} = \sigma_{ji}$. The convexity of the potential is equivalent to positive definiteness of $\partial J_i/\partial E_j$, i.e. positivity of the symmetric matrix whose (i, j)th element is given by (7). This reduces in the Ohmic case to the familiar condition that σ_{ij} be positive definite.

It is not always convenient to work in such generality. A special but very important case arises when the composite is locally isotropic, i.e. $\sigma(x, y, E)$ is scalar-valued. The existence of a potential implies that a scalar-valued σ can depend on E only through $|E|$:

$$J = \sigma(x, y, |E|)\, E \tag{8}$$

Our convexity hypothesis becomes

$$\sigma(|E|) > 0 \qquad \text{and} \qquad \sigma(|E|) + |E|\, \sigma'(|E|) > 0 \tag{9}$$

[2] We use the term "potential" for both Φ (the electric potential) and U (the potential energy) trusting that no confusion will arise.

This implies that $|J| = \sigma(|E|)\,|E|$ is a monotone function of $|E|$, so one can solve (8) uniquely for E as a function of J.

The discussion is naturally not limited to conductivity. It applies to any transport property connecting a divergence-free field (the "current") with a curl-free field (the "electric field"). Examples include thermal conductivity, magnetic permeability, and dielectric behavior.

2.1. Duality via Field Equations

We begin with the simplest case: a macroscopically isotropic non-Ohmic composite made from locally isotropic non-Ohmic materials. In this case the local J–E relation is given by (8) and the macroscopic J–E relation has the form $\langle J\rangle = \sigma^*(|\langle E\rangle|)\langle E\rangle$. Our goal is to determine a physically different "dual composite" whose effective behavior is related to that of the original ("primal") composite. We suppose that for the primal composite σ^* is defined using a constant-electric-field boundary condition. As we shall see, for the dual composite the effective behavior must be defined instead using a constant-current boundary condition.

Following Mendelson[5] and others, we recognize that $J^\perp = (-J_2, J_1)$ is a gradient: there is a function Ψ such that

$$J^\perp = -\nabla\Psi \tag{10}$$

Also, $E^\perp = (-E_2, E_1)$ is divergence-free, and it satisfies $E^\perp = E_0^\perp$ at the boundary if $E = E_0$ there. From the local relation (8) we get

$$|J| = \sigma(|E|)\,|E| = J(|E|)$$

with the convenient abuse of notation $J(t) = \sigma(t)\,t$. (Thus $J(|E|)$ represents the magnitude of the current, while $J(E)$ represents the current vector.) Our convexity hypothesis implies that $J(t)$ is monotone, so we can invert the preceding relation to give

$$|E| = J^{-1}(|\nabla\Psi|)$$

where J^{-1} is the inverse function of J. Here we write $J^{-1}(|\nabla\Psi|)$ instead of $J^{-1}(|J|)$ to avoid using the two different interpretations of J in a single formula. Since σ is scalar-valued we have

$$E^\perp = \frac{1}{\sigma(|E|)}\,J^\perp = \frac{1}{\sigma(J^{-1}(|\nabla\Psi|))}\,J^\perp \tag{11}$$

We now give the constitutive law for the dual composite. It is chosen so that the current field is $E^{\perp}$ when the electric field is $J^{\perp}$. In view of (11), this is achieved by defining the local conductivity of the dual composite to be

$$\sigma_d(x, y, |\nabla\Psi|) = \frac{1}{\sigma(x, y, J^{-1}(|\nabla\Psi|))} \tag{12}$$

when the electric field is $-\nabla\Psi$. If the effective conductivity σ_d^* of the dual composite is defined using the constant-current boundary condition $E^{\perp} = E_0^{\perp}$, then averaging of (11) gives

$$\langle E^{\perp}\rangle = \sigma_d^*(|\langle J\rangle|)\langle J^{\perp}\rangle$$

We are assuming that σ_d^* is scalar, so the preceding formula can be rewritten

$$\langle E\rangle = \sigma_d^*(|\langle J\rangle|)\langle J\rangle$$

Remembering that $E_0 = \langle E\rangle$ and writing $J_0 = \langle J\rangle = \sigma^*(|E_0|)\, E_0$, we have shown that

$$\sigma_d^*(|J_0|) = 1/\sigma^*(|E_0|)$$

It is convenient to write this in less compact notation, emphasizing the dependence of σ^* on the applied field and the form of the local conductivity:

$$\sigma^*\left(\frac{1}{\sigma(x, y, J^{-1}(|\nabla\Psi|))}; |J_0|\right) = \frac{1}{\sigma^*(\sigma(x, y, |\nabla\Phi|); |E_0|)} \tag{13}$$

This is the duality relation for effective conductivities of non-Ohmic inhomogeneous materials in the isotropic context.

We turn now to the general case, when both the local conductivity and the effective conductivity are permitted to be anisotropic. The local J–E relation is (1) and the effective conductivity is defined by

$$\langle J\rangle = \sigma^*(\langle E\rangle)\langle E\rangle \tag{14}$$

We suppose as before that σ^* is defined for the primal composite using the constant-electric-field boundary condition $E = E_0$.

The key to the isotropic case was (11). It generalizes easily: if $J = \sigma(E)\, E$ then

$$E^{\perp} = R\sigma^{-1}(E)\, R^T J^{\perp} \tag{15}$$

where R represents rotation by 90°, i.e. $R\xi = \xi^{\perp}$. We are assuming that the J–E relation is invertible, so it can be used to write E as a function of $J^{\perp}$. We write this dependence as $E = \mathscr{E}(J^{\perp})$.

The relation (15) dictates the constitutive law for the dual composite: its local conductivity is

$$\sigma_d(x, y, -\nabla\Psi) = R\sigma^{-1}(x, y, \mathscr{E}(x, y, -\nabla\Psi))\, R^T \tag{16}$$

when the electric field is $-\nabla\Psi$. This is chosen so that the current field is $E^{\perp}$ when the electric field is $J^{\perp}$. If we define the effective conductivity σ_d^* using the constant-current boundary condition $E^{\perp} = E_0^{\perp}$ then averaging of (15) gives

$$\langle E^{\perp} \rangle = \sigma_d^*(\langle J^{\perp} \rangle)\langle J^{\perp} \rangle \tag{17}$$

Now, $\langle J^{\perp} \rangle = \langle RJ \rangle = R\langle J \rangle$ and similarly $\langle E^{\perp} \rangle = R\langle E \rangle$. Taking this into account, and noting that $R^T = -R$, we see that (17) is equivalent to

$$\langle E \rangle = R\sigma_d^*(\langle J^{\perp} \rangle)\, R^T \langle J \rangle$$

Remembering that $E_0 = \langle E \rangle$ and writing $J_0 = \langle J \rangle = \sigma^*(E)\, E_0$, we have shown that

$$R\sigma_d^*(J_0^{\perp})\, R^T = [\sigma^*(E_0)]^{-1}$$

In less compact notation, emphasizing the dependence of σ^* on the applied field and the form of the local conductivity:

$$R\sigma^*\left(\frac{1}{R\sigma(x, y, \mathscr{E}(-\nabla\Psi))\, R^T}; J_0^{\perp}\right) R^T = \frac{1}{\sigma^*(\sigma(x, y, -\nabla\Phi); E_0)} \tag{18}$$

This is the general duality relation for the effective conductivities of non-Ohmic composites.

2.2. Duality via Variational Principles

One can define effective behavior using variational principles rather than field equations. Recall from (6) that the local J–E relation is assumed to come from a convex potential: $J = \partial U/\partial E$. We define the associated dual potential by

$$\hat{U}(J) = \max_E \{J \cdot E - U(E)\} \tag{19}$$

and we observe that

$$J=\frac{\partial U}{\partial E} \qquad \text{is equivalent to} \quad E=\frac{\partial \hat{U}}{\partial J} \tag{20}$$

We discussed two slightly different definitions of the effective conductivity, one using the constant-electric-field boundary condition $E=E_0$, the other using the constant-current boundary condition $J=J_0$. The former corresponds to the variational principle for the macroscopic potential at applied electric field E_0:

$$U^*(E_0)=\min_{\substack{\tilde{E}=E_0-\nabla\tilde{\Phi}\\ \tilde{\Phi}=0 \text{ at bdry}}} \frac{1}{V}\int U(x, y, \tilde{E})\, dV \tag{21}$$

The latter corresponds to the variational principle for the dual macroscopic potential at applied current field J_0:

$$\hat{U}^*(J_0)=\min_{\substack{\nabla\cdot\tilde{J}=0\\ \tilde{J}\cdot n=J_0\cdot n \text{ at bdry}}} \frac{1}{V}\int \hat{U}(x, y, \tilde{J})\, dV \tag{22}$$

Elaborating on this point, consider first the variational principle (21). If E and $J=\sigma(E)\,E$ solve the field equations with boundary condition $E=E_0$, then the first variation of (21) vanishes at E, so by convexity E achieves the minimum. Differentiating both sides of (21) with respect to E_0 we find that

$$\frac{\partial U^*}{\partial E}(E_0)=\frac{1}{V}\int \frac{\partial U}{\partial E}(E)\, dV=\langle J\rangle$$

Thus we have

$$J_0=\sigma^*(E_0)\,E_0=\frac{\partial U^*}{\partial E}(E_0)$$

with $J_0=\langle J\rangle$. Thus the effective behavior of the composite is associated with the potential U^*. A similar argument shows that when σ^* is defined using boundary condition $J=J_0$, the effective behavior of the composite is associated with the potential $\hat{U}^*$.

What does our "dual composite" look like in this variational setting? The answer is simple: if the primal composite has potential $U(x, y, E)$ then the dual composite has potential

$$U_d(x, y, -\nabla\Psi)=\hat{U}(x, y, (\nabla\Psi)^{\perp}) \tag{23}$$

To show this, we need merely verify that if E minimizes the constant-electric-field variational principle (21) for the primal composite then $E^{\perp}$ minimizes the constant-current variational principle (22) for the dual composite.

The desired application of (22) has integrand $\hat{U}_d$. Starting from the definition (19) we have

$$\begin{aligned}\hat{U}_d(\eta) &= \max_{\xi} \{\eta \cdot \xi - U_d(\xi)\} \\ &= \max_{\xi} \{\eta \cdot \xi - \hat{U}(-\xi^{\perp})\} \\ &= \max_{\xi} \{\eta \cdot (\xi^{\perp}) - \hat{U}(\xi)\} \\ &= \max_{\xi} \{-\eta^{\perp} \cdot \xi - \hat{U}(\xi)\} \\ &= U(-\eta^{\perp})\end{aligned}$$

using for the last step the convexity of U. Recognizing that

$$\nabla \cdot \tilde{J} = 0,\ \tilde{J} \cdot n = E_0^{\perp} \cdot n \text{ at bdry} \Leftrightarrow -\tilde{J}^{\perp} = E_0 - \nabla\tilde{\Phi},\ \tilde{\Phi} = 0 \text{ at bdry}$$

we see that the constant-current variational principle for the dual composite evaluated at $E_0^{\perp}$ is

$$\begin{aligned}\hat{U}_d^*(E_0^{\perp}) &= \min_{\substack{\nabla \cdot J = 0 \\ J \cdot n = E_0^{\perp} \cdot n \text{ at bdry}}} \frac{1}{V} \int \hat{U}_d(x, y, \tilde{J})\, dV \\ &= \min_{\substack{\tilde{E} = E_0 - \nabla\tilde{\Phi} \\ \tilde{\Phi} = 0 \text{ at bdry}}} \int \frac{1}{V} U(x, y, \tilde{E})\, dV\end{aligned}$$

Thus it is *equivalent* to the constant-electric-field variational principle for the primal composite. It follows that the optimal J for the dual composite is $E^{\perp}$, as asserted.

2.3. Function Space Issues

We have said nothing about how singular J or E may be. It is usually unnecessary to specify such information because the answer is obvious: each field should have finite energy for the associated variational principle

(21)–(22). This determines the appropriate function spaces for E and J in many cases. Suppose, for example, the potential satisfies

$$-c_0 + c_1 |E|^\alpha \leqslant U(x, y, E) \leqslant c_0 + c_2 |E|^\alpha \tag{24}$$

for some constants $c_0, c_1, c_2 > 0$ and $\alpha > 1$. Then the dual potential $\hat{U}$ satisfies a similar condition with α replaced by $\alpha' = \alpha/(\alpha - 1)$, and we expect

$$\int |E|^\alpha \, dV < \infty, \qquad \int |J|^{\alpha'} \, dV < \infty \tag{25}$$

For specific microstructures the solutions can be more regular. But singularities can arise, for example at multigrain junctions. The singularities are determined by the form of the local conductivity, and we have no right to constrain them.

The situation is different and more subtle, however, when the growth law of the potential is spatially heterogeneous. Suppose, for example, the potential satisfies

$$-c_0 + c_1 |E|^{\alpha_1} \leqslant U(x, y, E) \leqslant c_0 + c_2 |E|^{\alpha_2} \tag{26}$$

rather than (24), with $\alpha_1 < \alpha_2$. Then it is not clear whether to impose the weaker condition that E have finite energy, or the stronger one that $\int |E|^{\alpha_2} \, dV < \infty$. Somewhat surprisingly, we have the right to choose—and the solution can depend on the choice! This has been explored by Jikov[27, 28] and in Chapters 14–15 of ref. 12.

The following example, due to Jikov, reveals the essence of the matter. Consider a composite made of two materials, with potentials

$$U^{(1)}(E) = |E|^{\alpha_1} \qquad \text{and} \qquad U^{(2)}(E) = |E|^{\alpha_2}$$

with $\alpha_1 < 2 < \alpha_2$. Suppose furthermore that the composite has a checkerboard microstructure, with material 1 in the white squares and material 2 in the black ones. Then the choice of function space amounts to a very concrete decision: do we require the electric potential to be continuous at the corners, or do we permit it to be discontinuous? Indeed, if we require $\int |\nabla \Phi|^{\alpha_2} \, dV$ to be finite then Φ must be continuous, by the Sobolev embedding lemma, since $\alpha_2 > 2$. But if we only require $\nabla \Phi$ to have finite energy then Φ can be discontinuous—its behavior near a corner can be as shown in Fig. 1.

Jikov has shown that we may constrain Φ to be continuous when minimizing (21). This leads to a *different* notion of effective energy than that obtained using discontinuous potentials with finite-energy. In particular, the two alternative choices lead to different solutions of the field equations.

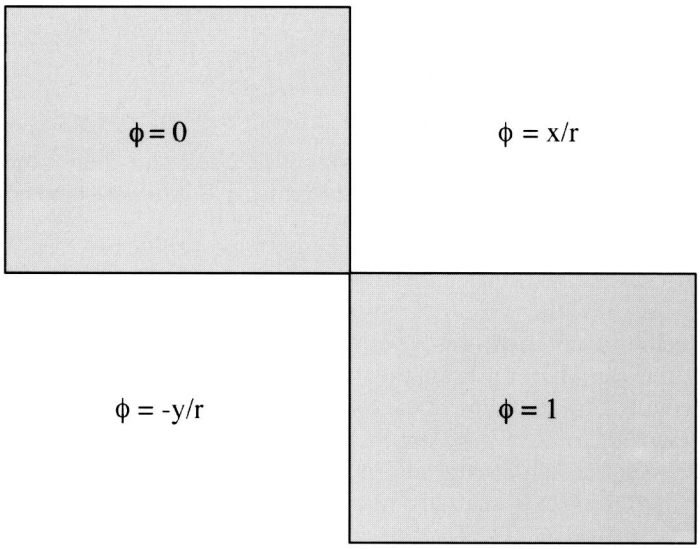

Fig. 1. A discontinuous, finite-energy $\nabla\Phi$, near the corner $(x, y) = (0, 0)$. In the two quadrants occupied by material 2 it takes the values 0 and 1 respectively. In the two quadrants occupied by material 1 it takes the values x/r and $-y/r$ respectively.

The same issue arises, of course, for the complementary variational principle (22). A more restrictive choice of function space for E naturally corresponds to a less restrictive choice of function space for J. Thus if we require the electric potential Φ to be continuous at a corner we should permit the current potential Ψ to be discontinuous, and vice versa.

It is natural to ask which solution is the "correct" one. This is a question of physics, not mathematics; we know no example where the answer is clear. One might hope that approximation would lead to insight. For example, our checkerboard has $U = |E|^{\alpha(x, y)}$ with $\alpha(x, y)$ discontinuous; one might hope that by smoothing the exponent slightly we could get an unambiguous definition of effective behavior. Alas, this does not work: similar function space issues arise even when the exponent is smooth, with the saddle points of $\alpha(x, y)$ playing the role of the "corners."

We shall avoid such function-space issues in the rest of this paper by considering mainly composites for which (24) holds.

3. DUALITY RESULTS FOR NONLINEAR COMPOSITE MEDIA

This section considers specific types of physically relevant nonlinearities, and explores the consequences of duality in each case. Following

ref. 15, we focus on three different classes of nonlinear behavior: (a) the weakly nonlinear regime, where attention is on small nonlinear corrections to predominantly linear response and the related phenomenon of flicker noise; (b) power-law behavior, where the conductivity is proportional to $|E|^\beta$ for some β; and (c) dielectric breakdown, where the conductivity vanishes for $|E|$ less then a critical value then increases linearly thereafter.

Our main focus is on behavior near percolation. The primal and dual composites describe different types of percolation problems—in the Ohmic setting, for example, if the primal mixes a normal conductor with a perfect insulator then the dual mixes a normal conductor with a perfect conductor. Thus duality couples the percolation exponents and scaling functions of physically different systems.

We shall consider mainly systems which are locally and macroscopically isotropic. Thus we use the scalar form of the duality relation (13) rather than its tensor generalization (18).

3.1. Weakly Nonlinear Composites

By a "weakly nonlinear" composite we really mean perturbation theory around the linear regime. It is therefore sufficient to keep the first nonlinear term in the dependence of J on E. We suppose that locally

$$J = \sigma E + b\,|E|^\beta E \tag{27}$$

with

$$b\,|E|^\beta \ll \sigma \tag{28}$$

All isotropic conductors with inversion symmetry (centrosymmetric crystal structure) exhibit such behavior, with $\beta = 2$, for an appropriate range of applied fields. Evidently σ represents the Ohmic conductivity and b is the coefficient of nonlinearity. We permit σ and b to vary from component to component, but we suppose all components have the same exponent β. The hypothesis of weak nonlinearity (28) requires that the applied field be sufficiently small.

The weakly nonlinear bulk behavior of an isotropic composite made from such materials has again the form (27), with the same exponent and effective coefficients σ_e and b_e:[15, 18]

$$\langle J \rangle = \sigma_e \langle E \rangle + b_e\,|\langle E \rangle|^\beta \langle E \rangle \tag{29}$$

This relation describes the nonlinear behavior of the composite, to first order when (28) holds. The effective coefficients have explicit formulas:

$$\sigma_e = \frac{1}{V} \int \sigma(r) \frac{|E_l(r)|^2}{|E_0|^2} dV \tag{30}$$

and

$$b_e = \frac{1}{V} \int b(r) \frac{|E_l(r)|^{\beta+2}}{|E_0|^{\beta+2}} dV \tag{31}$$

where $E_l(r)$ is the local electric field associated with applied field E_0 in the corresponding linear composite (with the same Ohmic conductivity in each components and the same microgeometry but $b = 0$).[18] Besides describing the weakly nonlinear behavior, these bulk moduli also have another interpretation when $\beta = 2$: then the ratio b_e/σ_e^2 determines the total power of the conductance fluctuation noise (known as flicker noise or $1/f$-noise) in the composite.[15, 18]

We now examine the consequences of duality. Consider a two-component, weakly nonlinear composite in which the local conductivities are

$$\sigma_1(|E|) = \sigma_1 + b_1 |E|^\beta \tag{32}$$

in one component and

$$\sigma_2(|E|) = \sigma_2 + b_2 |E|^\beta \tag{33}$$

in the other. The dual composite is another two-component composite with the same microgeometry. Keeping only the lowest order terms in b, we find the weakly nonlinear behavior of its component materials:

$$\sigma_1(|J|) = \frac{1}{\sigma_1(|E(J)|)} = \frac{1}{\sigma_1} - \frac{b_1}{\sigma_1^{\beta+2}} |J|^\beta \tag{34}$$

and

$$\sigma_2(|J|) = \frac{1}{\sigma_2(|E(J)|)} = \frac{1}{\sigma_2} - \frac{b_2}{\sigma_2^{\beta+2}} |J|^\beta \tag{35}$$

The effective behavior of these two composites can be expressed as

$$\sigma^*(\sigma_1(|E|), \sigma_2(|E|); E_0) = \sigma_e + b_e E_0^\beta$$

and

$$\sigma_d^*(\sigma_1(|J|), \sigma_2(|J|); J_0) = \sigma_{e,d} + b_{e,d} J_0^\beta$$

where

$$J_0 = \sigma^*(\sigma_1(|E|), \sigma_2(|E|); E_0)\, E_0 \tag{36}$$

is the magnitude of the current flowing through the primal composite, which is also the magnitude of the volume-averaged "electric field" in the dual composite. According to the duality relation (13), these effective conductivities satisfy

$$\sigma_e + b_e E_0^\beta = \frac{1}{\sigma_{e,d} + b_{e,d} J_0^\beta} = \frac{1}{\sigma_{e,d}} - \frac{b_{e,d}}{\sigma_{e,d}^2} J_0^\beta \tag{37}$$

to first order in the local nonlinearity coefficient b. This leads to

$$\sigma_e = \frac{1}{\sigma_{e,d}} \tag{38}$$

which is the well known duality result for linear composite materials, and

$$b_e E_0^\beta = -\frac{b_{e,d}}{\sigma_{e,d}^2} J_0^\beta$$

Substituting J_0 from (36), we find a new duality result for the bulk effective nonlinearity coefficients:

$$\frac{b_e}{\sigma_e^\beta} = -\frac{b_{e,d}}{\sigma_{e,d}^2} \tag{39}$$

In the special case $\beta = 2$ this gives a relation for the flicker-noise coefficients:

$$\frac{b_e}{\sigma_e^2} = -\frac{b_{e,d}}{\sigma_{e,d}^2} \tag{40}$$

We turn now to the consequences for percolation. The flicker-noise coefficient of a high-contrast composite is expected to have a characteristic power-law behavior near the percolation threshold.[29-32] When mixing a good conductor with a perfect insulator we expect

$$\frac{b_e}{\sigma_e^2} \approx \frac{b_M}{\sigma_M^2} \Delta p^{-\kappa} \tag{41}$$

above the percolation threshold of the good conductor. Here σ_M and b_M describe the good conductor; $\Delta p = |p - p_c|$ where p_c is the percolation threshold; and κ is a critical exponent. Similarly, when mixing a poor conductor with a perfect conductor we expect

$$\frac{b_e}{\sigma_e^2} \approx \frac{b_I}{\sigma_I^2} \Delta p^{-\kappa'} \tag{42}$$

above the percolation threshold of the poor conductor. Here σ_I and b_I describe the poor conductor, $\Delta p = |p - p_c|$ where p_c is the percolation threshold; and κ' is a critical exponent.[3] These are nonlinear analogues of the familiar scaling laws from the percolation theory of two-component Ohmic composites:

$$\sigma_e \approx \sigma_M \Delta p^t \qquad \text{or} \qquad \sigma_e \approx \sigma_I \Delta p^{-s} \tag{43}$$

for conductor—insulator mixtures and superconductor—normal conductor mixtures respectively. (Of course $t = s$ in the present two-dimensional setting, as a consequence of the linear duality result (38).)

Returning to our earlier notation (32)–(33), we take material 2 to be a perfect insulator ($\sigma_2 = 0$). Then (41) becomes

$$\frac{b_e}{\sigma_e^2} \approx \frac{b_1}{\sigma_1^2} \Delta p^{-\kappa}$$

where $\Delta p = p_1 - p_c > 0$ with p_c the percolation threshold of material 1. The dual composite is a mixture of a normal conductor described by (34) and a perfect conductor ($1/\sigma_2 = \infty$), and (42) gives

$$\frac{b_{e,d}}{\sigma_{e,d}^2} \approx -\frac{b_1}{\sigma_1^2} \Delta p^{-\kappa'}$$

Substituting these two expressions in the duality result (40) we conclude that the critical exponents describing the behavior above and below the threshold are the same:

$$\kappa = \kappa' \tag{44}$$

This result was previously obtained for random resistor networks by Wright *et al.*[32] They used the links-nodes-blobs model of the percolation

[3] We do not mean to claim that (41) and (42) hold for every continuum percolation problem. Rather, our assertion is that if they are valid then duality imposes certain restrictions on the exponents κ, κ'.

cluster and the duality relation for square linear networks to study the critical behavior of the resistance fluctuations above and below percolation. By using the nonlinear duality relation (13) we have shown that this relation is also valid for continuum composites of arbitrary microgeometry, subject only to the existence of scaling laws of the form (41) and (42).

This discussion can be easily extended to composites where the Ohmic conductivity ratio is finite. Following standard practice in percolation theory, we assume a law for the effective noise coefficient near the percolation threshold which interpolates between the two extremes considered so far:[33, 34]

$$\frac{b_e}{\sigma_e^2} \approx \frac{b_I}{\sigma_I^2} \Delta p^{-\kappa'} \mathscr{F}_I(z) + \frac{b_M}{\sigma_M^2} \Delta p^{-\kappa} \mathscr{F}_M(z) \tag{45}$$

where the Ohmic conductivity of the poor conductor is σ_I and that of the good conductor is σ_M. The scaling functions $\mathscr{F}_I$ and $\mathscr{F}_M$ are evaluated at $z = (\sigma_I/\sigma_M)/\Delta p^{t+s}$ with t and s determined by (43). In writing (45) we employ the usual abuse of notation: there are, in fact, *two pairs* of scaling functions, $\mathscr{F}_M^+$, $\mathscr{F}_I^+$ and $\mathscr{F}_M^-$, $\mathscr{F}_I^-$, one to be used when the good conductor percolates, the other to be used when the poor conductor percolates. Returning to our usual notation (32)–(33), we consider the case when $\sigma_1 \gg \sigma_2 > 0$. Then the bulk effective noise coefficient of the primal composite is

$$\frac{b_e}{\sigma_e^2} \approx \frac{b_2}{\sigma_2^2} \Delta p^{-\kappa'} \mathscr{F}_I(z) + \frac{b_1}{\sigma_1^2} \Delta p^{-\kappa} \mathscr{F}_M(z)$$

with $z = (\sigma_2/\sigma_1)/\Delta p^{t+s}$. Arguing similarly for the dual composite, its bulk effective noise coefficient is

$$\frac{b_{e,d}}{\sigma_{e,d}^2} \approx -\frac{b_1}{\sigma_1^2} \Delta p^{-\kappa'} \mathscr{F}_I^{(d)}(z) - \frac{b_2}{\sigma_2^2} \Delta p^{-\kappa} \mathscr{F}_M^{(d)}(z)$$

with the same definition of z. Substituting these formulas into the duality relation (40) we find, in addition to the equality (44), two equalities between the four scaling functions:

$$\mathscr{F}_M(z) = \mathscr{F}_I^{(d)}(z) \qquad \text{and} \qquad \mathscr{F}_I(z) = \mathscr{F}_M^{(d)}(z) \tag{46}$$

These relations must be interpreted appropriately: when the LHS uses $\mathscr{F}^+$ the RHS uses $\mathscr{F}^{(d)-}$, and vice versa.

3.2. Power Law Composites

The simplest "strong nonlinearity" is a power law relation between the local electric current and the local electric field:

$$J = \sigma |E|^{\beta} E \tag{47}$$

where the "nonlinear conductivity" σ is constant and $\beta > -1$. Such behavior is observed in certain classes of conductors including ZnO ceramics.[15] More generally, it describes the response of a material when the magnitude of the applied field is very large, so that the linear approximation breaks down completely.

We consider composites in which the nonlinear conductivity σ varies from component to component but the exponent β is the same for each component. The effective behavior of such a composite is again of power-law type, with the same exponent β. Therefore the composite is characterized by its effective nonlinear conductivity σ_e:[15]

$$\langle J \rangle = \sigma_e |\langle E \rangle|^{\beta} \langle E \rangle \tag{48}$$

where the angular brackets denote volume averaging and $\langle E \rangle = E_0$ is the externally applied electric field. There is a general formula for σ_e:

$$\sigma_e = \frac{1}{V} \int \sigma(r) \left| \frac{E(r)}{E_0} \right|^{\beta+2} dV$$

There are a few simple microgeometrics for which σ_e can be given explicitly.[20] Other approaches to its calculation include effective medium theories[35, 36] and a perturbation analysis around the case of a homogeneous medium.[19]

We now examine the consequences of duality. The relations we obtain should be satisfied by any physically reasonable mean field theory. Consider a two-component, strongly nonlinear composite in which the local conductivities are

$$\sigma_1(|E|) = \sigma_1 |E|^{\beta} \tag{49}$$

in the first component and

$$\sigma_2(|E|) = \sigma_2 |E|^{\beta} \tag{50}$$

in the second. The dual composite is another two-component composite with the same microgeometry. Its components have local conductivity laws

$$\sigma_1(|J|) = \frac{1}{\sigma_1(|E(J)|)} = \sigma_1^{-1/(\beta+1)} |J|^\gamma \tag{51}$$

in the first component and

$$\sigma_2(|J|) = \frac{1}{\sigma_2(|E(J)|)} = \sigma_2^{-1/(\beta+1)} |J|^\gamma \tag{52}$$

in the second, where

$$\gamma = -\frac{\beta}{\beta+1}, \qquad \text{or equivalently} \quad \gamma + 1 = \frac{1}{\beta+1} \tag{53}$$

The effective conductivities of these two composites can be expressed as

$$\sigma^*(\sigma_1(|E|), \sigma_2(|E|); E_0) = \sigma_e E_0^\beta$$

and

$$\sigma_d^*(\sigma_1(|J|), \sigma_2(|J|); J_0) = \sigma_{e,d} J_0^\gamma$$

where

$$J_0 = \sigma^*(\sigma_1(|E|), \sigma_2(|E|); E_0)\, E_0 \tag{54}$$

is the magnitude of the current flowing through the primal composite, which is also the magnitude of the volume-averaged "electric field" in the dual composite. According to the duality relation (13) these effective conductivities satisfy

$$\sigma_e E_0^\beta = \frac{1}{\sigma_{e,d} J_0^\gamma} \tag{55}$$

Substituting J_0 from (54), we obtain

$$\sigma_e^{1/(\beta+1)} = \frac{1}{\sigma_{e,d}} \tag{56}$$

This is the duality result for the nonlinear effective conductivities of composites made from power-law materials.

We turn now to the consequences for percolation. As before, we consider the high-contrast limit. When mixing a good conductor (nonlinear

conductivity σ_M, exponent β) with a perfect insulator (nonlinear conductivity $\sigma_I = 0$), we expect the nonlinear effective conductivity to behave as

$$\sigma_e \approx \sigma_M \, \Delta p^{t(\beta)} \tag{57}$$

above the percolation threshold of the good conductor.[37, 14] Similarly, when mixing a poor conductor (nonlinear conductivity σ_I, exponent β) with a perfect conductor (nonlinear conductivity $\sigma_M = \infty$) we expect

$$\sigma_e \approx \sigma_I \, \Delta p^{-s(\beta)} \tag{58}$$

below the percolation threshold of the perfect conductor. Here $t(\beta)$ and $s(\beta)$ are critical exponents, the power-law analogues of the familiar critical exponents for Ohmic composites (43).

Returning to our earlier notation (49)–(50), we take material 2 to be a perfect insulator ($\sigma_2 = 0$). Then (57) becomes

$$\sigma_e \approx \sigma_1 \, \Delta p^{t(\beta)}$$

when $\Delta p = p_1 - p_c > 0$, with p_c the percolation threshold of material 1. The dual composite is a mixture of a normal conductor described by (51) and a perfect conductor (since $\sigma_2^{-1/(\beta+1)} = \infty$), and (58) gives

$$\sigma_{e,d} \approx \sigma_1^{-1/(\beta+1)} \, \Delta p^{-s(\gamma)}$$

Substituting these two expressions in the duality result (56) we find a relation between the two critical exponents $s(\gamma)$ and $t(\beta)$:

$$t(\beta) = (\beta+1)\, s(\gamma) = (\beta+1)\, s\left(-\frac{\beta}{\beta+1}\right) \tag{59}$$

When $\beta = \gamma = 0$ this reduces to the well-known result $t = s$ for two-dimensional Ohmic composites. The relation (59) was previously obtained by Straley and Kenkel for strongly nonlinear square random resistor networks.[14] By using the nonlinear duality relation (13) we have shown that it also applies for continuum composites of arbitrary microgeometry, subject only to the existence of scaling laws of the form (57) and (58).

When the conductivity ratio is finite, the behavior described by (57) and (58) breaks down for Δp near 0, since σ_e cannot vanish or become infinite. Instead, we expect the nonlinear effective conductivity to satisfy a law of the form[38]

$$\sigma_e \approx \sigma_M \, \Delta p^{t(\beta)} \mathcal{F}_\beta(z) \tag{60}$$

with

$$z = \frac{\sigma_I/\sigma_M}{\Delta p^{t(\beta)+s(\beta)}}$$

This applies near the percolation threshold, for mixtures of two materials with nonlinear conductivities $\sigma_M \gg \sigma_I$ and the same exponent β. The function $\mathscr{F}_\beta(z)$ takes into account the effect of having a finite conductivity ratio σ_I/σ_M, while the prefactor is determined by the limiting case (57). In writing (60) we adopt the usual abuse of notation: in truth there are *two* scaling functions, $\mathscr{F}_\beta^+$ and $\mathscr{F}_\beta^-$, one to be used when the good conductor percolates, the other to be used when the poor conductor percolates. Returning to our usual notation (49)–(50), we find that when $\sigma_1 \gg \sigma_2 > 0$ the primal composite has nonlinear effective conductivity

$$\sigma_e \approx \sigma_1 \Delta p^{t(\beta)} \mathscr{F}_\beta \left(\frac{\sigma_2/\sigma_1}{\Delta p^{t(\beta)+s(\beta)}} \right)$$

The dual has local nonlinear conductivities $\sigma_1^{-1/(\beta+1)} \ll \sigma_2^{-1/(\beta+1)}$ and exponent γ, so its nonlinear effective conductivity is

$$\sigma_{e,d} \approx \sigma_2^{-1/(\beta+1)} \Delta p^{t(\gamma)} \mathscr{F}_\gamma \left(\frac{(\sigma_2/\sigma_1)^{1/(\beta+1)}}{\Delta p^{t(\gamma)+s(\gamma)}} \right) \tag{61}$$

Substituting these relations into (56) we find, in addition to the relation (59) between the critical exponents, an explicit relation between the two scaling functions $\mathscr{F}_\beta$ and $\mathscr{F}_\gamma = \mathscr{F}_{-\beta/(\beta+1)}$:

$$\mathscr{F}_{-\beta/(\beta+1)}(z^{1/(\beta+1)}) = \left[\frac{z}{\mathscr{F}_\beta(z)} \right]^{1/(\beta+1)} \tag{62}$$

This relation must be interpreted appropriately: when the LHS uses $\mathscr{F}^+_{-\beta/(\beta+1)}$ the RHS uses $\mathscr{F}_\beta^-$, and vice versa.

These exact relations can be used to test the reasonableness of any mean field theory for strongly nonlinear composites near the percolation threshold. Two such theories have been proposed recently, by Bergman[35] and by Wan *et al.*[36] Bergman's theory predicts

$$t(\beta) = \beta + 1 \qquad \text{and} \qquad s(\beta) = 1 \tag{63}$$

while that of Wan *et al.* predicts

$$t(\beta) = s(\beta) = \frac{\beta}{2} + 1 \tag{64}$$

In both cases the critical exponents satisfy the duality relation (59). It would be interesting to know whether these theories also satisfy the additional requirement (62).

3.3. Dielectric Breakdown and Critical Current

We turn to a different type of nonlinearity—or more precisely, a pair of types, dual to one another. It can be viewed as a simple model of dielectric breakdown. There is also an interpretation involving current-induced destruction of superconductivity. Either way, this nonlinearity differs from those considered above because the relationship between E and J changes abruptly at a certain critical field.

We concentrate first on dielectric breakdown. In this phenomenon, the material is a perfect insulator until the magnitude of the electric field reaches a critical value E_{cr}, when it begins to conduct (see Fig. 2a). The associated nonlinear conductivity law is

$$J(E) = \begin{cases} 0 & |E| \leqslant E_{cr} \\ \sigma(|E| - E_{cr}) \dfrac{E}{|E|} & |E| > E_{cr} \end{cases} \tag{65}$$

For non-smooth laws such as this it is convenient to consider the associated convex potential, defined by $J = \partial U/\partial E$. One verifies that

$$U(E) = \tfrac{1}{2}\sigma(|E| - E_{cr})_+^2$$

with the convention $t_+ = \max\{t, 0\}$. Its convex dual is

$$\hat{U}(J) = \max_E \{J \cdot E - U(E)\} = |J| E_{cr} + \frac{1}{2\sigma}|J|^2$$

which is non-differentiable at $J = 0$. Therefore the inverse of (65), the law for E in terms of J, takes the form

$$E = \frac{\partial \hat{U}}{\partial J} = \begin{cases} \text{undetermined, of magnitude} \leqslant E_{cr} & J = 0 \\ E_{cr} \dfrac{J}{|J|} + \dfrac{1}{\sigma} J & J \neq 0 \end{cases} \tag{66}$$

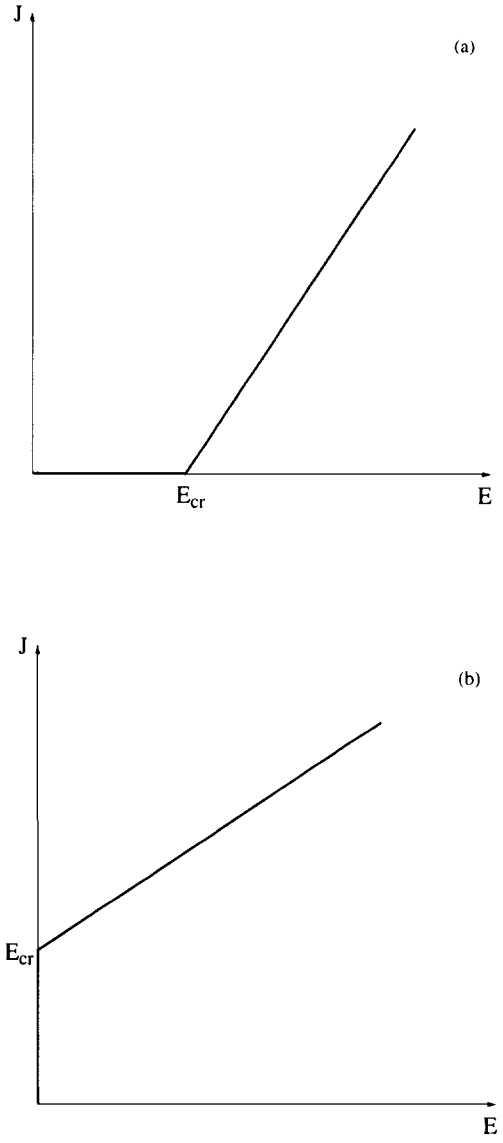

Fig. 2. Current vs. electric field curves for materials with a breakdown type nonlinearity: (a) Dielectric breakdown at a threshold field E_{cr}. (b) Breakdown of perfect conductivity at a critical current E_{cr}.

Current-induced destruction of superconductivity can be modeled very similarly.[39] In this setting the material is a perfect conductor until the magnitude of the current field reaches a critical value J_{cr}, when it develops nonzero resistance. This is described by the nonlinear conductivity law

$$E(J) = \begin{cases} 0 & |J| \leqslant J_{cr} \\ \rho(|J| - J_{cr}) \dfrac{J}{|J|} & |J| > J_{cr} \end{cases} \tag{67}$$

We emphasize that (67) has the same form as (65) but with J and E interchanged. Its inverse, the law giving J in terms of E in a superconductor, is therefore analogous to (66):

$$J(E) = \begin{cases} \text{undetermined, of magnitude} \leqslant J_{cr} & E = 0 \\ J_{cr} \dfrac{E}{|E|} + \dfrac{1}{\rho} E & E \neq 0 \end{cases} \tag{68}$$

When we pass from a primal composite to the associated dual composite, we treat $J^{\perp}$ as an "electric field" and $E^{\perp}$ as a "current field." Suppose each material in the primal composite satisfies a dielectric breakdown law of the form (65), with $\sigma(r)$ and $E_{cr}(r)$ varying from component to component: Then from (66) we have the local relation (Fig. 2b)

$$E^{\perp} = \begin{cases} \text{undetermined, of magnitude} \leqslant E_{cr} & J^{\perp} = 0 \\ E_{cr} \dfrac{J^{\perp}}{|J^{\perp}|} + \dfrac{1}{\sigma} J^{\perp} & J^{\perp} \neq 0 \end{cases}$$

Thus, each component of the dual composite is a superconductor, with critical current $E_{cr}(r)$ and resistivity $1/\sigma(r)$.

We turn now to the consequences of duality. Consider a two component composite mixing a material of dielectric-breakdown type with an Ohmic conductor. We suppose material 1 is described by (65) with $\sigma = \sigma_1$, and material 2 has Ohmic conductivity σ_2. We suppose furthermore that the Ohmic material does not percolate. Then the composite exhibits dielectric-breakdown type behavior: it behaves as a perfect insulator when the applied field E_0 is below a critical value $E_{0,cr}$. At this critical applied field the conductive region—consisting of material 2 and the part of material 1 which has undergone breakdown—begins to percolate, and the composite begins to conduct. The effective behavior of the composite is not as simple

as (65), because the average current J_0 will not depend linearly on E_0 for $|E| > E_{0,cr}$. Instead the effective law has the form

$$J_0(E_0) = \begin{cases} 0 & |E_0| \leqslant E_{0,cr} \\ \sigma^*(|E_0|)\, E_0 & |E_0| > E_{0,cr} \end{cases}$$

for some nonlinear function $\sigma^*(t)$ defined for $t \geqslant E_{0,cr}$ with $\sigma^*(E_{0,cr}) = 0$.

The dual composite mixes a superconductor with an Ohmic conductor: In material 1 the current field is given as a function of the electric field by (68) with $J_{cr} = E_{cr}$ and $[\rho = \sigma_1]$. In material 2 the Ohmic conductivity is $1/\sigma_2$. The superconductor percolates. Therefore, arguing as above (but applying a current rather than an electric field) we expect the composite to behave once again like a superconductor, with an effective critical current $J^{(d)}_{0,cr}$. As before, the effective law is not as simple as (68); instead it has the form

$$J_0(E_0) = \begin{cases} \text{undetermined, of magnitude} \leqslant J^{(d)}_{0,cr} & E_0 = 0 \\ \sigma^*_d(|E_0|)\, E_0 & E_0 \neq 0 \end{cases}$$

for some nonlinear function $\sigma^*_d(t)$ defined for $t > 0$ with $t\sigma^*_d(t) \to J^{(d)}_{0,cr}$ as $t \to 0$.

We claim, as a consequence of duality, that these two composites—one made from an insulator capable of dielectric breakdown, the other made from a superconductor—have the same critical fields:

$$E_{0,cr} = J^{(d)}_{0,cr} \tag{69}$$

This is clear from Section 2, since the current field in the dual composite and the electric field in the primal composite are identical up to rotation by 90°. An alternative derivation is provided by the duality relation (13) linking σ^* and σ^*_d. It says

$$\sigma^*(|E_0|)\, \sigma^*_d(|J_0|) = 1$$

with $J_0 = \sigma_*(|E_0|)\, E_0$. This amounts to the statement that

$$\sigma^*(t)\, \sigma^*_d(\sigma^*(t)\, t) = 1$$

for all $t > E_{0,cr}$. Therefore

$$1 = \lim_{t \to E_{0,cr}} \frac{\sigma^*_d(\sigma^*(t)t)\, \sigma^*(t)t}{t} = \frac{\lim_{s \to 0} \sigma^*_d(s)\, s}{E_{0,cr}} = \frac{J^{(d)}_{0,cr}}{E_{0,cr}}$$

confirming the conclusion (69).

As usual, this relation has consequences for the scaling laws near percolation. Our primal composite is a mixture of an Ohmic material and an insulator capable of dielectric breakdown. Suppose it is random, with fraction p of Ohmic conductor. As $p \to p_c$ and the Ohmic conductor percolates, the critical field of the composite tends to 0. It is standard in percolation theory to assume a characteristic scaling law:[40, 41]

$$E_{0,cr} \propto \Delta p^{y}$$

Our dual composite is a mixture of an Ohmic material and a superconductor. It is also random, with the same microgeometry and the same p_c. Its critical field also tends to 0 as $p \to p_c$, and we expect a characteristic scaling law:[41]

$$J_{0,cr} \propto \Delta p^{v}$$

Clearly, as a consequence of duality, the two exponents are the same:

$$v = y \tag{70}$$

Lobb *et al.*[41] previously derived this result for linear square random resistor networks. They also derived it for the "swiss cheese–inverse swiss cheese" version of continuum percolation via a links-nodes-blobs model of the percolation cluster. Our more general derivation shows that this relation does not depend on a specific model for continuum percolation; it holds in complete generality, as a consequence of the general relation (69).

3.4. Mixed and Self-Dual Composites

In Section 3.2 the "growth law" of J versus E was assumed to be homogeneous: $|J| \propto |E|^{\beta+1}$ with a uniform value of β throughout the composite. Therefore the function space issues raised in Section 2.3 were not relevant. However it is also of interest to consider some examples with inhomogeneous growth laws.

Consider a composite consisting of nonlinear inclusions embedded in a linear host. Such "mixed" composites are interesting because their nonlinearity may be strongly enhanced relative to bulk samples of the nonlinear component.[42–47] Because of the matrix-inclusion structure there are no multigrain junctions, so we do not expect the ambiguities discussed in Section 2.3 to arise. A mean field theory was recently developed, extending the Maxwell Garnett theory to composites with inclusions of arbitrary nonlinearity.[25, 26] Duality places certain restrictions upon the form of any

mean field theory. We now show that the nonlinear Maxwell Garnett theory passes this test.

According to refs. 25 and 26 the effective conductivity function of a two-dimensional mixture of cylindrical inclusions is given by

$$\frac{\sigma_e(E_0)-\sigma_h}{\sigma_e(E_0)+\sigma_h}=p_i\frac{\sigma_i(E_i)-\sigma_h}{\sigma_i(E_i)+\sigma_h} \tag{71}$$

where σ_h is the Ohmic conductivity of the host, $\sigma_i(E_i)$ is the field dependent conductivity of the inclusions, p_i is the volume fraction of the inclusions, E_0 is the volume averaged applied field, and E_i is the field inside the inclusions, which is determined by

$$(\sigma_i(E_i)-\sigma_h)\,p_hE_i+2\sigma_hE_i=2\sigma_hE_0$$

The dual composite has nonlinear inclusions with conductivity $1/\sigma_i(E_i(J_i))$ embedded in a linear host with conductivity $1/\sigma_h$. The effective conductivity of the dual composite, calculated by the same nonlinear Maxwell Garnett theory, is

$$\frac{\sigma_{e,d}(J_0)-\dfrac{1}{\sigma_h}}{\sigma_{e,d}(J_0)+\dfrac{1}{\sigma_h}}=p_i\frac{\dfrac{1}{\sigma_i(J_i)}-\dfrac{1}{\sigma_h}}{\dfrac{1}{\sigma_i(J_i)}+\dfrac{1}{\sigma_h}} \tag{72}$$

where J_i, the local current inside the inclusions, is determined by

$$\left(\frac{1}{\sigma_i(J_i)}-\frac{1}{\sigma_h}\right)p_hJ_i+2\frac{1}{\sigma_h}J_i=2\frac{1}{\sigma_h}J_0$$

and $J_0=\sigma_e(E_0)\,E_0$ is the total current flowing through the original composite. Comparing (71) and (72) we see that

$$\sigma_e(E_0)=\frac{1}{\sigma_{e,d}(J_0)}$$

Thus the nonlinear Maxwell Garnett theory obeys the duality relation (13), as it should.

We turn finally to the topic of "self-dual" composites, focusing as usual on the isotropic case. A composite is said to be self-dual if it is statistically indistinguishable from its dual. As noted by Kozlov,[13] such a composite must have Ohmic effective behavior—with conductivity 1—no matter how

nonlinear its local response. The proof is simple: from (13) we know that $\sigma^*(|E_0|)\,\sigma_d^*(|J_0|)=1$ with $J_0=\sigma^*(|E_0|)\,E_0$, and the hypothesis of self-duality says $\sigma_d^*=\sigma^*$. It follows that

$$\sigma^*(t)\,\sigma^*(t\sigma^*(t))=1 \tag{73}$$

for every $t\geqslant 0$. Multiplying by t we get

$$j(j(t))=t \tag{74}$$

where $j(t)=t\sigma^*(t)$. Our convexity hypothesis (9) says that $t\mapsto j(t)$ is monotone; (74) says that j is its own inverse; and our goal is to show that $j(t)=t$ for all t. Indeed, fix any t_0 and let $s_0=j(t_0)$. From (74) we have $j(s_0)=t_0$, whence

$$(t_0-s_0)(j(t_0)-j(s_0))=-(t_0-s_0)^2\leqslant 0$$

However monotonicity requires

$$(t-s)(j(t)-j(s))\geqslant 0 \qquad \text{for all } t, s \tag{75}$$

It follows that $s_0=t_0$, i.e. $j(t_0)=t_0$, as desired.

When is a two-component composite self-dual? If the materials in the primal composite have nonlinear conductivities $\sigma_i(|E|)$, $i=1,2$, then the materials in the dual composite have nonlinear conductivities

$$\sigma_{i,d}(|J|)=\frac{1}{\sigma_i(|E|\,(|J|))}$$

So the composite is self-dual if

$$\sigma_1(|E|)=\frac{1}{\sigma_2(E(|J|))}=\sigma_2^{(d)}(|J|) \tag{76}$$

and

$$\sigma_2(|E|)=\frac{1}{\sigma_1(E(|J|))}=\sigma_1^{(d)}(|J|) \tag{77}$$

and the microstructure is statistically invariant under interchange of materials 1 and 2. If the function-space issues of Section 2.3 are relevant—e.g. if the composite has multigrain junctions—then the continuity conventions must also be statistically invariant.

The following simple example of a self-dual nonlinear composite is due to Kozlov[13] and Jikov *et al.*[12] Consider a periodic checkerboard in

which the white squares have conductivity $\sigma_1(|E|) = \sigma |E|^\beta$ and the black squares have conductivity $\sigma_2(|E|) = \sigma^{-1/(\beta+1)} |E|^\gamma$ with γ defined by (53). We resolve the ambiguity discussed in Section 2.3 by requiring the electric potential to be continuous at alternate corners, with the current potential continuous at the remaining corners. The dual composite is another periodic checkerboard, with conductivity $\sigma_{1,d}(|J|) = \sigma^{-1/(\beta+1)} |J|^\gamma$ in the white squares and $\sigma_{2,d}(|J|) = \sigma |J|^\beta$ in the black squares. Like the primal, its electric potential is continuous at alternate corners, as is its current potential. Thus the composite is self-dual. It is not manifestly isotropic, since 90° symmetry implies isotropy only in the linear setting, so strictly speaking the analysis given above does not apply. But our analysis is easily adapted, using (18) in place of (13), and making use of the 90° symmetry. Relation (74) is replaced by the assertion that $E \mapsto J(E) = \sigma^*(E) E$ is its own inverse. Monotonicity (75) is replaced by the assertion that

$$(\xi - \eta) \cdot (J(\xi) - J(\eta)) \geqslant 0 \qquad \text{for all vectors} \quad \xi, \eta$$

The conclusion is as before: $J(E) \equiv E$, i.e. the effective behavior of the self-dual periodic checkerboard is Ohmic (and isotropic) with conductivity 1.

The same two materials can also be combined to form other self-dual composites. For example, consider the "cell material" obtained by packing space randomly with non-overlapping circles of all sizes, then filling each circle with material 1 or 2 by independent flips of an unbiased coin. We conjecture that the ambiguities of Section 2.3 do not arise in this case. Then the composite is clearly statistically interchangeable and self-dual.

Another, quite different example arises in the weakly nonlinear context, when the local conductivities are

$$\sigma_1(|E|) = \sigma + b |E|^\beta \tag{78}$$

in one component, and

$$\sigma_2(|J|) = \frac{1}{\sigma_1(|E(J)|)} = \frac{1}{\sigma} - \frac{b}{\sigma^{\beta+2}} |J|^\beta \tag{79}$$

in the other. If in addition the two components are statistically interchangeable then the weakly nonlinear effective behavior is Ohmic, i.e. the effective coefficient of weak nonlinearity b_e, defined by (31), vanishes. This implies the following simple relation between the average electric fields in the two components:

$$\langle |E|^{\beta+2} \rangle_1 = \frac{1}{\sigma^{\beta+1}} \langle |E|^{\beta+2} \rangle_2 \tag{80}$$

where the angular brackets denote volume averaging over the individual components and we have used the fact that each component has area fraction 1/2.

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