CRITICAL PATH ANALYSIS OF TRANSPORT IN HIGHLY DISORDERED RANDOM MEDIA

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In problems of conduction and fluid flow through complex random media, systems with a broad distribution in the local properties are often encountered. Here we introduce a continuum percolation model for such media, which is exactly solvable for the effective transport properties in the high disorder limit. The model represents such systems as fluid flowing through consolidated granular media and fractured rocks, as well as electrical conduction in some matrix-particle composites near critical volume fractions. Moreover, the results provide a rigorous basis for the widely used Ambegaokar, Halperin, and Langer critical path analysis [1]. This method rests on the proposition that transport in media with a broad range of local conductances g is dominated by a critical conductance g_c, which is the smallest conductance such that the set $\{g \mid g > g_c\}$ percolates, and in our model this proposition is rigorously established.

A central issue in the theory of transport in disordered materials is to determine the effective properties, such as electrical conductivity and fluid permeability, from some knowledge of the microstructural features of the material. In many problems of technological importance, one meets systems which display a wide distribution in the local properties characterizing the system. For example, in porous media there is often a wide range of pore and neck sizes through which the fluid must flow [2, 3, 4, 5, 6, 7, 8]. As another example, in resistor network models of hopping conduction in amorphous semiconductors, the bonds of the network are assigned a wide range of conductivities [1, 9, 10]. A powerful idea which has been widely used [2] to estimate the effective properties of such systems is the *critical path analysis*, which was first introduced by Ambegaokar, Halperin and Langer (AHL) [1]. They proposed that transport in a medium with a broad range of local conductances g is dominated by a critical value g_c, which is the smallest conductance such that the set $\{g \mid g > g_c\}$ percolates, or forms a connected cluster which spans the sample. This cluster is called the critical path. Then the problem of estimating transport in a highly disordered medium with a wide range of local conductances is reduced to a percolation problem with threshold value g_c. Critical path analysis was further developed in the context of amorphous semiconductors in [9] and [10], and its accuracy was numerically confirmed for various conductance distributions in [11]. It has also been used to analyze the permeability and electrical conductivity of porous media, such as sandstones, as obtained through mercury injection [12, 13, 14, 15], as well as fractured rocks with a broad distribution of fracture apertures [16], and porous media saturated with a non-Newtonian fluid [17].

While critical path analysis has been applied with substantial success, there has been little rigorous work on the fundamental observation that the critical conductance g_c dominates the effective behavior. Here we introduce a continuum percolation model of conducting and porous random media which is exactly solvable in the high disorder limit, and the result we obtain for the effective conductivity or permeability rigorously establishes the AHL principle for this model. Furthermore, our model closely represents an important class of porous materials, including consolidated granular media and some fractured rocks, where the easiest flow paths or channels exhibit a complex random topology, similar to a Voronoi network, as in Fig. 1 [2, 18, 19, 20, 6].

We obtain our model as a "long-range" generalization of the random checkerboard in $\mathbf{R}^2$ [21, 22, 23, 24, 25, 26, 27], where the squares are assigned the conductivities 1 with probability p and $\delta > 0$ with probability $1-p$. The random checkerboard has been used to model conducting materials which exhibit critical behavior too rich to be accurately handled by random resistor networks, such as graphite (conducting) particles embedded in a polymer (insulating) matrix. For example, the presence of both corner and edge connections between squares produces two percolation thresholds, with distinct asymptotic behaviors of the effective conductivity as $\delta \to 0$ (or $\delta \to \infty$) in the different regimes of p separated by these thresholds [22, 24, 25, 26]. In our model we allow arbitrarily long-range connections between squares, which leads to infinitely many thresholds, and rather complex asymptotic behavior, which we can nevertheless obtain exactly. Our analysis is based on the variational formulation of effective properties, which allows us to obtain bounds required for the asymptotics, by constructing trial fields which exploit relevant percolation structures [25, 26, 27]. A key com-

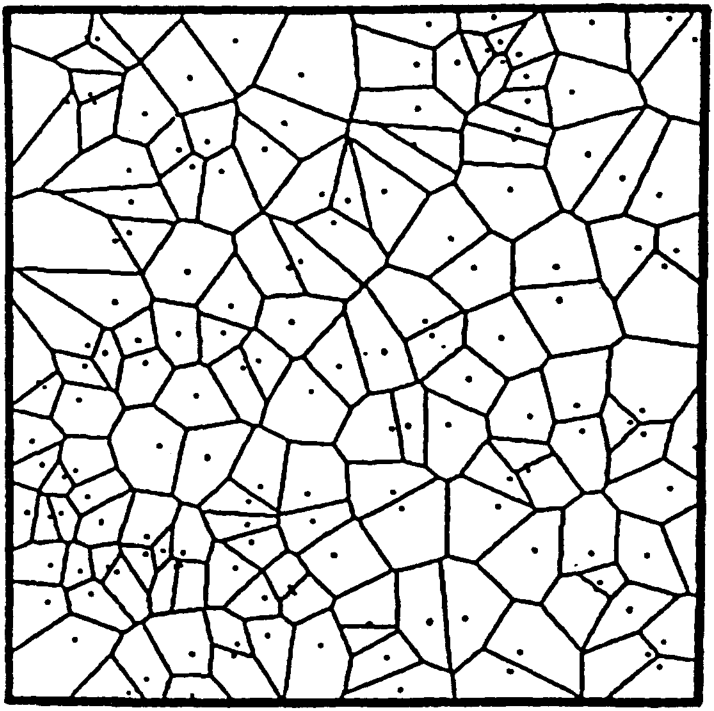

Figure 1: Two dimensional Voronoi tessellation (from [2]). The boundaries of the polygonal grains are formed from points which are equidistant to the dots in two neighboring grains. These boundaries form the channels of easiest flow in consolidated granular media and fractured rocks

ponent of our percolation analysis is to connect our model to a Poisson distribution of discs in the plane.

Whereas the standard checkerboard model allows for only two types of connections between conducting particles (squares), our generalization allows for arbitrarily many. This leads us to expect that our model may serve as a good representation for high contrast matrix-particle composites, particularly in the regime where the conducting particles "percolate," yet are of low enough volume fraction so that the conducting phase is only "partially connected" [27], where there is a broad distribution of connection types between the conducting particles. In such regimes the effective conductivity has been observed to vary over many orders of magnitude [28] over small volume fraction ranges, as does our model. More precisely, in the high disorder limit, the paths of easiest current flow in our model form a (continuum) Voronoi network with a broad distribution of "bond" conductivities. This network can be identified with the one obtained from joining the centers of "touching" conducting particles. The quality of the connection between the particles determines the conductivity of the path joining them. Our model can be modified to handle such problems, and our variational analysis, which is quite general, can be suitably applied to this and other related problems.

We now formulate the mathematical model. For simplicity, we give the formulation for electrical conductivity in $d = 2$, but the model and result can be carried over to $d = 3$, as well as fluid flow in a porous medium obeying Darcy's law, which will be discussed later. Consider the checkerboard of unit white squares in $\mathbf{R}^2$, with centers of the squares being the points of the lattice $\mathbf{Z}^2$. Randomly color the squares red with probability p, where the probability of coloring one square is independent from any other. Then for $x \in \mathbf{R}^2$, let $S(x)$ be the distance from x to the boundary of the nearest red square, with $S(x) = 0$ if x is inside a red square. Then define the local conductivity $\sigma(x)$ as

$$\sigma(x) = e^{\lambda S(x)}. \tag{1}$$

It is useful to think of the red squares as insulating particles, as we will be considering asymptotics as $\lambda \to +\infty$ (although we could just as easily consider $\lambda \to -\infty$). The medium defined by (1) can be thought of as being divided into grains associated with each red square, where each grain is the set of all points for which the distance to its red square is smaller than the distance to any other red square. The boundaries between these grains, where the distance to a red square is maximal, form the "channels" through which current (or fluid) passing through the medium will tend to flow. For small p, the set of boundaries forms a Voronoi network, as shown in Fig. 1. The points in the figure represent the red squares.

Our goal is to find the $\lambda \to +\infty$ asymptotics of the effective conductivity $\sigma^*(p)$ of the medium in (1), which is defined as follows (e.g. [29, 30]). Let $E(x)$ and $J(x)$ be the stationary random electric and current fields in the medium satisfying $J(x) = \sigma(x)E(x), \nabla \cdot J = 0, \nabla \times E = 0$, and $< E(x) >= e_k$, where e_k is a unit vector in the k^{th} direction, and $< \cdot >$ denotes ensemble or infinite volume average. Then the effective conductivity σ^* is defined via

$$< J >= \sigma^* < E > . \tag{2}$$

In order to state the results for the asymptotics and to more fully describe our model, we first consider standard nearest neighbor site percolation on $\mathbf{Z}^2$ [31, 32, 33], which is equivalent to the percolation of nearest neighbor red squares (connected along an edge), with percolation threshold $p_c^s \approx 0.59$. Now we relax the nearest neighbor restriction for connectedness, and consider a generalized definition of percolation of the red squares. We say that two red squares $\hat{x}$ and $\hat{y}$ with centers x and y in $\mathbf{Z}^2$, are r-connected if there is a sequence $\hat{x}_0,\ \hat{x}_1, \cdots, \hat{x}_n,\ \hat{x}_0 = \hat{x},\ \hat{x}_n = \hat{y}$ of red squares connecting them such that $dist\{\hat{x}_i,\ \hat{x}_{i+1}\} \leq r$, where $dist\{\hat{x}_i,\ \hat{x}_{i+1}\}$ means the shortest distance between the boundaries of $\hat{x}_i$ and $\hat{x}_{i+1}$. With $\theta_r(p)$ the infinite cluster density of r-connected red squares, we define $p_c(r)$ as the percolation threshold for $\theta_r(p)$. We shall be concerned with a particular sequence $r_j,\ j \geq 1$, defined by squares which are increasingly distant from $\hat{0}$, the square centered at the origin, with $r_1 = 0,\ r_2 = 1,\ r_3 = \sqrt{2},\ r_4 = 2,\ r_5 = \sqrt{5},\ r_6 = \sqrt{8}, \cdots$. Note that it suffices to consider squares with centers $(m, n) \in \mathbf{Z}^2$ with $n \geq m \geq 0,\ n > 0$. For simplicity we denote $p_c(r_j)$ as $p_c(j)$, and we also replace the term r_j-connected by j- connected. Note that the $j = 1$ $(r_1 = 0)$ case includes both the nearest neighbor case above and the next nearest neighbor (diagonal) case (since the distance between connected squares is 0 in both cases), so that $p_c(1) = 1 - p_c^s \simeq 1 - 0.59 = 0.41$ [25, 26, 27]. Furthermore $p_c(j+1) < p_c(j)$, which can be obtained from [34]. From analysis of a

Poisson distribution of discs in the plane (see below), one can also find that

$$p_c(j) \sim \frac{\pi \mu_c}{j} , \qquad j \to \infty , \tag{3}$$

where μ_c is the critical percolation intensity for unit discs [33].

The last ingredient we need to state the results is the notion of critical values of $S(x)$, which are associated with the $p_c(j)$. These values $S_c(j)$ are defined by the observation that for $p > p_c(j)$, the set $R_j = \{x \in \mathbf{R}^2 : S(x) \leq S_c(j)\}$ percolates in $\mathbf{R}^2$, where for j associated with $(m, n) \in \mathbf{Z}^2$ as above,

$$S_c(j) = r_j/2 = \sqrt{(1 - \delta_{m0})(m-1)^2 + (n-1)^2}\Big/2 , \tag{4}$$

where δ_{m0} is the Kroneker delta. By percolating in $\mathbf{R}^2$ we mean that R_j contains an infinite polygonal line joining vertices of the red squares in R_j. Note that R_1 is just the set of red squares, which percolates in the above sense when $p > p_c(1)$. Now in terms of the $S_c(j)$, define a step function $S_c(p)$ via

$$S_c(p) = S_c(j) , \quad p_c(j) < p < p_c(j-1) , \; j \geq 1, \tag{5}$$

where $p_c(0) = 1$. These critical distances $S_c(j)$ in our model correspond via (1) to the critical conductances g_c in the AHL theory.

The principal results of our investigation are now stated as follows. For the effective conductivity $\sigma^*(p)$ of the medium $\sigma(x) = e^{\lambda S(x)}$, we have for $p \neq p_c(j)$,

$$\frac{1}{\lambda} \, log \; \sigma^*(p) \sim S_c(p), \qquad \lambda \to \infty, \tag{6}$$

which establishes the validity of the AHL critical path analysis in the high disorder limit for our model. Furthermore, we have the following $p \to 0$ asymptotics for the exponents,

$$S_c(p) \sim \sqrt{\frac{\mu_c}{p}} , \qquad p \to 0 , \tag{7}$$

where p is the density of red squares (i.e., with units of inverse square length). Via (6) and (7), we see that $\sigma^*(p)$ for our long range checkerboard model exhibits infinitely many thresholds $p_c(j) \to 0$ as $j \to \infty$

with an infinite set of asymptotics as $\lambda \to \infty$. If instead of $\lambda \to \infty$ we wish to put $\lambda = 1$ so that $\sigma(x) = e^{S(x)}$, and consider the asymptotics of $\sigma^*(p)$ as $p \to 0$, we also find that

$$\sqrt{p}\; log\; \sigma^*(p) \sim \sqrt{\mu_c}\;, \qquad p \to 0\;. \tag{8}$$

We now give the analysis which leads to these results. The idea is to exploit the variational definition of σ^* equivalent to (2) and its dual, to obtain upper and lower bounds on σ^*. Let $\Lambda_N = [0, N] \times [0, N] \subset \mathbf{R}^2$. Then the variational form of (2) is

$$\sigma^* = \lim_{N\to\infty} \frac{1}{N^2} \inf_{u\in\mathcal{P}} \int_{\Lambda_N} \sigma(x)\; |\nabla u|^2 dx\;, \tag{9}$$

where $\mathcal{P} = \{$continuous potentials u on $\Lambda_N : u(0, x_2) = 0\;,\; u(N, x_2) = N, \forall x_2 \in [0, N]\}$. We obtain bounds by inserting trial u into (9). To describe the construction, we recall certain properties of standard site percolation. It has been shown [35, 36] that for $p > p_c^s$ the number α_N of disjoint paths through the occupied phase, (formed from sequences of nearest neighbor, occupied sites) which cross Λ_N horizontally or vertically, or disjoint crossings, satisfies (roughly speaking) $\alpha_N = O(N)$ as $N \to \infty$. In our generalized model, recall that for $p > p_c(j), R_j = \{x \in \mathbf{R}^2 : S(x) \leq S_c(j)\}$ percolates in $\mathbf{R}^2$. In this case, the number $\alpha_N(j)$ of disjoint crossings of Λ_N by j-connected red squares also satisfies $\alpha_N(j) = O(N)$ as $N \to \infty$. We call the associated disjoint subsets of R_j that cross Λ_N pink "j-chains" (we say "pink" because such sets contain both red squares and parts of white ones). It will be necessary for our purposes to consider only those chains which cross vertically. Note that these j-chains can be viewed as ribbons which at some points may have zero width, for example at corner connections between red squares with $j = 1$ and $r_1 = 0$, with $p_c(1) = 1 - p_c^s < p < p_c^s$. In this case, note that both R_1 and its complement in $\mathbf{R}^2$ percolate, with similar behavior for $j > 1$. (In standard nearest neighbor lattice percolation models, double connectivity, or simultaneous percolation of the two phases, does not occur in two dimensions, but only in higher dimensions.) Now, the trial u is constructed as follows, using a similar technique to that which has been used for the random checkerboard [25]. On the j-chains u increases linearly across the chain, in such a way that the total contribution to (9) of $|\nabla u|^2$ on the pink j-chains is $O(N^2)$. In each region between the j-chains, u is a constant (so that

$\nabla u = 0$), which, along with additive constants for u on the j-chains, are chosen so that u is continuous, and it "steps up" as one moves across Λ_N. Constructing u in the neighborhood of points where the j-chains have zero thickness is handled with asymptotic expansions, which are patched continuously to the rest of u, as in [25] for the so-called Laplace-Dirichlet integral. For such u then, the integrand in (9) is zero off the j-chains, and for x in the pink j-chains,

$$\sigma(x) \leq e^{\lambda S_c(j)}, \tag{10}$$

which leads to the inequality

$$\sigma^*(p) \leq C_1 e^{\lambda S_c(j)} \ , \qquad p > p_c(j) \ , \tag{11}$$

for some $C_1 > 0$ (depending on p), with $j \geq 1$. Another trial potential which gives the same type of bound (pointed out to us by a referee) is the actual potential for a medium which has unit conductivity in R_j and is superconducting off R_j.

To get the lower bound, we first note that a dual formulation of (9) can be obtained by replacing σ^* and $\sigma(x)$ by $(\sigma^*)^{-1}$ and $\sigma^{-1}(x)$, respectively. The key observation in the analysis now is that for $p < p_c(j-1), j \geq 1$ (with $p_c(0) = 1), W_j = \{x \in \mathbf{R}^2 : S(x) \geq S_c(j)\}$ percolates in $\mathbf{R}^2$, which can be seen as follows. When $p < p_c(j-1)$, R_{j-1} cannot percolate, $j \geq 2$. In this case, easy geometric reasoning shows that there must exist infinite chains of white cells, such that the minimal thickness r_j of these white chains (meaning that discs of radius $r_j/2$ percolate in these chains) is $2S_c(j)$. Then $Wj = \{x \in \mathbf{R}^2 : S(x) \geq r_j/2 = S_c(j)\}$, which contains this set of white chains, percolates in $\mathbf{R}^2$. Now constructing u similar to that above, one obtains

$$[\sigma^*(p)]^{-1} \ \leq \ C_2 e^{-\lambda S_c(j)} \ , \qquad p < p_c(j-1) \ , \tag{12}$$

for some $C_2 > 0$ (depending on p) , with $j \geq 1$. Combining (11) and (12) yields (6).

In order to obtain the asymptotic behaviors of the thresholds in (3) and the exponents in (7), we connect our work to the analogous problem for a Poisson distribution of discs in $\mathbf{R}^2$. Let $\{x_k\}_{k=1}^{\infty}$ be a set of Poisson distributed red points in the plane, with intensity μ. First we define, analogously to (1), $S_\mu(x) = dist\{x, \text{ nearest } x_k\}$ and $\sigma_\mu(x) = e^{\lambda S_\mu(x)}$. Let S_μ^c be the smallest h for which $\{x \in \mathbf{R}^2 : S_\mu(x) \leq h\}$ percolates.

Then S^c_μ coincides with r^c_μ, the minimum radius such that the discs of radius r^c_μ centered at the x_k percolate. The above arguments used for the long range checkerboard yield for the effective conductivity σ^*_μ in this case

$$\frac{1}{\lambda}\, log\ \sigma^*_\mu \sim S^c_\mu = r^c_\mu\ , \qquad \lambda \to \infty. \tag{13}$$

We remark that, via the scaling properties of the Poisson model, we can replace $\lambda S_\mu(x)$ by $\frac{\lambda}{\sqrt{\mu}}\ S_1(x)$, so that we may set $\lambda = 1$ and consider asymptotics as $\mu \to 0$, with a result analogous to (13).

It is now useful to note that the above Poisson model can be obtained by rescaling our checkerboard model, where the red squares of our model correspond to the x_k of the Poisson model, as follows. On the scaled lattice $h\mathbf{Z}^2, h > 0$, let the density of red squares be $p/h^2 = \mu$. As $p \to 0$, with $h = \sqrt{p/\mu} \to 0$ as well, $\hat{S}(x,p) = h\ S(x/h, p) \to S_\mu(x)$. Then the critical values also converge, $\hat{S}_c(p) \to S^c_\mu$ as $p \to 0$. Then with $\hat{S}_c = hS_c$ and $h = \sqrt{p/m}$, setting $\mu = 1$ yields $\sqrt{p}S_c(p) \to r^c_1$ as $p \to 0$, which is equivalent to (7), since $\mu_c = (r^c_1)^2$. Furthermore, the critical density of red points on the rescaled lattice is $p_c(j)/h^2$, so that $\mu_c = \lim_{j\to\infty} \frac{p_c(j)}{h^2}$. Note that $p_c(j)$ is the critical p for which disks of radius r_j percolate. So if we rescale the lattice with $h \sim 1/r_j$ as $j \to \infty$, then unit discs percolate, so that

$$\mu_c = \lim_{j\to\infty}\ p_c(j) r_j^2\ . \tag{14}$$

To relate r_j to j, we note that there are $O(j)$ integer points inside the disc of radius r_j as $j \to \infty$, so that

$$j \sim \pi r_j^2\ , \quad j \to \infty\ , \tag{15}$$

which combined with (14) yields (3).

In closing we wish to make a few remarks. Presumably an effective medium approach as in [22] could give an accounting of the behavior of our model for small j. However, as j grows, the number of configurations of squares that must be considered grows extremely rapidly, and numerical calculations become intractable.

An interesting question is the transition between different exponents for large λ as p crosses the threshold $p_c(j)$. We remark that, for example, the constant C_1 in estimate (11) diverges like $\xi(p)$ as $p \to p_c(j)^+$, where $\xi(p)$ is the correlation length for $j-$percolation.

Finally, for fluid flow (with unit viscosity) in porous media [2] obeying Darcy's law $v = -K(x)\nabla P$, where $K(x)$ is the local permeability, v is the fluid velocity satisfying $\nabla \cdot v = 0$, and P is the pressure (including gravity), one is interested in the effective permeability K^*, defined analogously to (2). As briefly mentioned earlier, if $K(x)$ has the form (1), then for large λ it is a close model for flow through consolidated granular media, where the grains themselves are permeable, with decreasing permeability as one approaches a hard core. Fig. 2 shows a computer simulation of a grain consolidation process [19]. The sequence (a) - (d) shows increasing consolidation, and correspondingly decreasing porosity. As $\lambda \to \infty$ the network of easiest flow paths in our model closely resembles the configuration in (d), which itself is similar to many types of sedimentary rocks, including Devonian sandstone [19].

Reminiscence: (from K.G.)

The work represented in this paper evolved over a period of a few years beginning in August 1991, when I visited Serguei Kozlov in Moscow, which turned into a very eventful time. During our initial investigations of the model considered here, our work was interrupted by the coup against Gorbachev, which led to the rise of Yeltsin, and the fall of the Soviet Union. During the first day, Serguei and I ventured into Red Square and saw the tanks assembled, on the last day we attended Yeltsin's speech from the Russian "White House," and there were some very tense times in between. It was a most memorable sequence of events, and I am grateful that I was able to experience it with Serguei and his family. We continued this work after Serguei moved to the south of France, and had some very enjoyable and productive periods there. During the times that we spent together, I came to greatly appreciate the depth, clarity, and power of Serguei's ideas, as well as his friendship. His untimely death is truly a loss, and he is deeply missed.

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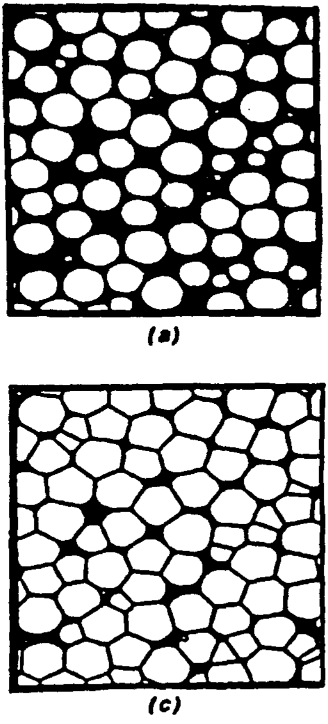

Figure 2: Computer simulation of the consolidation of spherical grains (from [19]), showing decreasing porosity φ, with (a) $\varphi = 0.364$, (b) $\varphi = 0.200$, (c) $\varphi = 0.100$ and (d) $\varphi = 0.030$

Provence, where most of this work was completed. S. M. Kozlov would have liked to thank G. Dagan for helpful discussions they had on porous media problems at Oberwolfach. L. Schwartz, M. Sahimi, VCH Publishers, and the Amercian Physical Society are kindly acknowledged for use of the figures that appear here. Finally, K. M. Golden gratefully acknowledges support from NSF Grants DMS-9307324, DMS-9622367, and OPP-9725038, and ONR Grant N000149310141.

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