

Exact result for the effective conductivity of a continuum percolation model

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(Received 11 March 1994)

A random two-dimensional checkerboard of squares of conductivities 1 and δ in proportions p and $1-p$ is considered. Classical duality implies that the effective conductivity obeys $\sigma^* = \sqrt{\delta}$ at $p = \frac{1}{2}$. It is rigorously found here that to leading order as $\delta \rightarrow 0$, this exact result holds for all p in the interval $(1-p_c, p_c)$, where $p_c \approx 0.59$ is the site percolation probability, not just at $p = \frac{1}{2}$. In particular, $\sigma^*(p, \delta) = \sqrt{\delta} + O(\delta)$, as $\delta \rightarrow 0$, which is argued to hold for complex δ as well. The analysis is based on the identification of a "symmetric" backbone, which is statistically invariant under interchange of the components for any $p \in (1-p_c, p_c)$, like the entire checkerboard at $p = \frac{1}{2}$. This backbone is defined in terms of "choke points" for the current, which have been observed in an experiment.

Composite conductors typically consist of conducting particles or inclusions randomly embedded in an insulator, such as metal particles in a polymer matrix, or sea ice, which consists of pure ice containing pockets of brine. Other materials with such structure include cermets, thick-film resistors, thermistors, and piezoresistors.¹ As the volume fraction p of conducting particles is increased from zero there is a minimal \hat{p} , which coincides with the formation of a "connected" matrix of conducting particles. Typically, just as this matrix is formed, there will be many places where the contact between conductors effectively occurs only at a point, such as for polyhedral particles. We say then that the minimal dimension d_m of the conducting matrix is one, since the dimension of the current passing through the system at these points is one. As p is increased further, the degree of connectedness increases, for example, as particles may start to percolate via edges and then by faces, so that typically d_m will increase as well, with $d_m \leq d$, where d is the dimension of the system. We say that the conducting matrix is fully connected if $d_m = d$. In real materials, the effective conductivity σ^* of such composites can vary over orders of magnitude depending on the connectedness of the conducting matrix.¹⁻³ In general it is difficult to accurately calculate σ^* for such materials. However, a very useful benchmark for isotropic random two-phase media in $d=2$ with conductivities σ_1 and σ_2 arises from Keller-Dykhne interchange duality,⁴⁻⁶ $\sigma^*(\sigma_1, \sigma_2)\sigma^*(\sigma_2, \sigma_1) = \sigma_1\sigma_2$. In fact, with $\sigma_1 = 1$ and $\sigma_2 = \delta > 0$ in proportions p and $1-p$, one has

$$\sigma^* = \sqrt{\delta}, \quad p = \frac{1}{2}, \tag{1}$$

when the geometry is invariant under interchange of the components at $p = \frac{1}{2}$.

A class of models which has been successfully used to study the behavior of composite conductors, and for which the above results hold, is the symmetric-cell ma-

terials of Miller.⁷ In these models, all of space \mathbb{R}^d is divided up into cells of various shapes, which are randomly assigned the conductivities 1 (white) and $\delta > 0$ (black) with probabilities p and $1-p$. One model for which it is possible to precisely formulate the above-described connectivity questions is the random checkerboard in $d=2$, where \mathbb{R}^2 is divided into unit squares (or cubes in \mathbb{R}^3). With $\delta \ll 1$, we think of the white squares as conductors in proportion p and the black squares as insulators in proportion $1-p$. When $p < 1-p_c$, where $p_c \approx 0.59$ is the site percolation probability for the square lattice in $d=2$, nearest-neighbor black squares percolate, i.e., the black squares form an insulating matrix and are connected along edges, which prevents the formation of any type of conducting matrix. When $1-p_c < p < p_c$, there is an infinite phase of conducting squares which coexists with an infinite phase of insulating squares, where the coexistence is made possible by allowing next-nearest-neighbor, as well as nearest-neighbor connections between the squares. This generalized notion of connectedness is called star connectedness. In this case, the conducting squares are connected at corner points, as well as along edges, so that the *minimal* dimension d_m of the conducting matrix is equal to one. For $p > p_c$, nearest-neighbor conducting squares (connected along edges) percolate, so that there is a fully connected conducting matrix with $d_m = 2$.

Checkerboard models, both random and periodic versions, have been studied by numerous authors, including the works in Refs. 5 and 8-14. In the $d=2$ random case formulated above, some of these works, particularly those of Sheng and Kohn,⁸ Molchanov,⁹ and the rigorous results of Kozlov,¹⁰ have established a three-step form for σ^* as $\delta \rightarrow 0$,

$$\sigma^*(p, \delta) = \begin{cases} O(\delta^1), & p \in [0, 1-p_c) \\ O(\delta^{1/2}), & p \in (1-p_c, p_c) \\ O(\delta^0), & p \in (p_c, 1] \end{cases} \tag{2}$$

More precisely, employing the variational formulation for σ^* , Kozlov¹⁰ proved that for p in the central interval $I=(1-p_c, p_c)$ there are constants $c_1(p)>0$ and $c_2(p)>0$ such that

$$c_1(p)\sqrt{\delta} \leq \sigma^*(p, \delta) \leq c_2(p)\sqrt{\delta}, \quad \delta \rightarrow 0, \quad (3)$$

and that this $O(\sqrt{\delta})$ behavior arises from corner connections between conducting squares. Here we present the surprising rigorous result that for p throughout the central interval I ,

$$\sigma^*(p, \delta) = \sqrt{\delta} + O(\delta), \quad p \in I, \quad \delta \rightarrow 0. \quad (4)$$

That is, the Keller-Dykhne duality result of $\sqrt{\delta}$ is *exact* to leading order as $\delta \rightarrow 0$ for all $p \in I$ where corner connections dominate, not just at $p = \frac{1}{2}$. One of the surprising features of this result is that while $\sigma^*(p, \delta)$ is monotonically increasing in p for $\delta > 0$, the leading-order coefficient in front of $\sqrt{\delta}$ is independent of p , and equals one. Recall that the duality result $\sigma^* = \sqrt{\delta}$ at $p = \frac{1}{2}$ was based on the interchange invariance of the geometry at $p = \frac{1}{2}$. The analysis that we use to obtain (4), roughly speaking, shows how to extend interchange invariance away from $p = \frac{1}{2}$ where the full geometry no longer has this property. More precisely, we introduce a “symmetric” backbone containing both conducting *and* insulating squares, which is present for all $p \in I$ and which is statistically interchange invariant even for $p \neq \frac{1}{2}$. The definition of the symmetric backbone is based on the identification of what we call “choke points,” which are special corner connections through which the current is forced to flow. As we discuss below, these “hot points” have been directly observed in experiments on actual mixtures of insulating and conducting particles and are physically characterized by high-electric-field concentration and corresponding joule heating. We hope that the exact result (4) will be used as a benchmark for systems with unequal-volume fractions as (1) has been used for equal-volume fractions.

It is important to remark that the checkerboard is not simply a specific model that has no bearing on the general class of composite conductors. The choke-point analysis, which is rigorously developed here for the checkerboard, gives a framework for studying effective properties of more general composites with only a partially connected conducting matrix, i.e., when $d_m < d$. In fact, based on analysis of two- and three-dimensional random checkerboards, a scaling law for the effective conductivity of partially connected systems has been proposed.¹⁵ More precisely, for systems in the same universality class as the random checkerboards, such as conducting polyhedral particles in an insulating matrix (in $d=3$), a natural generalization of (2) has been investigated. In particular, it is proposed that $\sigma^* \sim \delta^q$ as $\delta \rightarrow 0$, where $q = (d - d_m)/2$ if $0 \leq d - d_m \leq 2$ and $q = 1$ if $d - d_m \geq 2$. In this generalization, d_m is related to the fractal dimension of the appropriate choke structure, thus applying to fractal materials, or even to checkerboards at their percolation thresholds, which is not addressed in (2). In this last regard, separate, physical arguments are used below to propose that for the $d=2$ random checkerboard at criticali-

ty, $\sigma^*(1-p_c, \delta) \sim \delta^{3/4}$ and $\sigma^*(p_c, \delta) \sim \delta^{1/4}$ as $\delta \rightarrow 0$.

We wish to mention here that findings similar to the exact result (4) have been obtained by Fannjiang and Papanicolaou¹⁶ for the case of advection-diffusion equations, where the fluid velocity field has a random checkerboard geometry. Motivated by our result, they found that for $p \in (1-p_c, p_c)$ and $\delta \rightarrow 0$ (where δ is the diffusivity of one of the fluids), the leading-order coefficient of the effective diffusivity as $\delta \rightarrow 0$ is independent of p (for a slightly modified version of the problem). The methods they use are different from ours, and do *not* depend on duality.

As a final remark before discussing how (4) is obtained, we note that it should certainly hold for complex δ , which is a case of great interest, and which would agree with some numerical results in Ref. 8. However, the arguments which yield the result for real δ do not immediately extend to the complex case, as some parts are variational in nature. A rigorous extension of (4) to complex δ rests on the following conjecture: that for any $p \in I$, $\sigma^*(p, \delta)$ is analytic in $z = \sqrt{\delta}$ in some neighborhood of $\delta = 0$ in the complex δ plane. Arguments supporting the conjecture are given below.

We now focus on the analysis of percolation geometry which leads to (4). As current passes through the conducting phase when $p \in I$, it is forced through a network of special corner connections between white squares which we call “choke points.” These connections cannot be avoided by easier, alternate routes such as a chain of white squares connected only by edges, which we call an edge chain. The absence of an easier way around means that the current must be “blocked” by a “perpendicular” star-connected chain (star chain) of insulating black squares. Thus a choke point is characterized as the central vertex at the intersection of a horizontal white-star chain with a vertical black-star chain, or vice versa, as in Fig. 1. Such star chains may be chosen to contain only edge connections and choke points, that is, with no corner connections that are not chokes. This is because if there were any corner connections that were not chokes, then there exists an alternate route (white-edge chain) around the corner connection, which can then form part of the white-star chain. Now, due to the black-white symmetry in our definition, for any $p \in I$ the choke-point

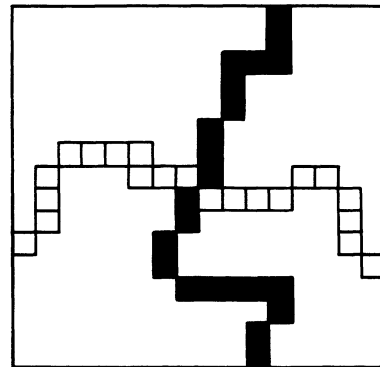


FIG. 1. Choke point at the intersection of a vertical black crossing and a horizontal white crossing (or vice versa.)

density $C(p)$ is symmetric in p , i.e., $C(p)=C(1-p)$. Note also that $C(p)\rightarrow 0$ as $p\rightarrow 1-p_c$ or $p\rightarrow p_c$. In other words, the average distance between choke points diverges as $p\rightarrow 1-p_c$ or $p\rightarrow p_c$.

The key structure that we use to extend the duality result (1) away from $p=\frac{1}{2}$ is a type of backbone appropriate to the current situation where we have two coexisting percolating phases. Associated with each choke point is a white- and black-star chain. For any realization of the square conductivities, we define $Q(p)$ for $p\in I$ to be the union of all the white- and black-star chains associated with the set of choke points. Due to the black-white symmetry inherent in the definition of choke point, the backbone $Q(p)$ is "symmetric," i.e., it is statistically invariant under the interchange of black and white ($p\rightarrow 1-p$), just as the checkerboard itself is statistically invariant under interchange at $p=\frac{1}{2}$. [A rigorous understanding of the invariance of $Q(p)$ can be obtained by noting that separated white chains which cross an $L\times L$ box alternate with black chains, which holds for all L .]

The above geometrical observations are used as follows to obtain the result (4). First, from the inequalities in (3) and the analytical properties of $\sigma^*(p,\delta)$ as a function of p and δ , it is clear that for $p\in I$,

$$\sigma^*(p,\delta)=\alpha(p)\sqrt{\delta}+O(\delta), \quad \delta\rightarrow 0, \quad (5)$$

with $\alpha(p)>0$. (However, a rigorous proof involves adapting techniques developed in Refs. 17 and 18.) Second, further exploitation of the variational method shows that the leading-order coefficient $\alpha(p)$ depends *only* on the choke-point configuration, or $Q(p)$. The contributions to σ^* from all other structures are $O(\delta)$, or more precisely, interchanging black and white in the islands off $Q(p)$ can only affect σ^* to $O(\delta)$. Finally, since the choke structure and $Q(p)$ are statistically invariant under the interchange of black and white, $\alpha(p)$ must be symmetric,

$$\alpha(p)=\alpha(1-p). \quad (6)$$

In view of the interchange theorem, which for our checkerboard takes the form

$$\sigma^*(p)\sigma^*(1-p)=\delta, \quad (7)$$

this symmetry of $\alpha(p)$ establishes the result (4).

It should now be quite clear that it is the choke points which determine the behavior of σ^* for $p\in I$. As mentioned above, such structures are not simply mathematical curiosities, but have been observed in an experiment which was explained in lectures given by Dykhne,^{9,19} and which we became aware of subsequent to our mathematical findings. The goal of the experiment was to measure σ^* for a random mixture of copper (conducting) and graphite (relatively insulating) granules in equal-volume fractions. A dense layer of the particles was pressed into a flat, soft plastic support by applying pressure with a metal plate. The particles were squashed into each other and the plastic support, so that the surface density of particles was greater than close-packing density. When current was passed through the system, the plastic support melted at a few points, which were sparsely distributed (with respect to the dimensions of the granules).

The melting of the plastic support made it impossible to measure σ^* for the system. These hot points occurred at places where two copper granules were just touching, being separated by two graphite granules, in a configuration analogous to that at the center of Fig. 1. Such a "corner connection" between two copper granules was found to be separating regions where the copper granules were in full contact, as they had been pressed into each other. The hot points observed in the experiment correspond exactly to our choke points.

The exact result (4) can be heuristically explained using a resistor-network interpretation of the white (conducting) half $Q_w(p)$ of the symmetric backbone $Q(p)$, as follows. Consider an $L\times L$ sample Λ of the random checkerboard with $p\in(1-p_c,p_c)=I$. We say that two horizontal (or vertical) white crossings of Λ (chains) are separated (or independent) if there is a horizontal black crossing of Λ between them, otherwise the two white chains would be joined by an edge-connected white chain, which is like a perfectly conducting wire as $\delta\rightarrow 0$, and vice versa for separated black crossings. The union of any set W of separated horizontal and vertical white crossings with any corresponding black set B forms the analog for the backbone $Q(p)$ of the node-link model of backbone structure.^{20,21} For $\delta\rightarrow 0$ asymptotics, we can view each chain of W as a series network of resistors of conductivity $\sqrt{\delta}$. Furthermore, we can distinguish primary chokes as those at the intersections of W and B . All other chokes in Λ are called secondary, as they arise from loops on and interconnections between crossings in W and B . The network of primary chokes forms a square lattice of these resistors, which has effective conductivity $\sqrt{\delta}$ as $L\rightarrow\infty$. As p varies in I , the average separation between resistors (chokes) varies (and diverges as $p\rightarrow p_c$ or $1-p_c$), but due to the scale invariance of σ^* in $d=2$, this analysis still gives the correct leading-order behavior as $\delta\rightarrow 0$ for $p\in I$. This situation, where the node-link picture is sufficient, should be contrasted with that when $p\rightarrow p_c$ with fixed δ , or $p=p_c$ as $\delta\rightarrow 0$ (see below), where a hierarchical node-link-blob picture is necessary,²²⁻²⁴ and one must consider secondary chokes as well.

As mentioned before, the result (4) appears to hold for complex δ , which is based on the conjecture of analyticity of $\sigma^*(p,\delta)$ in $z=\sqrt{\delta}$ near $\delta=0$. The mathematical reason behind the analyticity, we believe, goes as follows. The analytic structure of $\sigma^*(p,\delta)$ near $\delta=0$ is determined by a spectral measure in an integral representation for $\sigma^*(p,\delta)$.^{25,26} This measure depends *only* on the geometry of the checkerboard. At $p=\frac{1}{2}$, $\sigma^*=\sqrt{\delta}$, so the spectral measure yields analyticity. But the upshot of our geometrical analysis is that $Q(p)$ is essentially invariant under change of scale, or as p changes. Furthermore, $\sigma^*(p,\delta)$ is analytic in p .^{27,28} Thus, the analyticity in $z=\sqrt{\delta}$ at $p=\frac{1}{2}$ should be true away from $p=\frac{1}{2}$ as well. We note also that applicability of (4) to complex δ can most likely be established directly (without analyticity in $\sqrt{\delta}$), using techniques of complex variables.²⁹

We close with a discussion of the three-dimensional checkerboard and how it, and the $d=2$ model, shed light on conduction in partially connected systems. Consider all of \mathbb{R}^3 divided into unit cubes with conductivities as-

signed as in the $d=2$ case. As p is increased from 0, we meet three thresholds $p_c^1 \approx 0.097$, $p_c^2 \approx 0.137$, and $p_c^3 \approx 0.31161$,³⁰ which correspond to the onset of corner, edge, and face percolation, with $d_m=1$ for $p \in I_1=(p_c^1, p_c^2)$, $d_m=2$ for $p \in I_2=(p_c^2, p_c^3)$, and $d_m=3$ for $p \in I_3=(p_c^3, 1]$. For $p \in I_1$, choke points are corner connections which lie at the intersection of a “horizontal” white chain and a vertical black “sheet,” which is the analog of the black chain in Fig. 1. This sheet would be of minimal thickness 1, except where it is “pierced” by corner connections between white cubes on either side (other chokes). A similar definition holds for unavoidable edge connections for $p \in I_2$. The choke structures defined in the above way control the asymptotics of $\sigma^*(p, \delta)$ as they did in $d=2$. By extending Kozlov’s variational method to $d=3$ with the above black sheets, and using the properties of the conductivity in the neighborhood of corner and edge connections in $d=3$,^{13,14,31,32} one can obtain the following analog of (2) to $d=3$ as $\delta \rightarrow 0$:

$$\sigma^*(p, \delta) = \begin{cases} O(\delta^1), & p \in [0, p_c^2) \\ O(\delta^{1/2}), & p \in (p_c^2, p_c^3) \\ O(\delta^0), & p \in (p_c^3, 1]. \end{cases} \quad (8)$$

Note that there is no transition at p_c^1 .

As mentioned above, a general scaling law for $\sigma^*(p, \delta)$, which encompasses the results in (2) and (8), was noted in Ref. 15. While this scaling law should be viewed as a rigorous result for random checkerboards in d dimensions with p not equal to any critical point, it is tempting to extend its meaning beyond these cases, to where d_m is nonintegral. Its applicability to checkerboards at criticality, to systems of polyhedral conducting particles, and to fractal structures, is explored in Ref. 15. It is interesting to view systems of polyhedral particles in $d=3$, or

polyhedral particles in $d=2$, as examples in the universality class of the checkerboards, to which the scaling law applies, and defined by the condition that connections between conductors in, say, $d=2$, are formed from corners and edges, or mixtures thereof. The existence of this universality class is made possible by the fact that the exponent $q=\frac{1}{2}$ arising locally at a corner connection in $d=2$ is independent of the contact angle, i.e., $q=\frac{1}{2}$ is true for parallelograms,^{14,10} and even random polygons,¹⁸ as well as squares.

In the case of the $d=2$ checkerboard, a physical argument can be used to conjecture the scaling behavior of $\sigma^*(p, \delta)$ at $p=1-p_c$ and p_c . At $p=1-p_c$, in light of the resistor-network interpretation of the checkerboard, and using the universality of percolation thresholds, we can view our checkerboard as a $d=2$ bond lattice with resistors of conductivity δ and $\sqrt{\delta}$ at criticality, i.e., with equal-volume fractions. Then duality yields $\sigma^*(1-p_c, \delta) \sim (\delta\sqrt{\delta})^{1/2} = \delta^{3/4}$ as $\delta \rightarrow 0$. Applying similar reasoning at $p=p_c$ to a resistor network with conductivities $\sqrt{\delta}$ and 1 gives $\sigma^*(p_c, \delta) \sim \delta^{1/4}$ as $\delta \rightarrow 0$. It is interesting to compare these results with the general scaling law. The implication is that $d_m=\frac{1}{2}$ at $p=1-p_c$ and $d_m=\frac{3}{2}$ at $p=p_c$. A rigorous definition of d_m at criticality is given in Ref. 15, and the plausibility of these implications is investigated through inequalities relating d_m to the fractal dimensions of the backbone and “red bonds” for lattice-percolation problems in $d=2$ and 3.

The authors would like to thank the following people for very helpful conversations: M. Aizenman, S. Kozlov, G. Milton, S. Molchanov, and A. Volberg. K.G. was supported in part by NSF Grants No. DMS-9204328 and No. DMS-9307324, and ONR Grant No. N000149310141.

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