ELECTRICAL TRANSPORT IN HIGH CONTRAST COMPOSITE MATERIALS

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A broad range of problems in the physics of materials involve highly disordered media whose effective behavior depends critically on the connectedness, or percolation properties of a particular component. Examples include smart materials such as piezoresistors and thermistors, smart insulators, radar absorbing composites, cermets, porous media, doped semiconductors, thin metal films, and sea ice. In numerous composite and smart materials, the microstructure can be characterized by conducting particles embedded in an insulating host, and one is interested in the effective DC conductivity (or complex permittivity for interactions with waves) near the critical volume fraction for percolation of the conducting phase. Such media frequently arise due to the desirability of light materials having the attractive mechanical properties of common polymers and the electrical conductivities of metals. In modeling transport in such materials, one often considers a two component random medium with component conductivities σ_1 and σ_2, in the volume fractions $1-p$ and p. The medium may be discrete, like the random resistor network, or continuous, like the random checkerboard and Swiss cheese models. In these systems, as $h = \frac{\sigma_1}{\sigma_2} \to 0$, the effective conductivity $\sigma^*(p,h)$ exhibits critical behavior near the percolation threshold p_c, $\sigma^*(p,0) \sim (p-p_c)^t$ as $p \to p_c^+$ (with $\sigma_1 = 0$ and $\sigma_2 = 1$), and at $p = p_c$, $\sigma^*(p_c,h) \sim h^{1/\delta}, h \to 0^+$.

In the lattice case of the random resistor network, it has been widely proposed that the scaling behavior of σ^* as a function of both p and h around $p = p_c$ and $h = 0$ is similar to a phase transition in statistical mechanics, like that exhibited by the magnetization $M(T,H)$ of an Ising ferromagnet around its Curie point at temperature $T = T_c$ and applied field $H = 0$. However, despite the numerous works based on this similarity, a rigorous understanding of it has been elusive. In the continuum, such as for the Swiss cheese model (a conducting host with random holes cut out), the situation is even more complex – while the underlying percolation exponents remain the same as for the lattice (and satisfy the standard scaling relations of statistical mechanics), the transport exponents, such as t in three dimensions, can be different from their lattice values [1]. For the random checkerboard in two dimensions, it has been argued [2] that the exponent δ is different from its lattice value, while the percolation exponents (and t) remain the same. These examples of non-universal behavior raise a fundamental question as to what features of the lattice problem remain true in the continuum.

In recent work [3] we have shown that although the critical exponents of transport in the continuum may be different from their lattice values, they still satisfy the standard scaling relations of statistical mechanics, as do their lattice counterparts. This is accomplished through a direct, analytic correspondence between transport in two component random media and the magnetization M of an Ising ferromagnet [4], which has been developed further and applied to critical behavior [3]. In particular, we obtain a new integral representation for $m = \sigma^*/\sigma_2$,

$$m(h) = 1 + (h-1)g(h), \quad g = \int_0^\infty \frac{d\phi(z)}{1+hz}, \qquad (1)$$

where g is a Stieltjes function of h, ϕ is a positive measure which for our percolation models with $p > p_c$ is supported only in $[0, S(p)]$, where $S(p) \sim (p-p_c)^{-\Delta}, p \to p_c{}^+$, and Δ is called the gap exponent. This formula is the direct analogue for transport of Baker's formula [5] for the magnetization M in the variable $\tau = \tanh(\beta H)$,

$$M(\tau) = \tau + \tau(1-\tau^2)G(\tau^2), \quad G = \int_0^\infty \frac{d\psi(z)}{1+\tau^2 z}, \qquad (2)$$

where G is a Stieltjes function of τ^2, ψ is a positive measure which for $T > T_c$ is supported only in $[0, S(T)]$, where $S(T) \sim (T-T_c)^{-2\Delta}, T \to T_c{}^+$, and Δ is a different gap exponent. These formulas make the connection of transport in random media to statistical mechanics almost transparent. Then, methods which have been used to analyze the critical behavior of the Ising model can be appropriately modified for transport in lattice and continuum percolation models to obtain

$$\delta = \frac{\Delta}{\Delta - \gamma}, \qquad t = \Delta - \gamma, \qquad (3)$$

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where γ is the susceptibility exponent defined by $\chi(p) = \frac{\partial m}{\partial h} \sim (p - p_c)^{-\gamma}, p \to p_c{}^+, h = 0$. These laws are satisfied by the analogous critical exponents for phase transitions in statistical mechanics.

The Stieltjes integral representation above has been used previously in a different form to obtain rigorous, general bounds on the effective complex permittivity ϵ^* of two component random media in the continuum [6, 7]. These bounds are obtained under statistical constraints on the microstructure, such as known volume fractions and statistical isotropy (Hashin-Shtrikman), and are valid in the quasistatic regime. For high contrast materials, the bounds are very broad, and give little practical information about the effective behavior. However, Bruno [8] found that if one assumes the condition that one phase is contained in separated inclusions in a matrix of the other material (matrix-particle assumption), then there is a gap in the support of the measure in the integral representation, and tighter versions of the Hashin-Shtrikman bounds in the case of real parameters can be obtained. In [9, 10] we have found complex versions of the fixed volume fraction and Hashin-Shtrikman bounds for matrix-particle composites. Even when the inclusions are fairly close to touching, characterized by a parameter $0 \leq q \leq 1$, which is 1 when the inclusions are allowed to touch, the new bounds give a dramatic improvement over the original complex bounds [6, 7], as illustrated in Figure 1.

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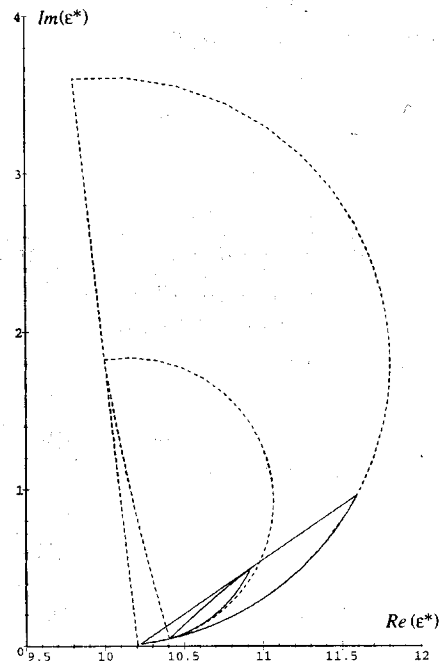

Figure 1: Bounds R_1 (outer, dotted), R_2 (inner, dotted), R_1^{mp} (outer, solid), and R_2^{mp} (inner, solid) on the complex permittivity ϵ^* of a PEO-PPY insulator-conductor composite. R_1 assumes only knowledge of the conductor volume fraction $p_1 = 0.02$, and R_2 assumes statistical isotropy as well. R_1^{mp} and R_2^{mp} further assume the material is a matrix-particle composite with $q = 0.9$. The complex permittivities of the components are $\epsilon_1 = 0 + i180$ and $\epsilon_2 = 10 + i0.00018$.