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Scaling law for conduction in partially connected systems

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Abstract

The electrical transport properties of systems of conducting particles embedded in an insulator are considered. For low volume fractions of the particles, the conducting matrix may only be "partially" connected, as particles may only touch at corners or edges. As a model where these connectedness questions can be precisely formulated, we consider a random checkerboard in dimensions $d=2$ and 3, where the squares in $d=2$ or cubes in $d=3$ are randomly assigned the conductivities 1 with probability p or $0<\delta \ll 1$ with probability $1-p$. To analyze connectedness, we introduce a new parameter, d_{m}, called the minimal dimension, which measures connectedness of the conducting matrix via the dimension of the dominant contacts between particles. Based on analysis of the checkerboards, we propose a general scaling law for the effective conductivity σ^* as $\delta \to 0$, namely $\sigma^* \sim \delta^q$, where $q=\frac{1}{2}(d-d_{\mathrm{m}})$ for $0 \leqslant d-d_{\mathrm{m}} \leqslant 2$ and $q=1$ for $d-d_{\mathrm{m}} \geqslant 2$. The applicability of this law to situations where d_{m} is non-integral, such as the checkerboards at criticality, is discussed in detail.

Composite conductors, such as cermets, thick-film resistors, and piezoresistors, typically consist of conducting articles embedded in an insulating matrix. If the particles are quite expensive, such as gold or silver, then it is useful to find the minimal volume fraction p required for the formation of a "connected" matrix of conductors which achieves the desired properties. However, for many systems, the *degree* of connectedness can vary significantly over a small range in p, which can be accompanied by the effective conductivity σ^* varying over orders of magnitude [1]. For example, if we consider polyhedral particles in dimension $d=3$, then as p is increased from zero, there is a minimal $\hat{p}$ for which there is a connected matrix of particles. Just as this matrix is formed, there will be many places where the contact between particles effectively occurs at a point, or vertex. As p is increased the degree of connectedness increases as the "predominant" contacts increase in dimension, through edges and faces. In this note we propose

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a general scaling law which describes σ^* for composite conductors when the conducting matrix is only partially connected.

As a model where the above connectedness questions can be precisely formulated, we consider the random checkerboard in $\mathbb{R}^d$, where for $d=2$, $\mathbb{R}^2$ is divided up into unit squares which are randomly assigned the conductivities 1 (white) with probability p and $\delta \ll 1$ (black) with probability $1-p$. When $p<1-p_c$, where $p_c \approx 0.59$ is the site percolation probability, nearest-neighbor black squares (connected by edges) percolate, which prevents the formation of any type of conducting matrix. When $p>p_c$, white nearest-neighbor squares percolate and the conducting matrix is fully connected. However, for $1-p_c<p<p_c$, there is an infinite phase of white squares which coexists with an infinite phase of black squares, where the coexistence is made possible by allowing next-nearest-neighbor (or corner connections), as well as nearest-neighbor connections between squares, which is called star-connectedness. In order to characterize the degree of connectedness of the conducting matrix, we introduce the minimal dimension d_m of the matrix, which is unity for $1-p_c<p<p_c$ since the current is forced to constrict to being one-dimensional where it passes through corner connections. For $p>p_c$, $d_m=2$ since it can always pass through edge connections between conducting squares, in which case we say the matrix is fully connected. For $p<1-p_c$, $d_m=0$.

The effective conductivity σ^* for checkerboard models, both random and periodic, has been studied in numerous works. For $d=2$ there is a classical, exact result $\sigma^*=\delta^{1/2}$ at $p=\frac{1}{2}$ [2], which arises from the duality relation $\sigma^*(p)\,\sigma^*(1-p)=\delta$ [3,2]. For general p, the following three-step form for σ^* as $\delta \to 0$ has been established [4–6]:

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

In fact, it has been recently shown that the exact result $\sigma^*=\delta^{1/2}$ holds to leading order as $\delta\to 0$ for all p in the interval $(1-p_c,p_c)$, not just at $p=\frac{1}{2}$ [7],

$$\sigma^*(p,\delta)=\sqrt{\delta}+O(\delta)\,, \qquad \delta\to 0\,, \quad p\in(1-p_c,p_c)\,. \tag{2}$$

The analysis used to obtain (2) is based on the identification of a network of special corner connections between white squares which we call "choke points." These connections cannot be avoided by easier, alternative 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 (i.e., one which connects the left side of an $L\times L$ box to the right side) with a vertical black star chain, or vice versa. Now, due to the black–white symmetry in our definition, for any $p \in (1-p_c, p_c)$ the choke-point density $C(p)$ is symmetric in p, i.e., $C(p) = C(1-p)$. Note also that $C(p) \to 0$ as $p \to 1-p_c$ or $p \to p_c$. In other words, the average distance between choke points diverges as $p \to 1-p_c$ or $p \to p_c$. We remark that choke points have been directly observed in an experiment involving copper and graphite granules [5,7], and were manifested as hot points which melted the plastic platform into which the particles were pressed.

The key structure associated with the choke network that we use to extend the duality result $\sigma^* = \delta^{1/2}$ away from $p = \frac{1}{2}$ is a new 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 $p \in (1-p_c, p_c)$ 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 $(p \to 1-p)$, just as the checkerboard itself is statistically invariant under interchange at $p = \frac{1}{2}$. [A rigorous understanding of this 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.]

For the $d = 3$ random checkerboard of cubes we meet three thresholds $p_c^{(1)} \approx 0.10$, $p_c^{(2)} \approx 0.14$ and $p_c^{(3)} \approx 0.31$, which correspond, respectively, to the onset of percolation by corners, edges and faces, 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 $p \in I_3 = (p_c^{(3)}, 1]$. By extending Kozlov's variational method [6] to $d = 3$ and using the properties of σ^* in the neighborhood of corner and edge contacts in $d = 3$ [8–11], one can show that σ^* has the same three-step form as in (1) with $p_c^{(2)}$ replacing $1 - p_c$, and $p_c^{(3)}$ replacing p_c. Note that there is no transition at $p_c^{(1)}$, so that percolating by corners is not enough to increase the order of σ^* beyond that for no conducting matrix.

In view of the scaling behavior established for the checkerboard in $d = 2$ and 3, it is interesting to investigate the dependence of the exponent q, with $\sigma^* \sim \delta^q$, as $\delta \to 0$, on the degree of connectedness of the conducting matrix, as measured by d_m. This dependence is shown in Fig. 1, which immediately leads to the following scaling law for σ^* as $\delta \to 0$:

$$\sigma^* \sim \delta^q , \quad q = \begin{cases} \frac{1}{2}(d - d_m) , & 0 \leq d - d_m \leq 2 , \\ 1 , & d - d_m \geq 2 . \end{cases} \tag{3}$$

The cut-off of $q = 1$ for $d - d_m \geq 2$ says that the contact between conductors must be of high enough dimension in order to achieve conduction greater than $O(\delta^1)$. While (3) should be viewed as a rigorous result for the random checkerboard in d dimensions with p not equal to any critical point, it is tempting to extend its meaning to situations where an appropriately defined d_m is non-integral, such as the checkerboard *at* criticality, or other fractal media. In order to define d_m for the checkerboard at criticality, let us focus on $d = 2$ at $p = 1 - p_c$, and note that

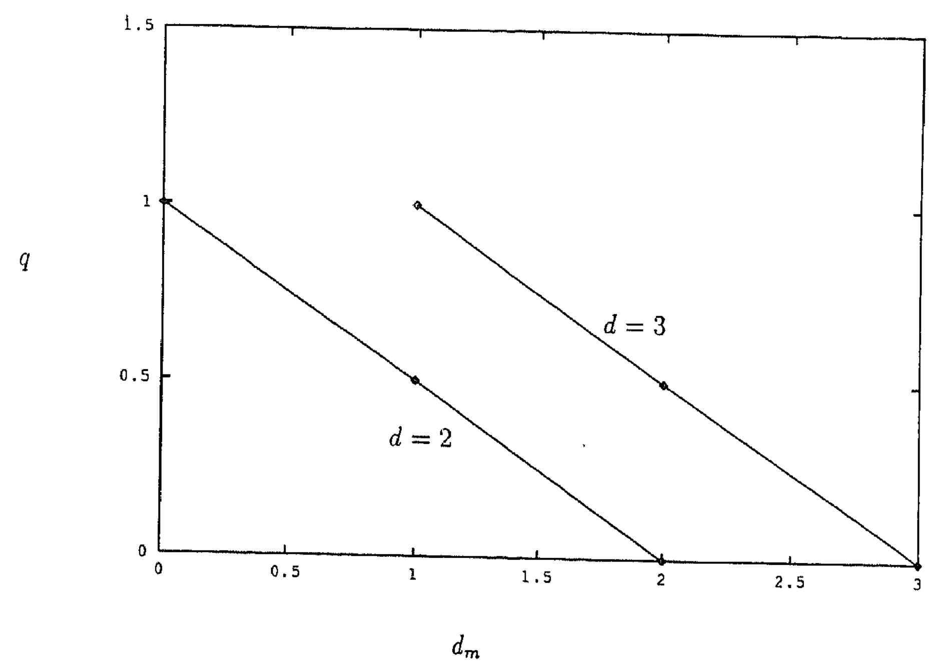

Fig. 1. Dependence of q, as $\delta \to 0$, on d_{m}.

any reasonable definition must yield $0 \leq d_{\mathrm{m}} \leq 1$, since $d_{\mathrm{m}} = 1$ for p just above $1 - p_{\mathrm{c}}$ and $d_{\mathrm{m}} = 0$ for p just below $1 - p_{\mathrm{c}}$. Now let $Q_{\mathrm{w}}(p)$ be the conducting (white) part of the symmetric backbone $Q(p)$, for $p \in (1 - p_{\mathrm{c}}, p_{\mathrm{c}})$, and let A be an $L \times L$ sample of the checkerboard. It is important to note that given a realization of the entire conducting backbone $B(p)$, any choice for $Q_{\mathrm{w}}(p)$ is strictly contained inside $B(p)$, since a cluster of star-connected squares in $B(p)$ may be reduced to some star-chain in $Q_{\mathrm{w}}(p)$ running through the cluster.

We view the relevant part of the partially connected conducting matrix at $p = 1 - p_{\mathrm{c}}$ to be the "incipient infinite cluster" left over from $Q_{\mathrm{w}}(p)$ as $p \to (1 - p_{\mathrm{c}})^{+}$, and define the fractal dimension d_Q of $Q_{\mathrm{w}}(p)$ as follows. Let m be the number of squares in $Q_{\mathrm{w}}(p)$, so that

$$m \sim L^{d_Q}, \qquad L \to \infty . \tag{4}$$

For $p \in (1 - p_{\mathrm{c}}, p_{\mathrm{c}})$, $d_Q = 2$. Then we define

$$d_{\mathrm{m}} = d_Q / d , \tag{5}$$

so that $d_{\mathrm{m}} = 1$ for $p \in (1 - p_{\mathrm{c}}, p_{\mathrm{c}})$ in $d = 2$, which can be interpreted as the fractal dimension of the crossings by $Q_{\mathrm{w}}(p)$ of, say, a vertical, unit width strip of A. Now, from the relation $Q_{\mathrm{w}}(p) \subseteq B(p)$ discussed above, we clearly have $d_Q \leq d_B$, where d_B is the fractal dimension of the backbone. Note that d_B is universal for $d = 2$ (or $d = 3$) lattice problems, independently or whether "connection" is defined via 1st, 2nd or whatever nearest neighbor, as is d_Q.

Through this universality, it is clear that the analog of $Q_w(p)$ for a $d=2$ bond problem contains the set of "red" or singly connected bonds, so that $d_Q \geq d_{red}$, where d_{red} is the fractal dimension of the red bonds. Combining our inequalities, we have

$$d_{red} \leq d_Q \leq d_B \,. \tag{6}$$

Using Coniglio's result that $d_{red} = 1/\nu$ [12], where ν is the correlation length exponent ($\nu = \frac{4}{3}$, $d=2$), and numerical estimates for d_B [13], we have for $d=2$, $0.38 \leq d_m \leq 0.81$ at $p = 1 - p_c$. We obtain inequalities for d_m at p_c by noting that $d_m(p_c) = d_m(1-p_c) + 1$, since we are simply adding a dimension in the contact. For $d=3$, we have from (6), $0.38 \leq d_m \leq 0.58$ at $p = p_c^{(1)}$. Again, $d_m(p_c^{(2)}) = d_m(p_c^{(1)}) + 1$, and so on.

It is very interesting to compare (3) with the following (conjectured) exact scaling results in $d=2$ obtained from physical argument [7]. At $p = 1 - p_c$, universality at critical points and a resistor network interpretation allow the checkerboard to be viewed as a bond lattice in $d=2$ at criticality ($p = \frac{1}{2}$) with resistors of conductivities δ and $\delta^{1/2}$, so that from duality $\sigma^* \sim (\delta\delta^{1/2})^{1/2} = \delta^{3/4}$. Comparing with (3) implies $d_m = \frac{1}{2}$ at $p = 1 - p_c$, which is consistent with the above inequality. In particular, this implies that the universal fractal dimension d_Q takes the value 1 in $d=2$. At $p = p_c$, similar arguments give $\sigma^* \sim \delta^{1/4}$, which implies $d_m = \frac{3}{2}$, which is consistent with $1.38 \leq d_m \leq 1.81$ at $p = p_c$. For $d=3$ we have no argument yielding exact results for q at $p_c^{(2)}$ and $p_c^{(3)}$, although (6) and (3) imply

$$0.71 \leq q \leq 0.81 \,, \quad p = p_c^2 \,, \qquad 0.21 \leq q \leq 0.31 \,, \quad p = p_c^3 \,. \tag{7}$$

We expect that the scaling law (3) should apply to a universality class of continuum percolation models which contains the checkerboards, and is defined by the condition that connections between conductors are formed from corners, edges and faces, or mixtures thereof. Examples of systems in the class should include polyhedral particles in $d=3$ or polygonal particles in $d=2$, Miller's cell materials [14] with appropriate cells, or systems of conducting particles with fractal connections, such as cubes whose contact is a Cantor set, rather than an edge. The existence of this universality class is made possible by the fact that the exponent $q = \frac{1}{2}$ at corners for $d=2$ is independent of the contact angle, so that it is valid for parallelograms [9,6], and even for random polygons [15], as well as squares.

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