Statistical mechanics of conducting phase transitions

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The critical behavior of the effective conductivity σ^* of the random resistor network in Z^d, near its percolation threshold, is considered. The network has bonds assigned the conductivities 1 and $\epsilon \geq 0$ in the volume fractions p and $1-p$. Motivated by the statistical mechanics of an Ising ferromagnet at temperature T in a field H, we introduce a partition function and free-energy for the resistor network, which establishes a direct correspondence between the two problems. In particular, we show that the free energies for the resistor network and the Ising model both have the same type of integral representation, which has the interpretation of the complex potential due to a charge distribution on $[0, 1]$ in the $s = 1/(1-\epsilon)$ plane for the resistor network, and on the unit circle in the $z = \exp(-2\beta H)$ plane for the ferromagnet. Based on this correspondence, we develop a Yang–Lee picture of the onset of nonanalytic behavior of the effective conductivity σ^*, so that the percolation threshold $p = p_c$ is characterized as an accumulation point of zeros of the partition function in the complex p-plane as $\epsilon \to 0$. A scheme is developed to find the locations of a certain sequence of zeros in the p-plane, which is based on Padé approximation of a perturbation expansion of $\sigma^*(p, \epsilon)$ around a homogeneous medium ($\epsilon = 1$). Furthermore, for $\epsilon > 0$, we construct a domain $\mathscr{D}_\epsilon$ containing $[0, 1]$ in the p-plane in which $\sigma^*(p, \epsilon)$ is analytic, and which collapses as $\epsilon \to 0$. The explicit construction of this domain allows us to obtain a lower bound on the size of the gap in zeros of the partition function around the percolation threshold $p = p_c$, which leads to the gap exponent inequality $\Delta \leq 1$. © *1995 American Institute of Physics.*

I. INTRODUCTION

Systems which exhibit phase transitions have occupied a central place in statistical physics for a long while. One large class of such systems appearing in a wide variety of applications are conducting random media near the percolation threshold. The transport properties of these systems exhibit critical behavior near the threshold, and the medium undergoes an insulating/conducting phase transition there. Examples of such media include doped semiconductors, thermistors, composite conductors, brine-filled rocks, and sea ice. Perhaps the oldest model used to study transport in these types of percolating systems is the random resistor network in Z^d, whose bonds have conductivity 1 with probability p or $\epsilon \geq 0$ with probability $1-p$.[1-4] When $\epsilon = 0$, the effective conductivity $\sigma^*(p, \epsilon)$ exhibits an insulating/conducting phase transition at the percolation threshold $p_c (=\frac{1}{2}$ in $d = 2)$, with $\sigma^*(p, 0) = 0$ for $p \leq p_c$, and $\sigma^*(p, 0) \sim (p - p_c)^t$ as $p \to p_c^+$. Straley[5,6] first proposed that the critical behavior exhibited by the effective conductivity $\sigma^*(p, \epsilon)$ at p_c was analogous to that exhibited by a ferromagnet at its Curie point. In particular, the magnetization $M(T, H)$ of an Ising ferromagnet at temperature T in a magnetic field H vanishes when $H = 0$ for $T \geq T_c$, yet obeys $M(T, 0) \sim (T_c - T)^\beta$ as $T \to T_c^-$. Connections between the scaling theories of these two models was further investigated in Ref. 7.

While many general techniques of statistical physics have been brought to bear on the random resistor network and related models, the very basic question of the development of singular behavior in the infinite volume limit, or as $\epsilon \to 0$, is not well understood for conducting random

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media. For Ising ferromagnets, the Yang–Lee theory[8–10] provides a beautiful picture of the onset of nonanalytic behavior in terms of the zeros of the partition function. Regions in the complex H- or T-planes (containing a real segment) which remain free of zeros in the infinite volume limit cannot contain phase transition points. Typically, these transition points on the real H- or T-axes are characterized as accumulation points of zeros in an appropriate limit. In the H-plane, the Lee–Yang theorem[9] states that the zeros must lie on the imaginary axis Re $(H)=0$, so that a phase transition can occur only at $H=0$. In the T-plane, the situation is somewhat more complex, as was pointed out independently by Fisher[11] and Jones.[12] For the two-dimensional Ising model in zero magnetic field, Fisher[11] has shown that these zeros must lie on two circles in the complex $v=\tanh(2\beta J)$ plane, where J is the interaction. In Ref. 13 the locations of some of these so-called "Fisher's zeros" were calculated numerically. Zeros of the partition function in the T-plane have also been investigated for other models, including random energy and hierarchical models.[14–19]

For conducting random media, it has been established[20–22] that $\sigma^*(p,\epsilon)$ is analytic in ϵ off the negative real axis, which is similar to the above Lee–Yang theorem in the H-plane. Analogous questions in the p-plane have been addressed[23] and appear to be more complex than in the ϵ-plane. In these previous works, however, it was not clear how the similarities between the conductivity and Ising problems could shed light on the development of singular behavior of $\sigma^*(p,\epsilon)$ near percolation. In this work, we show that the statistical mechanical framework of the partition function and free energy can be set up directly for conductivity, so that a Yang–Lee picture for the onset of nonanalytic behavior can be developed for these types of problems. Our definitions of the partition function and free energy for conductivity, where there is no Hamiltonian, are based only on the Herglotz property of $\sigma^*(p,\epsilon)$, established in Refs. 20–22. Consequently, our framework for critical behavior holds also for other transport problems lacking a Hamiltonian, such as porous media, heat conduction, electrical permittivity, and advection/diffusion in a turbulent fluid,[24,25] where the effective parameters are Herglotz functions of appropriate variables.

Through the introduction of our partition function and free energy for conductivity, we are able to establish a direct connection between Ising and conductivity models. The Lee–Yang theorem for Ising ferromagnets enables one to represent the finite volume partition function $Z_N(z)$ with "activity" $z=\exp(-2\beta H)$ in terms of a product of factors $(z-z_n)$, where the zeros z_n lie on the unit circle in the z-plane. The thermodynamic limit f of the corresponding free energies f_N has an integral representation in the z-plane involving a positive measure ν on the unit circle. This representation gives f the interesting interpretation[9] as the complex potential in two dimensions due to charges distributed according to ν. The magnetization $M(T,H)$ is related to f via $M=-\partial f/\partial H$. In this framework, zeros of $Z_N(z)$ correspond to poles of the magnetization. For conductivity we now have a similar picture. For finite samples of the resistor network, the effective conductivity has a finite number of poles s_n lying on $[0,1]$ in the $s=1/(1-\epsilon)$ plane.[20–22] Analogous to the Ising model, we introduce a finite volume partition function $\mathscr{Z}_N(s)$ as a product of the factors $(s-s_n)$. The infinite volume limit Φ of the corresponding free energies Φ_N then has in the s-plane the same type of integral representation as f for the Ising model in the z-plane, except ν is replaced by a positive measure μ on the interval $[0,1]$. The conductivity $\sigma^*=1-F$ is related to Φ via $F=\partial\Phi/\partial s$.

Our framework then allows us to pursue a Yang–Lee analysis for conduction problems. In the s-plane the zeros are of course restricted to the unit interval $[0,1]$. However, in the p-plane the situation is more complicated, as with the Ising model in T. For conduction, the positions of the zeros s_n in $[0,1]$ depend on p, which correspond to singularities of the effective conductivity in the p-plane, located at the roots of the equations $s-s_n(p)=0$. For $\epsilon>0$, these roots, which are the zeros of $\mathscr{Z}_N(p)$, must lie off of $[0,1]$ in the complex p-plane. As one takes the infinite volume and $\epsilon\rightarrow 0$ limits, the percolation threshold $p=p_c$ is characterized as an accumulation point of zeros of $\mathscr{Z}_N(p,\epsilon)$ in the p-plane, which form analogs of Fisher's zeros for conductivity. Analogously, $\epsilon=0$ is characterized as an accumulation point of the zeros of $\mathscr{Z}_N(p,\epsilon)$ in the ϵ-plane.

While the direct calculation of the zeros of $\mathcal{Z}_N(p)$ in the p-plane as formulated above is a difficult problem, we will briefly look at numerically characterizing the onset of nonanalytic behavior in an alternate way. Since singular behavior on the real p-axis is attained only in the infinite volume and $\epsilon \to 0$ limits, there are various ways of constructing sequences of functions which converge to $\sigma^*(p, \epsilon = 0)$. Different sequences of approximants may have different locations of zeros in the p-plane, but we are only interested in those for which the zeros accumulate around $p = p_c$. Here we look at Padé approximants to a perturbation series expansion of $\sigma^*(p, \epsilon)$ around a homogeneous medium. This sequence of rational functions in $(1-\epsilon)$ has coefficients which are polynomials in p. The zeros of the denominators correspond to the zeros of the corresponding sequence of partition functions $\mathcal{Z}_N(p, \epsilon)$. Here, we only consider the perturbation expansion up to fourth order in $d=2$, which yields two zeros in the p-plane. Nevertheless, we are able to see these zeros move in the direction of $p = p_c = \frac{1}{2}$ as $\epsilon \to 0$. Further numerical work on this approach is required. In this same spirit, we examine the simple but illustrative $d=1$ case, which can be solved exactly, so that the behavior of the zero can be examined directly.

As transition points on the real axis are "pinched" by the zeros, a natural question is how fast do the zeros approach the real axis in a particular singular limit. For models in statistical mechanics, this question has been examined in detail, for example, in Ref. 26. The approach of the zeros is measured by the decay of a "gap" centered around a critical point on the real axis. This gap measures the distance from the critical point to the nearest zero. For conducting random media, we study the behavior of the gap in the p-plane as $\epsilon \to 0$ (where the infinite volume limit has already been taken). In particular, we find a domain $\mathcal{D}_\epsilon$ in the p-plane on which $\sigma^*(p, \epsilon)$ is analytic in p for $\epsilon > 0$ (or more generally, for $|\epsilon - 1| < 1$ with complex ϵ). As $\epsilon \to 0$, $\mathcal{D}_\epsilon \to [0, 1]$ so that analyticity in p is lost. Any nonanalytic behavior of $\sigma^*(p, \epsilon)$ in the p-plane for $\epsilon > 0$ [which corresponds to the limiting configuration of zeros of $\mathcal{Z}_N(p, \epsilon)$ as $N \to \infty$] must occur outside of this domain, so that $\mathcal{D}_\epsilon$ around $p = p_c$ provides a "lower bound" on the gap around $p = p_c$ extending in the Im (p) direction. More precisely, if we assume that the gap can be measured by a parameter θ, which decays to 0 as $\epsilon \to 0$ like $\theta \sim \epsilon^\Delta$, then a consequence of the explicit construction of $\mathcal{D}_\epsilon$ is the inequality $\Delta \leq 1$. Furthermore, the result establishes rigorously that no phase transition can occur when $\epsilon > 0$. The proof relies on establishing uniform convergence of the perturbation expansion of $\sigma^*(p, \epsilon)$ around a homogeneous medium $\epsilon = 1$. Weaker forms of this result were proven in Refs. 27 and 28.

We remark that Bruno[29] has proven a type of "gap" theorem in the ϵ-plane, which holds for separated inclusions of zero conductivity, away from the percolation threshold. In the ϵ-plane, the singularities of σ^* on the negative real axis correspond to the spectrum of a self-adjoint operator determined by the microstructural geometry. For certain types of inclusions, the collapse of the gap (to the left of zero on the negative real axis) as they begin to touch can be studied. Bruno used the spectral gap to derive bounds on σ^* in the real case, and Sawicz and Golden[30] have used it to obtain bounds on the complex permittivity of sea ice.

It is also interesting to remark that if one views recent results on the random checkerboard in two dimensions[31,32] in terms of the ferromagnetic analogy, then we see for continuum problems that the phase transition point can be "smeared out" over an interval on the real p-axis. In particular, the quantity which corresponds to the magnetic susceptibility $\chi = \partial M / \partial H$ diverges for p throughout an interval, not just at a particular $p(=p_c)$ as in the case of the lattice.

Finally, we note that for any finite graph, the generating function for spanning trees[33] has been observed[34] to play a role similar to that of the partition function in statistical mechanics. There the effective resistance of the network can be written as a logarithmic derivative of the generating function, where the variable of differentiation is the conductivity of a specially designated "battery bond," which plays the role of the applied field for the Ising model. In our formulation the variable is the conductivity of the nearly insulating **phase** in the random medium. Our $\mathcal{Z}_N(p, \epsilon)$ appear to be different from the tree generating functions.

II. ANALYTIC PROPERTIES OF EFFECTIVE CONDUCTIVITY AND MAGNETIZATION

We first formulate the effective conductivity problem for a general stationary random medium in the continuum, and then point out the modifications for the random resistor network.[22,28] Subsequently, we briefly review Ising ferromagnets.

Let (Ω, P) be a probability space and $\sigma(x, \omega)$ be a stationary stochastic process in $x \in R^d$ and $\omega \in \Omega$. The space Ω represents the set of all realizations of our random medium, and P is a probability measure on Ω which is compatible with the stationarity, i.e., it is invariant under the translation group $\tau_y : \Omega \rightarrow \Omega$ defined by $\tau_y \omega(x) = \omega(x + y)$, $\forall x, y \in R^d$, $\omega \in \Omega$. We consider two-component media, so that $\sigma(x, \omega)$ takes two values σ_1 and σ_2, and can be written as

$$\sigma(x, \omega) = \sigma_1 \chi_1(x, \omega) + \sigma_2 \chi_2(x, \omega), \tag{1}$$

where the characteristic function $\chi_j(x, \omega)$ equals 1 for all realizations ω which have medium j at x, $j = 1, 2$, and equals 0 otherwise. Let $E(x, \omega)$ and $J(x, \omega)$ be stationary random electric and current fields satisfying

$$J(x, \omega) = \sigma(x, \omega) E(x, \omega), \tag{2}$$

$$\nabla \cdot J(x, \omega) = 0, \tag{3}$$

$$\nabla \times E(x, \omega) = 0, \tag{4}$$

$$\langle E(x, \omega) \rangle = \int_\Omega P(d\omega) E(x, \omega) = e_k, \tag{5}$$

where e_k is a unit vector in the kth direction. In (2) and (4) the differential operators $\partial / \partial x_i$ are replaced by the infinitesimal generators D_i of the unitary group T_x acting on $L^2(\Omega, P)$ via $(T_x f)(\omega) = f(\tau_x \omega) = f(x, \omega)$, for any $f \in L^2(\Omega, P)$, which is a stationary process on R^d and Ω. By stationarity, we may focus attention at $x = 0$, and subsequently drop the x-notation.

In view of (2), the effective conductivity tensor σ^* may now be defined as

$$\langle J(\omega) \rangle = \sigma^* \langle E(\omega) \rangle, \tag{6}$$

so that the coefficients σ_{jk}^* can be written as

$$\sigma_{jk}^* = \int_\Omega P(d\omega) \sigma(\omega) E_j^k(\omega), \tag{7}$$

where $E_j^k(\omega)$ is the jth component of E^k satisfying (2)–(5). We shall restrict our attention to isotropic media, where $\sigma_{jk}^* = \sigma^* \delta_{jk}$, and pick out a diagonal coefficient

$$\sigma^* = \sigma_{kk}^* = \int_\Omega P(d\omega) [\sigma_1 \chi_1(\omega) + \sigma_2 \chi_2(\omega)] E_k^k(\omega). \tag{8}$$

Since σ^* is homogeneous of degree 1, i.e.,

$$\sigma^*(\lambda \sigma_1, \lambda \sigma_2) = \lambda \sigma^*(\sigma_1, \sigma_2), \tag{9}$$

it suffices to let

$$\sigma_1 = \epsilon, \quad \sigma_2 = 1, \tag{10}$$

and consider the function

$$m(\epsilon) = \sigma^* = \int_\Omega P(d\omega)[\epsilon\chi_1(\omega) + \chi_2(\omega)]E_k^k(\omega). \tag{11}$$

It has been established[20-22] that

$m(\epsilon)$ is analytic everywhere in the ϵ-plane except $(-\infty, 0]$, the negative real axis. (12)

Furthermore, from the symmetric, or variational formulation of σ^*,

$$m(\epsilon) = \int_\Omega P(d\omega)[\epsilon\chi_1 + \chi_2]E^k \cdot \overline{E^k}, \tag{13}$$

where the overbar denotes complex conjugation, we see that m maps the upper half-plane to the upper half-plane, i.e.,

$$\text{Im}(\epsilon) > 0 \Rightarrow \text{Im}(m(\epsilon)) > 0. \tag{14}$$

The two key properties (12) and (14) of $m(\epsilon)$ allow it to be classified as a Herglotz function. A most useful consequence of the Herglotz property is that such functions have a convenient integral representation. To exhibit the formula, it is simpler to consider the function

$$F(s) = 1 - m(h), \quad s = \frac{1}{1-\epsilon}, \tag{15}$$

which is analytic off $[0, 1]$ in the s-plane. It has been proven[22] that $F(s)$ has the integral representation

$$F(s) = \int_0^1 \frac{d\mu(t)}{s-t}, \quad s \notin [0, 1], \tag{16}$$

where μ is a positive Borel measure on $[0, 1]$. This representation can be proved either as a consequence of the Herglotz theorem in analytic function theory[35] or by applying the spectral theorem to the resolvent representation

$$F(s) = \int_\Omega P(d\omega)\chi_1[(s + \Gamma\chi_1)^{-1}e_k] \cdot e_k. \tag{17}$$

In (17), $\Gamma = \nabla(-\Delta)^{-1}\nabla\cdot$, with the differential operators again replaced by the generators of translations on Ω. In the Hilbert space $L^2(\Omega, P)$ with χ_1 in the inner product, $\Gamma\chi_1$ is a bounded self-adjoint operator with norm less than or equal to 1. Then (16) is the spectral representation of the resolvent $(s + \Gamma\chi_1)^{-1}$, where μ is the spectral measure of the family of projections associated with $\Gamma\chi_1$. It is important to note that in (16), the parameter information $s = 1/(1-\epsilon)$ is separated from the geometry information (χ_1), which is all contained in the measure μ.

In the investigation of analyticity of σ^*, it will be necessary to consider a perturbation expansion of σ^* around a homogeneous medium $\epsilon = 1$ or $s = \infty$. By expanding the integrand of (16) for $|s| > 1$ in powers of $1/s$, we obtain

$$F(s) = \frac{\mu_0}{s} + \frac{\mu_1}{s^2} + \frac{\mu_2}{s^3} + \cdots, \tag{18}$$

where the μ_j are the moments of the measure μ,

$$\mu_j = \int_0^1 z^j \, d\mu(z) \geqslant 0. \tag{19}$$

By equating (18) to the same expansion of (17) around $s = \infty$, one obtains

$$\mu_j = (-1)^j \int_\Omega P(d\omega) [\chi_1 (\Gamma \chi_1)^j e_k] \cdot e_k. \tag{20}$$

Clearly, for any medium, we have

$$\mu_0 = p_1, \tag{21}$$

the relative volume fraction of $\sigma_1 = \epsilon$. When the medium is isotropic, μ_1 can be calculated as well,[22,28] with

$$\mu_1 = \frac{p_1 p_2}{d}. \tag{22}$$

In general, μ_n depends on the $(n+1)$-point correlation function of the medium under consideration.

In the case of the random resistor network in Z^d, where the bonds are independently assigned the conductivities 1 with probability p and $\epsilon \geqslant 0$ with probability $1 - p$, then the above formulation holds with minor but important differences. The key feature of the resistor network which distinguishes it from general random media in the continuum is that once p is fixed, the measure μ in (16) (as well as all of its moments) is completely determined. There is no further geometry to specify, as in the general continuum case. This exclusive dependence on p is true also for cell materials in the continuum, such as the random checkerboard in $d = 2$, where all space is divided up into unit squares, which are then assigned conductivities in the same way as the bonds for the random resistor network. In general, once the geometry of the cell is fixed, σ^* depends only on p.

The setup for the resistor network problem is the same except the unitary translation group is generated by composition of the operators $T_i^+ = T_{+e_i}$ and $T_i^- = T_{-e_i}$, where e_i is a unit vector in the ith direction. Then the differential operators in (3) and (4) are replaced by forward and backward difference operators,

$$D_i^+ = T_i^+ - I, \tag{23}$$

$$D_i^- = T_i^- - I, \tag{24}$$

where I is the identity operator and $i = 1, \ldots, d$. Equations (3) and (4) become Kirchhoff's laws:

$$\sum_{i=1}^d D_i^- J_i(\omega) = 0, \tag{25}$$

$$D_i^+ E_j(\omega) - D_j^+ E_i(\omega) = 0. \tag{26}$$

In the resolvent representation (17) and the moment formulas (20), the operator Γ is replaced by

$$\Gamma = \nabla^+ (-\Delta)^{-1} \nabla^- \cdot, \tag{27}$$

where the difference operators in (23) and (24) replace the differential operators in ∇. The inverse Laplacian $(-\Delta)^{-1}$ can be expressed as convolution with the lattice Green's function. For the $d = 2$ square lattice, the first four moments of μ have been calculated explicitly,[28]

$$\mu_0 = 1-p, \quad \mu_1 = \frac{p(1-p)}{2}, \quad \mu_2 = \frac{p(1-p)}{4}, \quad \mu_3 = \frac{p(1-p)[1+p(1-p)]}{8}, \quad (28)$$

where $p = p_2$ and the probability of $\sigma_2 = 1$.

In order to motivate our analysis of the development of singular behavior of $\sigma^*(p,\epsilon)$, we formulate the problem of finding the magnetization $M(T, H)$ of an Ising ferromagnet at temperature T in a field H. We consider a finite box $\Lambda \subset Z^d$ containing N sites. At each site there is a spin variable s_i which can take the value $+1$ or -1. We consider a Hamiltonian with ferromagnetic pair interaction $J \geq 0$ between nearest-neighbor pairs,

$$\mathcal{H}_\omega = -J \sum_{\langle i,j \rangle} s_i s_j - H \sum_i s_i, \quad (29)$$

for any configuration $\omega \in \Omega = \{-1,1\}^N$ of the spin variables. The canonical partition function Z_N is given by

$$Z_N(T, H) = \sum_{\omega \in \Omega} \exp(-\beta \mathcal{H}_\omega) = \exp(-\beta N f_N), \quad (30)$$

where $\beta = 1/kT$, k is Boltzmann's constant, and f_N is the free energy per site,

$$f_N(T, H) = \frac{-1}{\beta N} \log Z_N(T,H). \quad (31)$$

We are interested in the infinite volume limit $f(T, H)$ of (31),

$$f(T, H) = \lim_{N \to \infty} f_N(T,H). \quad (32)$$

Then the average magnetization in the infinite volume limit

$$M(T, H) = \lim_{N \to \infty} M_N(T, H) = \lim_{N \to \infty} \frac{1}{N} \left\langle \sum_{i=1}^N s_i \right\rangle_\omega, \quad (33)$$

where $\langle \cdot \rangle_\omega$ denotes average over $\omega \in \Omega$ with Gibbs' weights, can be expressed in terms of the free energy as

$$M(T, H) = -\frac{\partial f}{\partial H}. \quad (34)$$

The magnetic susceptibility $\chi(T, H)$ is given by

$$\chi(T, H) = \frac{\partial M}{\partial H} = -\frac{\partial^2 f}{\partial H^2} \geq 0. \quad (35)$$

Now, to present the Lee–Yang theorem for ferromagnets,[10] we consider the slightly modified Hamiltonian

$$\mathcal{H}'_\omega = -J \sum_{\langle i,j \rangle} s_i s_j - H \sum_i (s_i - 1). \quad (36)$$

It is easy to see that the partition function Z_N in (30) for this Hamiltonian can be written as an Nth-order polynomial

$$Z_N(z) = \sum_{n=0}^{N} a_n z^n, \quad a_n \geq 0, \tag{37}$$

in the "activity"

$$z = \exp(-2\beta H). \tag{38}$$

This polynomial has the following remarkable property, which is the analog of (12) for Ising ferromagnets.

Theorem 1 (Lee–Yang): If $J \geq 0$, then $Z_N = 0$ implies z lies on the unit circle $|z| = 1$ or, equivalently, H lies on the imaginary axis Re $(H) = 0$ (with real β). Then (37) can be written as

$$Z_N(z) = a_N \prod_{n=1}^{N} (z - z_n), \quad |z_n| = 1, \tag{39}$$

where $a_N = \exp(c_N \beta J)$ and c_N is the number of nearest neighbor pairs in Λ.

The product representation (39) can be used to derive a useful integral representation for the free energy $f(T, H)$. Inserting (39) into the definition of the finite volume free energy $f_N(T, H)$ in (31), we obtain

$$f_N(T, H) = \frac{-1}{\beta} \sum_{n=1}^{N} \frac{1}{N} \log(z - z_n) - \frac{1}{\beta N} \log a_N. \tag{40}$$

In the thermodynamic limit, the zeros z_n get "smeared out" according to some positive measure ν. Using the fact that $c_N \sim 2dN$ as $N \to \infty$ for nearest neighbors, the free energy in the thermodynamic limit can be expressed as

$$f(T, H) = \frac{-1}{\beta} \int_{|t|=1} \log(z - t) d\nu(t) - 2d\beta J. \tag{41}$$

The integral in (41) has the interpretation[9] of the complex electrostatic potential in two dimensions due to charges distributed according to the measure ν on the circle. [The constant in (41) is different than in Refs. 9 and 26 since we have a slightly modified Hamiltonian.] In fact, we have via (34) the following

$$M(T, H) = 2z \int_{|t|=1} \frac{d\nu(t)}{z - t}, \tag{42}$$

which shows how zeros of the partition function are connected to poles of the magnetization.

It is interesting to note that, based on the above considerations, a Herglotz representation for the magnetization has been found and exploited by Baker[26] [for the Hamiltonian in (29)],

$$G(\tau) = \int_0^A \frac{d\psi(t)}{1 + \tau^2 t}, \tag{43}$$

where $A \geq 0$, ψ is a positive measure on $[0, A]$,

$$G(\tau) = \frac{M - \tau}{\tau(1 - \tau^2)}, \tag{44}$$

and

$$\tau = \tanh(\beta H). \tag{45}$$

The integral representation in (43) immediately leads to the inequalities

$$G \geqslant 0, \tag{46}$$

$$\frac{\partial G}{\partial u} \leqslant 0, \tag{47}$$

$$\frac{\partial^2 G}{\partial u^2} \geqslant 0, \tag{48}$$

where $u = \tau^2$. It is interesting to compare (46)–(48) with analogous inequalities derived from (16) for $s > 1$ or $0 < \epsilon \leqslant 1$,

$$F \geqslant 0, \tag{49}$$

$$\frac{\partial F}{\partial s} \leqslant 0, \tag{50}$$

$$\frac{\partial^2 F}{\partial s^2} \geqslant 0. \tag{51}$$

In the magnetic case, (48) is just the GHS inequality.[36] In the conducting case, (51) is a macroscopic version of the fact that the effective resistance of a finite network is a concave downward function of the resistances of the individual network elements.[37] It is interesting to note that this theorem was discovered after many unsuccessful attempts at obtaining a rheostat having resistance that was a concave upward function of the shaft angle, which was needed in the development of the computer.

As a final note in this section, we observe that some recent results on continuum percolation models, namely the random checkerboard in $d = 2$, suggest that in the continuum, a "phase transition" can be smeared out, rather than occur at a single point. In the random checkerboard model, the plane is divided into unit squares, each of which is assigned the conductivities 1 with probability p and ϵ with probability $1 - p$. Recent work[32] has shown that there is a surprising exact result for the leading-order term of the effective conductivity $\sigma^*(p, \epsilon)$ as $\epsilon \to 0$, when $p \in (1 - p_c^{\ s}, p_c^{\ s})$, where $p_c^{\ s} \approx 0.59$ is the site percolation probability. Namely, we have found that $\sigma^*(p, \epsilon) = \sqrt{\epsilon} + O(\epsilon)$ as $\epsilon \to 0$ for all p in this interval. Now, for ferromagnets, the susceptibility is $\chi = \partial M / \partial H$, and the corresponding quantity for conduction is $\chi_c = \partial \sigma^* / \partial \epsilon$. For the Ising ferromagnet on the lattice, and the random resistor network in $d = 2$, the susceptibilities diverge only at the critical point. For the bond lattice resistor network in $d = 2$, this occurs only at $p = p_c = \frac{1}{2}$, where $\sigma^*(p = \frac{1}{2}, \epsilon) = \sqrt{\epsilon}$, so that $\chi_c \to \infty$ as $\epsilon \to 0$. However, for the random checkerboard in $d = 2$, in view of the above result, we see that $\chi_c \to \infty$ as $\epsilon \to 0$ for all $p \in (1 - p_c^{\ s}, p_c^{\ s})$.

III. DEVELOPMENT OF SINGULAR BEHAVIOR FOR THE EFFECTIVE CONDUCTIVITY

To help understand the onset of nonanalytic behavior of σ^* near the percolation threshold $p = p_c$, $\epsilon = 0$, we now introduce a partition function and free energy for conductivity problems. For any finite resistor network, the effective conductivity has a finite number of poles $s_n \in [0, 1]$ in the $s = 1/(1 - \epsilon)$ plane.[20–22] In view of (39), we define a partition function for conductivity as

$$\mathscr{Z}_N(s) = \prod_{n=1}^{N} (s - s_n), \quad s_n \in [0, 1], \tag{52}$$

so that these poles become the zeros of $\mathscr{Z}_N(s)$. In view of (31), we define the corresponding finite volume free energy as

$$\Phi_N(s) = \frac{1}{N} \log \mathscr{Z}_N(s). \tag{53}$$

We then define $\Phi(s)$ to be the thermodynamic limit of the $\Phi_N(s)$,

$$\Phi(s) = \lim_{N \to \infty} \Phi_N(s), \tag{54}$$

or

$$\Phi(s) = \lim_{N \to \infty} \sum_{n=1}^{N} \frac{1}{N} \log(s - s_n). \tag{55}$$

For the random resistor network the locations of the poles (or zeros) s_n depend on p. As the thermodynamic limit is taken, the poles may become smeared out, according to some positive measure μ, which of course depends on p, so that we now may write explicitly

$$\Phi(p,s) = \int_0^1 \log(s-t) d\mu(t). \tag{56}$$

This representation for the free energy is the analog of (41) for conductivity and has the interpretation of the complex electrical potential in two dimensions due to charges on $[0, 1]$ distributed according to the measure μ. As for the Ising model in (34), we can recover the effective conductivity $\sigma^* = 1 - F$ via the relation

$$F(p,s) = \frac{\partial \Phi}{\partial s}, \tag{57}$$

so that

$$F(p,s) = \int_0^1 \frac{d\mu(t)}{s-t}. \tag{58}$$

We note that our definitions of the partition function and free energy can be defined for any Herglotz function with a representation like (58)

It is important to remark that any conductivity function of the form (58) or any free energy of the form (56) can be obtained from a limit of sums of Dirac point measures of the form

$$\mu_N = \sum_{n=1}^{N} \alpha_n \delta(t - s_n), \tag{59}$$

where the "residues" α_n and support points s_n satisfy

$$\sum_{n=1}^{N} \alpha_n = 1 - p, \quad \alpha_n \geq 0, \quad s_n \in [0, 1). \tag{60}$$

For a finite sample of the random resistor network, the α_n and s_n are functions of p, and the sample size. [The condition that $s_n \neq 1$ is a reflection of the physical condition that $F(s=1) \leqslant 1$.] Mathematically, the measures in (59) are important because any positive Borel measure on $[0, 1]$ can be expressed as a weak*-limit of these measures. In particular, the measure μ characterizing the infinite volume limit of the random resistor network is a limit of such finite volume approximants.

Now, in order to create a scheme which can illustrate the onset of nonanalytic behavior in the p-plane, we consider a sequence of approximants to $F(p,s)$ based on the simple Dirac measures in (59). The sequence that we consider, however, is not based on increasing volume, but is a sequence of Padé approximants to the perturbation series in (18). Then the accumulation of zeros toward the percolation threshold should, in principle, take place as one incorporates higher and higher order correlation information about the medium, and as $\epsilon \to 0$. We note that in this approach, we are already working in the infinite volume limit. This sequence of functions has been discussed in some detail in the context of bounds on effective parameters in a number of works, including Refs. 38 and 39.

We construct a sequence $F_N(p,s)$ of approximants to $F(p,s)$ as follows. Let

$$F_N(p,s) = \sum_{n=1}^{N} \frac{\alpha_n(p)}{s - s_n(p)}. \tag{61}$$

To determine the masses $\alpha_n(p)$ and pole locations $s_n(p)$, we equate $F_N(p,s)$ to the perturbation expansion

$$F(p,s) = \frac{\mu_0(p)}{s} + \frac{\mu_1(p)}{s^2} + \frac{\mu_2(p)}{s^3} + \cdots , \tag{62}$$

assumed known to order $2N$. Since the $\mu_n(p)$ for the random resistor network are polynomials in p, the $s_n(p)$ are polynomials in p as well, and the roots of the equations

$$s_n(p) = s, \quad n = 1,\dots,N, \tag{63}$$

form the poles of F_N in the p-plane, or the corresponding zeros of $\mathscr{Z}_N$. Since $s_n(p) \in [0, 1)$ when $p \in [0, 1]$, $s \geqslant 1$ implies that the poles must stay away from the interval $[0, 1]$ in the p-plane for finite N.

To illustrate the procedure, we calculate the zeros for $N=1$ and $N=2$ for the $d=2$ random resistor network. For $N=1$, we assume that the perturbation expansion (62) is known only to second order,

$$F(p,s) = \frac{1-p}{s} + \frac{p(1-p)/2}{s^2} + \cdots . \tag{64}$$

For F_1 we obtain

$$F_1(p,s) = \frac{1-p}{s - p/2}, \tag{65}$$

which is just the $[0, 1]$ Padé approximant to the series in (64). (An $[n, m]$ Padé approximant to a power series expansion is a rational function whose numerator is a polynomial of order n and whose denominator is a polynomial of order m.) In more standard Padé notation, in the variable $u = 1/s$, (65) is the $[1, 1]$ Padé approximant to this series. Clearly the lone zero of the denominator is at $p = 2s$, so that even when $\epsilon = 0$ or $s=1$, the closest this zero comes to the interval $[0, 1]$ is $p=2$.

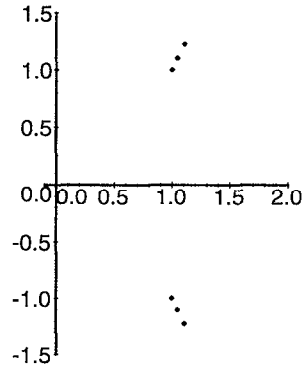FIG. 1. Three pairs of zeros of $\mathscr{Z}_2(p,\epsilon)$ in the complex p-plane for $\epsilon = 0.1$, 0.05, and 0. As $\epsilon \to 0$, the zeros move in the direction of $p_c = \frac{1}{2}$.

For $N=2$, we assume that the perturbation expansion is known to fourth order,

$$F(p,s) = \frac{1-p}{s} + \frac{p(1-p)/2}{s^2} + \frac{p(1-p)/4}{s^3} + \frac{p(1-p)[1+p(1-p)]/8}{s^4} + \cdots . \tag{66}$$

Now, to obtain $F_2(p,s)$, which is the $[1, 2]$ Padé approximant to the series in (66), we use Maple, resulting in

$$F_2(p,s) = \frac{2(1-p)s - (1-p)}{2s^2 - (1-p)s + p^2/2}, \tag{67}$$

so that

$$\mathscr{Z}_2(p,s) = s^2 - \frac{(1-p)}{2} s + \frac{p^2}{4}. \tag{68}$$

Again, in more standard Padé notation, in the variable $u = 1/s$, (67) is the $[2, 2]$ Padé approximant to the series (66). Now, to obtain the zeros of $\mathscr{Z}_2(p,s)$ in the p-plane, we view the denominator as a quadratic polynomial in p, and use Maple again to find its roots as functions of s (or ϵ). In Fig. 1, we have plotted these zeros for $\epsilon = 0.1$, 0.05, and 0.0. Note that as ϵ approaches 0, the zeros approach the interval $[0, 1]$ in the direction of $p = \frac{1}{2}$, and the closest they come is $1 \pm i$ when $\epsilon = 0$.

As a final example in this section, we consider the $d=1$ random resistor network, which can solved exactly rather simply, and exhibits "trivial" critical behavior. Nevertheless, it is illustrative to analyze its behavior in light of the above picture. It is easy to see that the exact solution for the $d=1$ case is

$$(\sigma^*(p,\epsilon))^{-1} = \left(\frac{1-p}{\epsilon} + \frac{p}{1} \right), \tag{69}$$

or in terms of F, it is

$$F(p,s) = \frac{1-p}{s-p}. \tag{70}$$

As $\epsilon \to 0$, the limiting function of p is $\sigma^*(p)=0$ if $p \in [0, 1)$ and $\sigma^*(p)=1$ if $p=1$. The "percolation threshold" occurs at $p=1$. In view of (70), the lone zero of the partition function [or pole of $F(p,s)$] is located at $p=s$. As $s \to 1$, or $\epsilon \to 0$, the location approaches the critical point $p=1$. However, the strength of this pole, or its residue, which from (70) is $1-p$, vanishes as $p \to 1$. This mechanism, we believe, should be characteristic of higher dimensions as well, whereby, for large N, poles in the p-plane approach the threshold $p=p_c$, yet their residues should vanish as $N \to \infty$ and $\epsilon \to 0$.

IV. LOWER BOUND ON THE DOMAIN OF ANALYTICITY OF THE EFFECTIVE CONDUCTIVITY IN THE COMPLEX VOLUME FRACTION PLANE

The magnetization $M(T, H)$ of an Ising ferromagnet displays a phase transition at a critical temperature T_c only in the limit as the applied magnetic field $H \to 0$, and only in the infinite volume limit. If one first takes the infinite volume limit, with $H>0$, then the zeros of the partition function take on some limiting configuration, all points of which must avoid the real T-axis. As one then takes the subsequent limit of $H \to 0$, then this "zero structure" converges from above and below the real T-axis to $T=T_c$, i.e., $T=T_c$ is "pinched," and $M(T, H)$ loses analyticity in T at $T=T_c$. In this and many similar problems of statistical physics, one is interested in how fast the critical point ($T=T_c$) is pinched as some parameter goes to its critical value, such as $H \to 0$. The typical situation is that there is a gap in the zero structure around the critical point which collapses in the appropriate limit. In this section we study this question for the (infinite volume) zero structure $\mathscr{S}_\epsilon$ of the conductivity partition function as $\epsilon \to 0$. This zero structure $\mathscr{S}_\epsilon$ corresponds to the singular set for $\sigma^*(p,\epsilon)$ in the p-plane, i.e., any point in the p-plane where $\sigma^*(p,\epsilon)$ is *not* analytic, hence the notation. Our first goal is to establish a domain $\mathscr{D}_\epsilon$ in the p-plane in which $\sigma^*(p,\epsilon)$ is analytic or, equivalently, which has empty intersection with $\mathscr{S}_\epsilon$. This will rigorously establish the absence of a phase transition for $\epsilon > 0$. Secondly, by the nature of our construction of the domain $\mathscr{D}_\epsilon$, we shall be able to obtain a "lower bound" on the gap in $\mathscr{S}_\epsilon$ around the percolation threshold $p=p_c$.

We are now ready to construct a domain of analyticity for $\sigma^*(p,\epsilon)$.

Theorem 2: Let $\sigma^*(p,\epsilon)$ be the effective conductivity of the random resistor network in $d \geq 1$ dimensions, with conductivities 1 and ϵ in the volume fractions p and $1-p$. For every ϵ such that $|\epsilon-1|<1$, there exists a domain $\mathscr{D}_\epsilon$ in the complex p-plane such that $\sigma^*(p,\epsilon)$ is analytic in $\mathscr{D}_\epsilon$. In particular, $[0, 1] \subset \mathscr{D}_\epsilon$, and $\mathscr{D}_\epsilon$ is the image in the p-plane of the annulus

$$|1-\epsilon|<|q|<1 \tag{71}$$

in the complex q-plane under the conformal mapping

$$p=\frac{1}{4}\left(2+q+\frac{1}{q}\right). \tag{72}$$

Proof: Fix ϵ such that $|\epsilon-1|<1$, or $|s|>1$, and consider the perturbation expansion in (16), which we write as

$$F(p,s)=\frac{a_1(p)}{s}+\frac{a_2(p)}{s^2}+\frac{a_3(p)}{s^3}+\cdots. \tag{73}$$

The idea of the proof is to construct a domain in the p-plane in which (73) converges uniformly. Now, for $p \in [0, 1]$, the coefficients $a_n(p)$ are the moments of a positive measure of mass $a_1(p)=1-p$, with $a_n(p) \geq a_{n+1}(p)$, so that

$$|a_n(p)| \leq 1, \quad p \in [0, 1]. \tag{74}$$

Clearly then for $p \in [0, 1]$, (73) converges uniformly, and we must stretch this convergence for p away from $[0, 1]$ as far as we can. So consider the slit plane $W = \{p \in C : p \notin [0, 1]\}$. Conformally map W onto the unit disk $D = \{|q| < 1\}$ in the complex q-plane via the Joukowski transformation in (72). This mapping takes $p = \infty$ to $q = 0$, and the unit interval $[0, 1]$ gets mapped to the unit circle $|q| = 1$. Now, the key fact about the random resistor network which the proof requires is that the moments $a_n(p)$ are polynomials in p. In general, $a_n(p)$ is a polynomial of order less than or equal to n. Then $a_n(q)$ has at worst an nth-order pole at $q = 0$. Thus $q^n a_n(q)$ is analytic in D. Since $|a_n(q)| \leq 1$ for $|q| = 1$, by the maximum modulus principle,

$$|a_n(q)| \leq \frac{1}{|q|^n}, \quad q \in D, \tag{75}$$

or

$$\left| \frac{a_n(q)}{s^n} \right| \leq \left(\frac{1}{|q||s|} \right)^n, \quad q \in D. \tag{76}$$

Then, for fixed $|s| > 1$, if we choose q so that

$$\frac{1}{|q||s|} < 1, \tag{77}$$

by (75) we are assured of geometric (and therefore uniform) convergence of (73). Thus if q is chosen to lie in the annulus

$$\frac{1}{|s|} < |q| < 1, \tag{78}$$

then $\sigma^*(p, \epsilon)$ is analytic in the image $\mathscr{D}_\epsilon$ of the annulus 78 under the mapping (72), which proves the theorem.

Remarks: The above Theorem and its proof hold for a large class of continuum materials as well, namely infinitely interchangeable media, which were introduced by Bruno.[40] This class is a generalization of Miller's cell materials,[41] where all space is divided up into cells, such as squares in the plane, which are then assigned conductivities, such as 1 and ϵ, with probabilities p and $1 - p$. In Ref. 28 we proved that the moments of the measure μ for infinitely interchangeable media are polynomials in p, which as mentioned in the proof, is the key fact required to make the proof work. We also note that this Theorem provides a rigorous basis for many volume fraction expansions of $\sigma^*(p)$, which have been widely used since the time of Maxwell.

In Fig. 2, we have plotted the domain $\mathscr{D}_\epsilon$ for $\epsilon = 0.3$ and $\epsilon = 0.1$. Note its collapse to the interval $[0, 1]$ as $\epsilon \to 0$. Note also that the locations of the poles in Fig. 1 are outside of the corresponding $\mathscr{D}_\epsilon$. We remark that the full domain of analyticity of $\sigma^*(p, \epsilon)$ in the p-plane is certainly larger than $\mathscr{D}_\epsilon$, and in this sense, we say that $\mathscr{D}_\epsilon$ forms a lower bound on the full domain of analyticity.

Finally, we are now ready to obtain a lower bound on the size of the gap in the singularity set $\mathscr{S}_\epsilon$ for $\sigma^*(p, \epsilon)$ in the p-plane around the percolation threshold $p = p_c$. Equivalently, we obtain a lower bound on the distance from the percolation threshold to the nearest points of the zero configuration of the conductivity partition function. Now, let θ measure the size of the gap in $\mathscr{S}_\epsilon$ around the percolation threshold $p = p_c$. More precisely, let θ be twice the distance from $p = p_c$ to the nearest point of $\mathscr{S}_\epsilon$. Furthermore, let us assume that the decay of the gap obeys a power law as $\epsilon \to 0$,

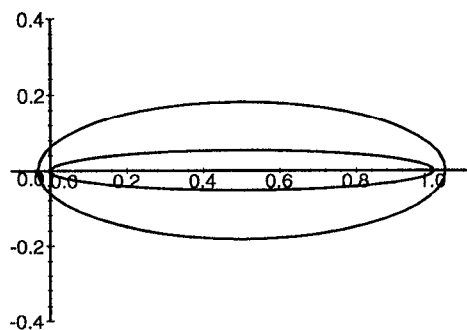$$\theta \sim \epsilon^\Delta, \quad \epsilon \to 0, \tag{79}$$

FIG. 2. The domain $\mathscr{D}_\epsilon$ in the complex p-plane for $\epsilon = 0.3$ (larger domain) and $\epsilon = 0.1$. The effective conductivity $\sigma^*(p,\epsilon)$ is analytic in p inside $\mathscr{D}_\epsilon$, which shrinks to $[0, 1]$ as $\epsilon \to 0$.

where Δ is called the "gap exponent." Then we have the following:

Corollary 1: Let $\sigma^*(p,\epsilon)$ be the effective conductivity of the random resistor network in $d \geqslant 2$ dimensions, with conductivities 1 and ϵ in the volume fractions p and $1-p$. Let Δ be the gap exponent measuring the distance from the nearest singularities of $\sigma^*(p,\epsilon)$ to the percolation threshold $p = p_c$, as defined in (79). Then

$$\Delta \leqslant 1. \tag{80}$$

Proof: We prove the Corollary in $d=2$, where $p_c = \frac{1}{2}$, but the same idea holds in higher dimensions as well. We simply note that the points on the boundary of $\mathscr{D}_\epsilon$ near $p = \frac{1}{2}$ are $O(\epsilon)$ away from it. Since $\mathscr{D}_\epsilon$ forms a lower bound on the domain of analyticity, possible singularities of $\sigma^*(p,\epsilon)$ must be further away than $O(\epsilon)$. For example, if $q = \pm i(1-\epsilon)$ in (72), then $p = \frac{1}{2} \mp i\epsilon/2 + O(\epsilon^2)$.

ACKNOWLEDGMENTS

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