Critical Behavior of Transport in Lattice and Continuum Percolation Models

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It has been observed that the critical exponents of transport in the continuum, such as in the Swiss cheese and random checkerboard models, can exhibit nonuniversal behavior, with values different than the lattice case. Nevertheless, it is shown here that the transport exponents for both lattice and continuum percolation models satisfy the standard scaling relations for phase transitions in statistical mechanics. The results are established through a direct, analytic correspondence between transport coefficients for two component random media and the magnetization of the Ising model, which is based on the observation we made previously that both problems share the Lee-Yang property. [S0031-9007(97)03093-7]

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A broad range of problems in the physics of materials involve highly disordered media whose effective behavior is dominated by the connectedness, or percolation properties, of a particular component. Examples include porous media, doped semiconductors, smart materials such as piezoresistors and thermistors, radar absorbing composites, thin metal films, snow, and sea ice. 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 [1–3], or continuous, like the random checkerboard [4,5] and Swiss cheese models [1,3,6]. In these systems, as $h = \sigma_1/\sigma_2 \to 0$, 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 [2,3,7–11] that the scaling behavior of σ^* as a function of both p and h around $p = p_c$ and $h = 0$ (including crossover between the above laws), 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, this behavior of $\sigma^*(p, h)$ has been explicitly obtained only in mean-field theory around the critical dimension $d_c = 6$ [12], and in the effective medium approximation [3], although renormalization arguments in two and three dimensions have supported its validity [13,14]. This situation should be contrasted with that for the underlying percolation model, where its Kasteleyn-Fortuin [15] representation as the $q \to 1$ limit of the q-state Potts model makes clear the connection to phase transitions. Indeed, the critical exponents of percolation have been shown to obey the standard scaling relations of statistical mechanics [16,17]. Similar efforts to use the connection between the random resistor network and the $q \to 0$ Potts model to analyze σ^* when $h > 0$ have apparently been unsuccessful [12,18]. Nevertheless, for $h = 0$, a number of scaling laws relating t to percolation exponents have been proposed, such as the Alexander-Orbach conjecture, although none of them seems to be exactly true [1,2].

In the continuum, such as for the Swiss cheese model, while the percolation exponents remain the same as for the lattice [19], the transport exponents, such as t in three dimensions, can be different from their lattice values [6]. For the random checkerboard in two dimensions, it was argued in [5] that the exponent δ is different from its lattice value, while the percolation exponents (and t) remain the same. These examples of nonuniversal behavior raise a fundamental question as to what features of the lattice problem remain true in the continuum.

In this Letter, we show 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 an analytic correspondence between effective transport coefficients for two component random media and the magnetization M of an Ising ferromagnet. The correspondence is based on the observation that both problems share the Lee-Yang property, which was originally found in [20], but is developed further here and applied to critical behavior. In particular, we obtain a new Stieltjes integral representation for σ^* which is the direct analog of Baker's formula for M [21], making the connection to statistical mechanics almost transparent. Then, methods which have been used to analyze the critical behavior of the Ising model [21–23] can be appropriately modified for transport in lattice and continuum percolation models. We also further investigate the zeros of the conductivity partition function in the complex p plane introduced in [20].

To present our results, we briefly review the relevant theory for the nearest neighbor Ising model of a ferromagnet in a field H and at temperature T. When $H = 0$, the magnetization $M(T) = -\partial f/\partial H \sim (T_c - T)^\beta$ as $T \to T_c^-$, where f is the free energy per site, and the magnetic susceptibility $\chi = \partial M/\partial H = -\partial^2 f/\partial H^2 \sim (T - T_c)^{-\gamma}$ as $T \to T_c^+$. Along the critical isotherm $T = T_c$,

$M(H) \sim H^{1/\delta}$ as $H \to 0^+$ (where δ and the other exponents have different numerical values from their analogs in transport). Now, in 1952 Lee and Yang [24] found that the zeros of the partition function of the Ising ferromagnet (or lattice gas) lie on the unit circle in the z plane, where $z = \exp(-2\beta H)$ is the "activity," $\beta = 1/kT$, and k is Boltzmann's constant. Equivalently they lie on the imaginary axis in the H plane. For $T > T_c$, there is a gap θ_H in these zeros around $H = 0$, which collapses as $T \to T_c^+$, with $\theta_H \sim (T - T_c)^{\Delta}$. In the T plane the situation is more complicated, although in $d = 2$ for $H = 0$, "Fisher's zeros" lie on two circles in the complex $v = \tanh(2\beta J)$ plane [25], where J is the interaction strength. In 1968, Baker [21] used the Lee-Yang property to show that the magnetization has the following special analytic structure in the variable $\tau = \tanh(\beta H)$:

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

where G is a Stieltjes (or Herglotz) function of τ^2, and ψ is a positive measure which for $T > T_c$ is supported only in $[0, S(T)]$, where $S(T) \sim (T - T_c)^{-2\Delta}, T \to T_c^+$. Note that M is analytic throughout the τ^2 plane except $(-\infty, 0]$. This integral representation was used to obtain the scaling relations $\beta = \Delta - \gamma$ and $\delta = \Delta/(\Delta - \gamma)$ [22,23] (which are satisfied by the mean field exponents $\beta = \frac{1}{2}, \gamma = 1, \delta = 3, \Delta = \frac{3}{2}$, and the exact exponents for $d = 2, \beta = \frac{1}{8}, \gamma = 1\frac{3}{4}, \delta = 15, \Delta = 1\frac{7}{8}$), as well as *Baker's inequalities* $\gamma_{n+1} - 2\gamma_n + \gamma_{n-1} \geq 0$ for the critical exponents γ_n of the higher field derivatives of the free energy f, or equivalently, of the moments ψ_n of ψ, $\psi_n \sim (T - T_c)^{-\gamma_n}, T \to T_c^+$, with $\gamma_0 = \gamma$ [21]. The sequence γ_n is actually linear in n, $\gamma_n = \gamma + 2\Delta n, n \geq 0$, with constant gap $\gamma_i - \gamma_{i-1} = 2\Delta$ [21,26].

It is observed here that the Lee-Yang-Baker critical theory outlined above applies to transport problems, and in particular, we rigorously establish direct analogs of (1) and the associated scaling relations and inequalities for lattice and continuum percolation models of conduction in two component random media. Our results apply as well to electrical permittivity, magnetic permeability, thermal diffusivity, fluid permeability for Darcy flow in a porous medium, and effective diffusivity for turbulent transport [27], which all share the Lee-Yang property.

We now formulate the effective conductivity problem in general for two component random media in the continuum $\mathbf{R}^d$, which includes the lattice $\mathbf{Z}^d$ as a special case [20,28]. Let the local conductivity $\sigma(x, \omega) = \sigma_1\chi_1(x, \omega) + \sigma_2\chi_2(x, \omega)$ be a two-valued stationary random field in $x \in \mathbf{R}^d$ and $\omega \in \Omega$, where Ω is the set of realizations of the random medium, $\chi_1(x, \omega) = 1$ if x is in medium 1, and 0 otherwise, and $\chi_2 = 1 - \chi_1$. Let $E(x, \omega)$ and $J(x, \omega)$ be stationary random electric and current fields which are related by $J = \sigma E$ and satisfy $\nabla \cdot J = 0$ and $\nabla \times E = 0$, with $\langle E(x, \omega)\rangle = e_k$, where e_k is a unit vector, and $\langle\cdot\rangle$ denotes ensemble average over Ω, or an appropriate infinite volume limit. For the random resistor network the differential equations become difference equations (Kirchoff's laws). For isotropic media, the effective conductivity σ^* is defined by $\langle J\rangle = \sigma^*\langle E\rangle$, or $\sigma^* = \langle(\sigma_1\chi_1 + \sigma_2\chi_2)E_k\rangle$. Since σ^* is homogeneous $\sigma^*(\lambda\sigma_1, \lambda\sigma_2) = \lambda\sigma^*(\sigma_1, \sigma_2)$, we consider $m(h) = \sigma^*/\sigma_2$, $h = \sigma_1/\sigma_2$.

The following analytic properties of $m(h)$ have been established [29–31]: (i) $m(h)$ is analytic everywhere in the h plane except $(-\infty, 0]$, and (ii) $\mathrm{Im}(m) > 0$ when $\mathrm{Im}(h) > 0$. These properties of m were used to prove [31] the following representation formula for $F(s) = 1 - m(h), s = 1/(1 - h)$ (based on earlier conjectures in [29,30]),

$$F(s) = \int_0^1 \frac{d\mu(u)}{s - u}, \tag{2}$$

where μ is a positive measure on [0,1] depending only on the geometry of the medium. Representation (2) was also proven by applying the spectral theorem to the resolvent representation $F(s) = \langle\chi_1[(s + \Gamma\chi_1)^{-1}e_k] \cdot e_k\rangle$, where $\Gamma = \nabla(-\Delta)^{-1}\nabla\cdot$, and μ is the spectral measure of $\Gamma\chi_1$. This formula has been used quite successfully to obtain bounds on effective transport coefficients under microstructural constraints [29–33]. It was shown in [20] how (2) could be derived from a free energy $\Phi(s) = \int_0^1 \ln(s - u)d\mu(u)$, with $F = \partial\Phi/\partial s$. For the Ising model $f(T, H)$ has a similar representation [24]. For a finite resistor network with N resonances $s_n \in [0, 1]$, $\Phi(s)$ is the infinite volume limit of finite volume free energies $\Phi_N(s) = \frac{1}{N}\ln Z_N(s)$, where $Z_N(s) = \prod_{n=1}^N (s - s_n)$ is the partition function (whose zeros become distributed according to μ), serving as the analog of $Z_N(z) = a_N\prod_{n=1}^N (z - z_n), |z_n| = 1$, for an Ising model with N sites. We remark that (2) leads to $\partial^2 m/\partial h^2 \leq 0$, the analog of the G. H. S. inequality $\partial^2 M/\partial H^2 \leq 0$.

We now focus on applying (2) to conductivity functions $\sigma(x, \omega)$ describing lattice and continuum percolation models. We assume the existence of the critical exponents t and δ, defined above, as well as γ, defined via a conductive susceptibility $\chi(p) = \partial m/\partial h \sim (p - p_c)^{-\gamma}, p \to p_c^+, h = 0$ (which is different from [34] and numerous subsequent works). Furthermore, for $p > p_c$, we assume that there is a gap $\theta_h \sim (p - p_c)^{\Delta}$ in the support of μ around $h = 0$ or $s = 1$, which is discussed further below. Now, one of the key features of (1) is that the coefficients in the Taylor expansion of G around $\tau^2 = 0$ are the moments of ψ, which is not the case for (2), when expanded around $h = 0$ or $s = 1$. However, a simple change of variables $u = y/(y + 1)$ in (2) yields the direct analog of (1) for conductivity

$$m(h) = 1 + (h - 1)g(h), \qquad g(h) = \int_0^{\infty} \frac{d\phi(y)}{1 + hy}, \tag{3}$$

which is a general formula holding for two component stationary random media in lattice and continuum settings. In (3), g is a Stieltjes function of h, and ϕ is a positive measure which for our percolation models with $p > p_c$ is supported only in $[0, S(p)]$, where $S(p) \sim (p - p_c)^{-\Delta}, p \to p_c^+$. The moments $\phi_n = \int_0^\infty y^n d\phi(y)$ satisfy the inequalities $\phi_n \phi_m \leq \phi_0 \phi_{n+m}$, and form the coefficients of the expansion of g around $h = 0$, $g(h) = \phi_0 - \phi_1 h + \phi_2 h^2 - \ldots$, where $(-1)^n n! \phi_n = \partial^n g / \partial h^n(0)$ [$\sim -\partial^n m / \partial h^n(0)$ as $p \to p_c^+$]. We assume that $\phi_n(p) \sim (p - p_c)^{-\gamma_n}$, so that $\gamma_0 = 0$ and $\gamma_1 = \gamma$. The moment inequalities yield *Baker's inequalities* for transport,

$$\gamma_{n+1} - 2\gamma_n + \gamma_{n-1} \geq 0, \qquad n \geq 1. \tag{4}$$

We now exploit (3) to show that the critical exponents of transport above satisfy the same scaling relations as their counterparts in statistical mechanics. Instead of directly analyzing M as in [23], where $\gamma_0 = \gamma$, since $\gamma_0 = 0$ for transport, we must consider the derivative $\partial m / \partial h \sim h^{(1/\delta)-1}, h \to 0, p = p_c$. For p near p_c and h near 0, $-\partial m / \partial h \sim \partial g / \partial h = \int_0^{S(p)} [d\lambda(y)/(1 + hy)^2]$, where $d\lambda(y) = y d\phi(y)$ and $\lambda_0(p) \sim (p - p_c)^{-\gamma}$. Now, under the assumption that the asymptotic behavior of $\partial g / \partial h$ near criticality is determined primarily by the mass $\lambda_0(p)$ of λ and the rate of collapse of the spectral gap, with $S(p) = Q(p - p_c)^{-\Delta}$, we let $d\Lambda = (p - p_c)^{\gamma} d\lambda$ and $q = (p - p_c)^{\Delta} y$. Then

$$\frac{\partial g}{\partial h} = (p - p_c)^{-\gamma} \int_0^Q \frac{d\Lambda(q)}{[1 + h(p - p_c)^{-\Delta} q]^2} \sim (p - p_c)^{-\gamma} \mathcal{F}(x), \tag{5}$$

where $x = h(p - p_c)^{-\Delta}$. As $x \to \infty$ (or $p \to p_c$), we must have $\mathcal{F}(x) \sim x^{-\gamma/\Delta}$, so that $\partial m / \partial h \sim h^{-\gamma/\Delta} \sim h^{(1/\delta)-1}$, yielding

$$\delta = \frac{\Delta}{\Delta - \gamma}. \tag{6}$$

A generalization to higher derivatives of this argument shows that, like the Ising model, γ_n for transport is linear in n, $\gamma_n = \gamma + \Delta(n - 1), n \geq 1$, with constant gap $\gamma_i - \gamma_{i-1} = \Delta$, which is consistent with the absence of multifractal behavior for the bulk conductivity [1]. To involve t in our relations, we must analyze $\partial m / \partial p \sim (p - p_c)^{t-1}, p \to p_c^+, h = 0$. A more involved calculation than that above yields $\partial m / \partial p \sim h(p - p_c)^{-\gamma - 1} \mathcal{F}(x)$. As $x \to 0$ (or $h \to 0$), we must have $\mathcal{F}(x) \sim 1/x$, which yields

$$t = \Delta - \gamma. \tag{7}$$

Relations (6) and (7) hold for lattice and continuum percolation models, and establish in general that the two-parameter scaling exhibited by phase transitions in statistical mechanics holds for transport as well. As mentioned above, this type of scaling behavior has been proposed before for the lattice, e.g., [2,3,9], and the connection to these works is made through the identifications $\Delta = s + t, \gamma = s, \delta = (s + t)/t$, where s is the superconducting exponent. Our relations are satisfied directly by the exponents in effective medium theory for the lattice [35], $t = 1, \gamma = 1, \delta = 2$, and $\Delta = 2$. Unfortunately, it appears as if not enough is known numerically at this point about exponents other than t for the models of interest to directly test their validity. For the $d = 2$ lattice, where $\delta = 2$, these relations imply $t = \gamma = \Delta/2$, so that with $t = 1.3$ [1], $\gamma = 1.3$ and $\Delta = 2.6$. The inequalities $1 \leq t \leq 2$ for $d = 2, 3$ and $2 \leq t \leq 3$ for $d \geq 4$ [28,36] imply inequalities for the other exponents. For the $d = 2$ checkerboard, where it is believed that $\delta = 4$, these relations imply $t = \gamma/3 = \Delta/4$. The relation $\Delta = 2t$ for the $d = 2$ lattice was found in [35] with a different argument. It should be remarked that while there has been much numerical and analytical work on the sequence of critical exponents $\tilde{\psi}(q)$ for the moments of the current distribution in the resistor network, e.g., [1,37,38], this sequence exhibits nonlinear dependence in q, or multifractal behavior, as opposed to our γ_n, and these results are not in a form suitable for comparison with our findings. It is interesting, though, that the $\tilde{\psi}(q)$ satisfy the inequalities in (4) [37].

We now briefly discuss the gap θ_h for $p > p_c$, which has been investigated in [2] for the lattice. While the spectrum actually extends all the way to $h = 0$, the part close to $h = 0$ corresponds to very large, but very rare connected regions of the insulating phase (Lifshitz phenomenon), and is believed to give exponentially small contributions to σ^*, and not affect power law behavior. In fact, it is predicted in [2] that in numerical simulations there will appear to be a gap, which is supported in [35]. For a modified Swiss cheese model where the holes are separated by a minimal distance ϵ, Bruno [32] has proven the existence of a spectral gap and studied how it vanishes as $\epsilon \to 0$ (like $p \to p_c^+$). For the actual model we expect behavior similar to the lattice case.

Finally, let us consider the zeros of the conductivity partition function in the complex p plane, which correspond to the poles of $F(p, s)$. In [20], we used Padé approximants to the perturbation expansion of (2) around $h = 1$,

$$F(p, s) = \frac{\mu_0(p)}{s} + \frac{\mu_1(p)}{s^2} + \frac{\mu_2(p)}{s^3} + \cdots, \tag{8}$$

where the $\mu_j(p)$ are the moments of μ, to obtain a sequence of approximants to the partition function for the $d = 2$ random resistor network. Using exact results for μ_0, μ_1, μ_2, and μ_3 [39], we calculated the zeros of the partition function approximants to orders $k = 2$ and $k = 4$ in (8). Here we extend these calculations of the zeros to order $k = 6$ using the numerical results on μ_4 and μ_5 for the $d = 2$ lattice in [35,40], which are shown in Fig. 1. These results provide further evidence that the percolation threshold $p_c = \frac{1}{2}$ is an accumulation point of the zeros of these approximants as $k \to \infty$ (with $h = 0$), where the real p axis is "pinched" at p_c as $h \to 0$. Even further evidence is provided within the effective medium

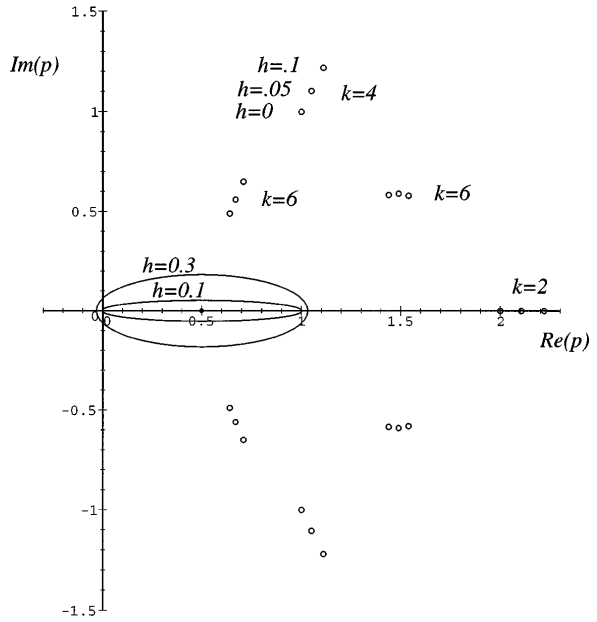

FIG. 1. Zeros of the partition function approximants for $k = 2, 4, 6$ in the complex p plane for the $d = 2$ random resistor network, with $h = 0.1$, 0.05, 0, and the zero-free region $\mathcal{D}_h$ for $h = 0.3$, 0.1.

approximation, where the zeros for $k = 8$ and $k = 10$ approach $p_c = \frac{1}{2}$ much more closely (not shown).

We close by noting that the following theorem proved in [20] rigorously establishes the existence of a gap θ_p in the zeros in the p plane around p_c, for "infinitely interchangeable media" (see, e.g., [39]), which include the lattice, and cell materials in the continuum such as the random checkerboard.

Theorem—For any $|h - 1| < 1$, $\sigma^*(p, h)$ *for an infinitely interchangeable medium in any dimension is analytic in a domain* $\mathcal{D}_h$ *in the complex p plane, where* $[0, 1] \subset \mathcal{D}_h$, *and* $\mathcal{D}_h$ *is the image of the annulus* $|1 - h| < |q| < 1$ *under the Joukowski conformal mapping* $p = \frac{1}{4}(2 + q + 1/q)$.

Assuming $\theta_p \sim (p - p_c)^{\Delta_p}$, the explicit construction of $\mathcal{D}_h$ establishes the general inequality $\Delta_p \leq 1$ for these media, which is satisfied in the effective medium approximation, where $\Delta_p = \frac{1}{2}$. In Fig. 1 we have plotted $\mathcal{D}_h$ for $h = 0.3$ and 0.1.

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