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Percolation Models for Porous Media

Kenneth M. Golden[1]

Recent progress in understanding the effective transport properties of percolation models for porous and conducting random media is reviewed. Both lattice and continuum models are studied. First, we consider the random flow network in $\mathbb{Z}^N$, where the pipes of the network are open with probability p and closed with probability $1 - p$. Near the percolation threshold p_c, the effective permeability $\kappa^*(p)$ exhibits the scaling behavior $\kappa^*(p) \sim (p - p_c)^e$, $p \to p_c^+$, where e is the permeability critical exponent. In the limit of low Reynolds number flow, this model is equivalent to a corresponding random resistor network. Here we discuss recent results for the resistor network problem which yield the inequalities $1 \leq e \leq 2$, $N = 2, 3$ and $2 \leq e \leq 3$, $N \geq 4$, assuming a hierarchical node-link-blob (NLB) structure for the backbone near p_c. The upper bound $t = 2$ in $N = 3$ virtually coincides with a number of recent numerical estimates. Secondly, we consider problems of transport in porous and conducting media with broad distribution in the local properties, which are often encountered. Here we discuss 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 matrix-particle composites near critical volume fractions. Moreover, the results for the model rigorously establish the widely used Ambegaokar, Halperin, and Langer critical path analysis [AHL71].

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2.1 Fundamentals of Percolation Theory

Porous media represent an interesting class of materials which exhibit a wide range of microstructures. For a given microstructure, the central problem of single-phase flow through the medium is to find its effective permeability κ^* [Sah95, Dul92, Dag89, Mat67, BB93, Has59, Pra61, BM85, JKS86, AT91]. The microstructure can be characterized in many ways, the simplest being the pore volume fraction, or porosity ϕ. In general, however, the macroscopic transport properties of porous media depend in a complex way on the details of the pore structure, not simply its volume fraction. Due to the wide range of relevant microstructures, there have been many approaches to estimating the effective permeability κ^* of porous media. One widespread approach is to treat the porous medium as a network of open pores of varying size and "throats" of varying cross section which connect the pores [Kop82, Fat56, Sah95, Dul92, BB93]. In the limit of low Reynolds number flow, this "pipe" network becomes equivalent to a linear electrical network. In each pipe, the fluid flux is linearly related to the pressure drop across the pipe, which is a local version of Darcy's law. For electrical conduction, the current in Ohm's law is proportional to the potential drop across the resistor. In this case, the problem of finding the effective permeability κ^* of the network is mathematically equivalent to finding the effective conductivity σ^* of the corresponding resistor network.

The simplest network model which, nevertheless, exhibits complex macroscopic behavior is where the pipes form the standard bond lattice in $\mathbb{Z}^N$ and they are either open or closed with probabilities p or $1-p$, respectively. This network is based on the classical percolation model of Broadbent and Hammersly [BH57], one of the original inspirations for which was the flow of gas through the porous substance inside a miner's gas mask. In this model, we assign a local permeability, or fluid conductance $\kappa(x)$, to the bonds, where $\kappa(x) = 1$ or $\varepsilon \geq 0$ with probabilitities p or $1-p$. When $\varepsilon = 0$, the effective permeability $\kappa^*(p) = 0$ for $p \leq p_c$, yet $\kappa^*(p) > 0$ for $p > p_c$, where p_c is the percolation threshold, which equals $1/2$ for $N = 2$. Furthermore, it is generally believed (though not rigorously proven), that $\kappa^*(p)$ exhibits critical scaling behavior near the percolation threshold [FHS87, BB93, Sah95],

$$\kappa^*(p) \sim (p - p_c)^e, \qquad p \to p_c^+, \tag{1.1}$$

where e is the permeablity critical exponent. Due to the above equivalence between the fluid and electrical conduction problems, we can just as well consider the random resistor network [Kir71, SA92, GK84, Gri89, Kes82, CC86], where the electrical conductivity $\sigma(x)$ of the bonds is defined similarly to $\kappa(x)$ above. Then, the effective conductivity $\sigma^*(p)$ is believed to behave as

$$\sigma^*(p) \sim (p - p_c)^t, \qquad p \to p_c^+, \tag{1.2}$$

where t is the conductivity critical exponent. At the percolation threshold, the system undergoes a phase transition from an insulator to a conductor. Analogously, the fluid sytem undergoes a phase transition from a medium, through which no fluid can pass, to a porous or permeable medium. Furthermore, for such network

models, the critical exponents for permeability and electrical conductivity are the same: [FHS87, BB93]

$$e = t \tag{1.3}$$

(although this relation is not generally true for continuum systems [FHS87]). These network models have been used extensively to study problems in porous media [Sah95] and electrical transport [SA92].

In the first part of this chapter, we will review recent progress in understanding the behavior of $\kappa^*(p)$ or $\sigma^*(p)$ near the percolation threshold [Gol90, Gol92]. In particular, assuming a hierarchical, node-link-blob (NLB) structure [Sta77, Con82, BB93] for the conducting backbone of the percolation cluster near p_c (and certain technical assumptions), we have rigorously shown that the permeability exponent e or the conductivity exponent t satisfies the following inequalities:

$$\begin{aligned} 1 \le e \le 2, \qquad & N = 2, 3, \\ 2 \le e \le 3, \qquad & N \ge 4. \end{aligned} \tag{1.4}$$

Our approach was motivated by the simple observation that, in numerical simulations [Kir71, NvVE86, WL74], the graph of $\sigma^*(p)$ for bond or site models in $N \ge 2$ is always convex near p_c. We, then, analyze the asymptotic behavior of $\frac{d^2\sigma^*}{dp^2}$ near the percolation threshold p_c and investigate the consequences for the critical exponent t or e under the above mentioned assumptions. In particular, in the key assumption about backbone structure, our NLB model contains both singly and multiply connected bonds, has "loops" on arbitrarily many length scales in a self-similar fashion, and incorporates the few features rigorously known [Con82, CC86] about the backbone on a macroscopic scale.

Our results for $N = 3$ are particularly intriguing. First, the inequality $t \le 2$ excludes roughly one fourth of published numerical estimates of t for $N = 3$, which have ranged from 1.5 to 2.36. Furthermore, this inequality is based on an exact calculation of $t = 2$ for one particular member of our class of hierarchical NLB backbones, which provides an upper bound on t for the full class. In view of this result, it is quite striking that Gingold and Lobb [GL90] have obtained the estimate $t = 2.003 \pm 0.047$ for $N = 3$ from simulation on lattices up to $(80)^3$ and Adler et al. [AMA+90] have obtained $t = 2.02 \pm 0.05$ from a 13th order series expansion. Moreover, in [BB93] by Monte Carlo simulation, it is found that the permeability exponent e has a "universal" value of 2.0 under certain conditions of the distribution of local conductances. In addition, our inequality is compatible with the results of an $\varepsilon = 6 - N$ expansion [HKL84] and the general view that "roughly $t = 2$" [SA92]. The recent numerical results, in conjunction with our work, suggest the possibility that $t = e = 2$ is an *exact* result for $N = 3$. To our knowledge, the results in [Gol90, Gol92] are the only ones which relate t or e directly and naturally to the number 2, rather than to other (unknown) critical exponents of percolation theory.

Before discussing the second part of this chapter, we will mention other recent results in homogenization that may be of interest to researchers in porous media. In [Gol89] we began investigating the behavior of $\sigma^*(p)$ in the complex $p-$plane

and were able to establish rigorously that, for $\varepsilon > 0$, $\sigma^*(p)$ is analytic in p in some open neighborhood containing [0, 1]. Investigating how this analyticity is lost and how singular behavior develops at criticality, as $\varepsilon \to 0$ or in the infinite volume limit, has led to a number of works, including [Gol91] and [BG90]. The investigation culminated in [Gol95b], where this question has been addressed in detail, by finding a direct correspondence between the random resistor network (or continuum analogs) and the Ising model for a ferromagnet, where there is a well-developed Lee–Yang theory [YL52, LY52, Gri72] for understanding the onset of critical behavior. The correspondence is established by introducing a partition function and free energy for conduction problems, which have equivalent representations to those for the Ising ferromagnet. Through this correspondence, the percolation threshold p_c for transport in the random flow or resistor network is characterized as an accumulation point of zeros of the partition function in the complex p-plane.

On a more practical note, we also mention that the author has been involved in studying the electromagnetic properties of sea ice, which is a composite of pure ice containing random brine and air inclusions. It is very interesting to note that sea ice exhibits a percolation threshold at a critical temperature $T_c \approx -5$ °C, above which the brine inclusions coalesce and the sea ice becomes porous, allowing transport of sea water and brine through the ice. The implications of this transition on homogenized electrical transport coefficients is discussed in [Gol95a] and [Gol94a]. Furthermore, we report briefly in [LG] on observations we made of transport and percolation processes in sea ice and their effect on microwave backscatter from sea ice in the Weddell Sea, Antarctica during July and August of 1994.

In the second part of this chapter, we will continue to review recent results in homogenization for percolation models which impact on the study of porous media. The work we now describe began in discussions with Serguei Kozlov in Moscow in 1991 and slowly evolved into [GK]. In many problems of technological importance, one meets systems which display 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 [Sah95, Dul92, Fat56, Kop82, FHS87, Dag89, Mat67]. In associated network models, one must consider a broad distribution in the local fluid conductances in the bonds. As another example, in resistor network models of hoppping conduction in amorphous semiconductors, the bonds of the network are assigned a wide range of conductivities [AHL71, Sha77, Kir83]. A powerful idea which has been widely used [Sah95] to estimate the effective properties of such systems is the *critical path analysis*, first introduced by Ambegaokar, Halperin and Langer (AHL) [AHL71]. 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. The critical path anal-

ysis was further developed in the context of amorphous semiconductors in [Sha77] and [Kir83], and its accuracy was numerically confirmed for various conductance distributions in [BOJG86]. The AHL idea has also been used to model the permeability and electrical conductivity of porous media, such as sandstones, obtained through mercury injection [KT86, KT87, BJ87, Dou89], and fractured rocks with a broad distribution of fracture apertures [CGR87]. Recently, it has been applied to porous media saturated with a non-Newtonian fluid [Sah93].

Although the AHL idea has been used with substantial success, there has been little analysis of their fundamental observation that the critical conductance g_c dominates the effective behavior. In [GK], 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 complex random topology, similar to a Voronoi network (see Fig. 2.2) [Sah95, Ker83, RS85, JHSD84, FHS87].

We obtain our model as a "long-range" generalization of the random checkerboard in $\mathbb{R}^2$ [Dyk71, SK82, Kel87, Mol91, Koz89, BG94, Gol94b], where the squares are assigned conductivities 1 with probability p and $\varepsilon > 0$ 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 polymeric (insulating) matrix. For example, the presence of both corner and edge connections between squares produces two percolation thresholds with distinct asymptotic behavior of the effective conductivity as $\varepsilon \to 0$ (or $\varepsilon \to \infty$) in the different regimes of p separated by these thresholds [SK82, Mol91, Koz89, BG94]. 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 [Koz89, BG94, Gol94b]. A key component 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 will 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" [Gol94b] 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 [MBN90] 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" con-

ductivities. 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.

2.2 Exponent Inequalities for Random Flow and Resistor Networks

The principal results of our investigation and the assumptions under which they are obtained are as follows. We will formulate the problem for the effective conductivity $\sigma^*(p)$, but, of course, everything carries over to the effective permeabitlity $\kappa^*(p)$. This connection will be examined in more detail below. First, the most serious assumption is that the conducting backbone near p_c has a hierarchical, self-similar node-link-blob (NLB) structure, as described in the Introduction. We further make some technical assumptions about $\sigma^*(p)$: it obeys the above scaling law (1.2) near p_c, has at least three derivatives for all $p > p_c$, and $\frac{d^2\sigma^*}{dp^2} + \frac{d\sigma^*}{dp} > 0$ at $p = 1$, which we have verified numerically. Under these assumptions, we prove exact asymptotics for $\frac{d^2\sigma^*}{dp^2}$ as $p \to p_c^+$. The proof employs a novel technique whereby $\frac{d^2\sigma^*}{dp^2}$, for the NLB model with $\varepsilon = 0$ and p near p_c, is computed using perturbation theory for $\sigma^*(p)$ (for two-and three-component resistor lattices) around $p = 1$, with a sequence of ε's converging to 1 as one goes deeper in the hierarchy. Our asymptotics yield not only convexity near p_c, which implies $t \geq 1$, but delineate in which dimensions $\frac{d^2\sigma^*}{dp^2} \to 0, +\infty$, or a positive constant as $p \to p_c^+$. Combining this information with the scaling law $\frac{d^2\sigma^*}{dp^2} \sim (p - p_c)^{t-2}$ yields the inequalities $1 \leq t \leq 2$ for $N = 2, 3$ and $2 \leq t \leq 3$ for $N \geq 4$. The inequality $t \leq 3$ for $N \geq 4$ is obtained by applying a similar analysis to $\frac{d^3\sigma^*}{dp^3}$ for the simpler node-link model, and can be viewed as a mean field bound, because it is believed that $t = 3$ for $N \geq 6$. We stress that the convexity and inequalities are not rigorous for the actual backbone near p_c for the original lattice, but *are* rigorous for the NLB model of the backbone, under the above technical assumptions.

Before we begin, we refer the reader to [Gol92]. In addition to containing the mathematical details of the results discussed here, we obtain, there, numerical and rigorous results concerning the regimes in ε and p of convexity of $\sigma^*(p)$ for bond and site models, the principal rigorous results being that, for the $N = 2$ bond problem, although $\sigma^*(p)$ cannot be convex for all p, when $\varepsilon = 0$, it is convex for every $\varepsilon > 0$ near $p_c = 1/2$.

We now formulate the bond conductivity problem for $\mathbb{Z}^N$, where, for simplicity, we begin with $N = 2$. Take an $L \times L$ sample G_L of the bond lattice with $M(\sim NL^N)$ bonds. Assigned to G_L are M independent random variables c_i, $1 \leq i \leq M$, the bond conductivities, which take the values 1 with probability p and $\varepsilon \geq 0$ with probability $1-p$. We attach perfectly conducting bus bars to two opposite edges and

let $\sigma_L(p)$ be the effective conductance of this network, averaged over realizations of the bond conductivities. This conductance is just the total current that flows through the network when there is a unit average potential difference between the bus bars. The potential at any lattice site in $\mathbb{Z}^N$ is determined by Kirchoff's laws. For $N \geq 1$, the bulk conductivity of the lattice is defined as

$$\sigma^*(p) = \lim_{L\to\infty} L^{2-N}\sigma_L(p). \tag{2.1}$$

For $\varepsilon > 0$, the infinite volume limit in (2.1) has been shown to exist [Koz78, PV82, GP83, Kün83], and for $\varepsilon = 0$ the existence of σ^* has been proven in the continuum [Zhi89].

At this point, it is useful to investigate the relationship between the conductivity and flow problems, particularly, in view of the fact [CC86] that, for effective permeability, we must replace (2.1) with

$$\kappa^*(p) = \lim_{L\to\infty} L^{1-N}\kappa_L(p), \tag{2.2}$$

where $\kappa_L(p)$ replaces $\sigma_L(p)$, and is the total fluid current in the network, which we now discuss. To elucidate the connection, we consider the following simple model [Dul92] in $N = 3$ of a cube of side L filled with n parallel cylindrical capillary tubes of length L and radius R, evenly spaced throughout the cube. Then in the Darcy regime, the total flow rate is given by

$$Q = n\,\frac{\pi R^4}{8\eta}\left(\frac{\Delta P}{L}\right), \tag{2.3}$$

where η is the fluid viscosity and ΔP is the pressure drop across the cube (or across each capilllary). If we identify $k = \frac{\pi R^4}{8}$ as the permeability of each capillary, then, because $n = L^2$, we can rewrite (2.3) as

$$\frac{Q}{L^2} = \frac{k}{\eta}\left(\frac{\Delta P}{L}\right), \tag{2.4}$$

where the left-hand side is the fluid current density. Now, we can associate this problem with the effective conductance of n parallel "networks" of L conductances in series, each with conductance

$$g = \frac{k}{\eta L}, \tag{2.5}$$

which is just the effective conductance of L bonds in series, each of conductance $\frac{k}{\eta}$. This relation accounts for the different scalings in (2.1) and (2.2).

Now, the calculation of $\frac{d^2\sigma^*}{dp^2}$ requires the following definition. For any graph B with bonds b_i of unit conductivity, define

$$\delta^2\sigma(B) = \sum_{b_i\neq b_j}\Big[\sigma_{ij}(1,1) + \sigma_{ij}(0,0) - \sigma_{ij}(1,0) - \sigma_{ij}(0,1)\Big], \tag{2.6}$$

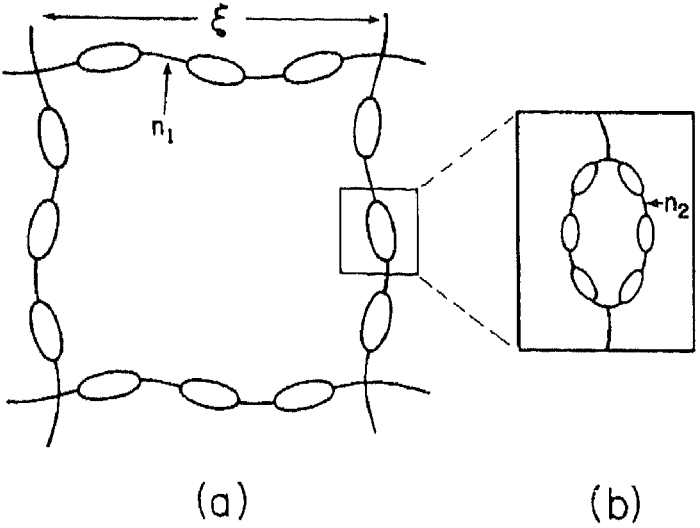

FIGURE 2.1. Node–link–blob model of the conducting backbone near p_c. In (a), the nodes are a correlation length ξ apart and are connected by necklaces of beads (blobs) and strings (links) with n_1 bonds connecting two beads. The beads have a self-similar structure, as shown in (b), with n_2 bonds connecting two beads.

where, in (2.6), $\sigma_{ij}(1, 1) = \sigma(B)$, the conductivity of B measured between the two bus bars, $\sigma_{ij}(0, 0)$ is the conductivity of B with b_i and b_j removed, and so on. This expression represents the discrete second derivative of σ with respect to p, as follows. Let G be the lattice in $N \geq 2$ with bond conductivities 1 and 0 and bulk conductivity function $\sigma^*(p)$. If $B = B(p)$ is a realization of occupied bonds of G at probability p, then [Gol92]

$$p^2 \frac{d^2\sigma^*}{dp^2} = \delta^2\sigma^*(B(p)), \tag{2.7}$$

where $\delta^2\sigma^*$ is the scaled infinite volume limit of (2.6), and the right-hand side in (2.7) is appropriately averaged. (We are assuming here that $\sigma^*(p)$ is twice differentiable for $p > p_c$ when $\varepsilon = 0$.) In (2.6), note that dangling bonds do not contribute, so that one may think of $B(p)$ as a realization of the backbone at bond fraction p. For clarity, note that, at $p = 1$, $B(p) = G$. We remark that analysis of simple graphs, typically, shows that positive contributions to (2.6) arise from series pairs, whereas negative contributions arise from pairs in parallel.

The idea now is to replace an actual backbone graph $B(p)$ for p near p_c by a node-link-blob (NLB) graph A, which is based on the work of Stanley [Sta77] and Coniglio [Con82]. This graph is a "super-lattice," constructed by replacing the bonds of the hypercubic lattice G in $N \geq 2$ by first-order necklaces composed

of strings (links) and first-order beads (blobs), and separating the nodes of G by a correlation length ξ, as in Fig. 2.1a. The beads themselves have a hierarchical structure, as shown in Fig. 2.1b, consisting of two second-order necklaces in parallel, and so on, in a self-similar fashion to order J, for an arbitrary, large integer J. We assume that any kth order necklace has $\beta - 1$ beads on it, for an arbitrary, large integer β, and that each pair of beads is joined by a string of n_k bonds, so that there are a total of βn_k string bonds on each necklace. The βn_1 string bonds on any first-order necklace are called singly connected – because removal of one of them breaks the connection between nodes separated by ξ. All the rest of the bonds in the NLB graph are multiply connected, and among these it is useful to identify the βn_2 string bonds on a 2^{nN}-order necklace as doubly connected, because it is possible to remove two of them (in parallel) and break a connection between nodes. Based on a result of Coniglio's [Con82] implying, in our context, that the number of singly and doubly connected bonds between the nodes both diverge with exponent 1 as $p \to p_c^+$, we assume that $n_1 = 2\beta n_2$. Due to self-similarity, we assume that

$$n_{j-1} = 2\beta n_j \quad , \quad j = 2, \ldots, J. \tag{2.8}$$

Eq. (2.8) can be used to solve for the n_j, $j > 1$, in terms of n_1, with $n_2 = n_1/2\beta$, $n_3 = n_1/4\beta^2$, and so on, and we refer to the NLB graph as $A(n_1)$. In this model, the percolation limit $p \to p_c^+$ is characterized by the limits $n_1, \beta, J, \xi \to \infty$, so that the lengths of all orders of necklaces and the numbers and sizes of all orders of blobs, diverge as $p \to p_c^+$.

Before we give the asymptotics of $\delta^2\sigma^*(A(n_1))$, we must discuss the conditions under which they are proven. Consider $\sigma^*(q_1, q_2)$ for the bond lattice in $\mathbb{Z}^N$ with three conductivities $1, \varepsilon_1$, and ε_2 in proportions p, q_1, and q_2, in addition to our standard two-component conductivity $\sigma^*(p)$. We require that $\sigma^*(q_1, q_2)$ has second-order partials at $q_1 = q_2 = 1$ for all $\varepsilon_1, \varepsilon_2 \geq 0$, and that $\sigma^*(p)$ has two derivatives at $p = 1$, for all $\varepsilon \geq 0$. For ε, ε_1, and $\varepsilon_2 > 0$, these conditions are satisfied by our general results [Gol92] that $\sigma^*(p)$ is analytic for all $p \in [0, 1]$ and $\sigma^*(q_1, q_2)$ is analytic for all $(q_1, q_2) \in [0, 1] \times [0, 1]$. The $\varepsilon = \varepsilon_1 = 0$ requirements will be assumed, although Kozlov [Koz89] has proven the existence of $\frac{d\sigma^*}{dp}\Big|_{p=1}$ for a class of continuum analogs.

The second main condition is that, given the hypercubic base lattice G for $A(n_1)$,

$$K(G) = \frac{d\sigma^*}{dp}\Big|_{p=1} + \frac{d^2\sigma^*}{dp^2}\Big|_{p=1} > 0. \tag{2.9}$$

In any $N \geq 2$, $\frac{d\sigma^*}{dp}\Big|_{p=1} = N/(N-1)$ [Kir71], whereas $\frac{d^2\sigma^*}{dp^2}\Big|_{p=1}$, if negative, is quite small, e.g., ≈ -0.21 in $N = 2$ [NvVE86, Gol92], indicating that $\sigma^*(p)$ is quite straight near $p = 1$, so that (2.9) is satisfied. Condition (2.9) amounts to a consequence of the long-held view that effective medium theory (giving a straight-line solution) provides an accurate description of $\sigma^*(p)$ near $p = 1$, which also holds for general lattices. In fact, the asymptotics below can be proven for a variety of periodic base lattices G which satisfy (2.9) and, presumably, hold even for random lattices.

We may now state our principal result. Under the above assumptions, for fixed, large n_1, β, and J,

$$\delta^2\sigma(A(n_1)) = \alpha_J K(G)\beta n_1 + \sum_{i=0}^{\infty} \frac{a_i n_1 + b_i}{\beta^i}, \tag{2.10}$$

where $(\alpha_J)^{-1} = \sum_{i=0}^{J}(\frac{1}{4})^i$ and the series in (2.10) converges, so that

$$\delta^2\sigma^*(A(n_1)) \sim \frac{\alpha_J K(G)\beta n_1}{\xi^{N-2}} > 0, \quad n_1, \beta, J, \xi \to \infty. \tag{2.11}$$

The idea of the proof of (2.10) is first to write

$$\delta^2\sigma(A(n_1)) = \sum_{k \geq j} \delta_{jk}, \tag{2.12}$$

where δ_{jk} is the sum of all contributions to $\delta^2\sigma(A(n_1))$ in (2.6) arising from pairs with one bond in a jth order string and the other in a kth order string, which is in either the same or a different first-order necklace. Now let z_k be the conductivity of a single first-order necklace with one bond removed from a kth order string, with $z_0 = \alpha_J/\beta n_1$ for no bond removed, $z_1 = 0$, and

$$z_k = z_0\left(1 + \frac{\gamma_k}{\beta^{k-1}}\right)^{-1}, \quad k \geq 2, \tag{2.13}$$

where $\gamma_k \to 0$ as $k \to \infty$ geometrically fast. There are analogous formulas for the various forms of z_{jk} with two bonds removed, say, in series or in parallel. Then, through representations like (2.6) and (2.7), we obtain formulas for the δ_{jk} in terms of derivatives of $\sigma^*(p)$ and $\sigma^*(q_1, q_2)$ at $p = 1$, such as

$$\delta_{11} = z_0\left[\beta n_1(\beta n_1 - 1)\frac{d\sigma^*}{dp}(p = 1, h_1) + (\beta n_1)^2\frac{d^2\sigma^*}{dp^2}(p = 1, h_1)\right] \tag{2.14}$$

and

$$\delta_{12} = z_0\left[(\beta n_1)^2\frac{d\sigma^*}{dp}(p = 1, h_1) + (\beta n_1)^2\frac{\partial^2\sigma^*}{\partial q_1 \partial q_2}(p = 1, h_1, h_2)\right], \tag{2.15}$$

where $\frac{d\sigma^*}{dp}(p = 1, h_1)$, e.g., is for G with bond conductivities 1 and $h_1 = 0$, with $h_k = z_k/z_0$. As $k \to \infty$, $h_k \to 1$, and, as $\beta \to \infty$, $h_k \to 1$ for all $k \geq 2$, and similarly for $h_{jk} = z_{jk}/z_0$. The necessary control of the δ_{jk} is, then, obtained either from (2.9) or from perturbation theory around a homogeneous medium ($\varepsilon = 1$ or $\varepsilon_1 = \varepsilon_2 = 1$), which establishes (2.10).

We wish to make the following remarks concerning the above result. First, a result similar to (2.11) holds if we replace (2.8) by $n_{j-1} = \eta_j\beta_j n_j$, where the blobs of order $j-1$ are made of η_j necklaces in parallel, with reasonable assumptions about η_j and β_j. Even if the blobs have a more complicated "super-lattice" structure

themselves, an analog of (2.11), presumably, holds. Also, as noted above, (2.11) can be proven for a variety of base lattices G. Finally, although the principal assumption of the NLB graph replacing the actual backbone is quite serious, our proof of (2.10) shows that the dominant contribution to (2.11) comes from δ_{11}, which comes from macroscopic contributions in the NLB graph, where the model reflects the actual structure well. A similar result will hold for any reasonable assumption about microscopic backbone structure.

Now, we proceed to the implications of (2.11). First, its positivity establishes convexity of $\sigma^*(p)$ for the NLB model, which implies (under our assumptions, including scaling and the existence of three derivatives of $\sigma^*(p)$ for all $p > p_c$ when $\varepsilon = 0$) that $t \geq 1$, for any $N \geq 2$. Now, let $\lambda(n_1)$ be the length of a first-order necklace, so that $\lambda(n_1) \approx \beta n_1 + \beta^2 n_2 + \ldots + \beta^J n_J = \theta_J \beta n_1$, $\theta_J = \sum_{i=0}^{J} 2^{-i}$. By (2.11), then

$$\delta^2\sigma^*(A(n_1)) \sim \frac{\rho_J \lambda(n_1)}{\xi^{N-2}} \quad , \quad n_1, \beta, J, \xi \to \infty, \tag{2.16}$$

where $\rho_J = \alpha_J K(G)/\theta_J$, so that $\rho_J \approx 2/3$ for large J in $N = 2$. Because all the parameters n_1, β, J and ξ are diverging as $p \to p_c^+$, we can define a whole class of NLB models by how fast $\lambda(n_1)$ scales to ∞ relative to ξ. By the structure of the model, clearly, $\lambda(n_1) \geq \xi$, and, typically, $\lambda/\xi \to \infty$. Thus, as a consequence of (2.16), in $N = 2$ and 3,

$$\delta^2\sigma^*(A(n_1)) \to +\infty \quad , \quad n_1, \beta, J, \xi \to \infty, \tag{2.17}$$

except in $N = 3$ when $\lambda(n_1) = C\xi$, $C \geq 1$, in which case,

$$\delta^2\sigma^*(A(n_1)) \to \rho C > 0, \tag{2.18}$$

where $\rho = \lim_{J\to\infty} \rho_J$. In $N \geq 4$, if λ and ξ are scaled so that $\lambda(n_1)/\xi^{N-2} \to 0^+$, then,

$$\delta^2\sigma^*(A(n_1)) \to 0^+. \tag{2.19}$$

Under our assumptions, in particular, that $\frac{d^2\sigma^*}{dp^2} \sim (p - p_c)^{t-2}$, then, collecting our results,

$$\begin{aligned} &1 \leq t \leq 2, \quad N = 2, 3 \\ &2 \leq t \leq 3, \quad N \geq 4. \end{aligned} \tag{2.20}$$

In (2.20), the last inequality $t \leq 3$ for $N \geq 4$ is obtained by a result that $\delta^3\sigma^*(A'(n_1)) \sim C'\lambda^2(n_1)/\xi^{N-2}$ for a simpler node-link graph $A'(n_1)$, which is believed to be adequate in higher dimensions [Har83]. For models in $N = 4, 5$ which satisfy $\lambda^2(n_1)/\xi^{N-2} \to \infty$,

$$\delta^3\sigma^*(A(n_1)) \to \infty,$$

so that $\frac{d^3\sigma^*}{dp^3} \sim (p - p_c)^{t-3} \to \infty$, which gives the inequality.

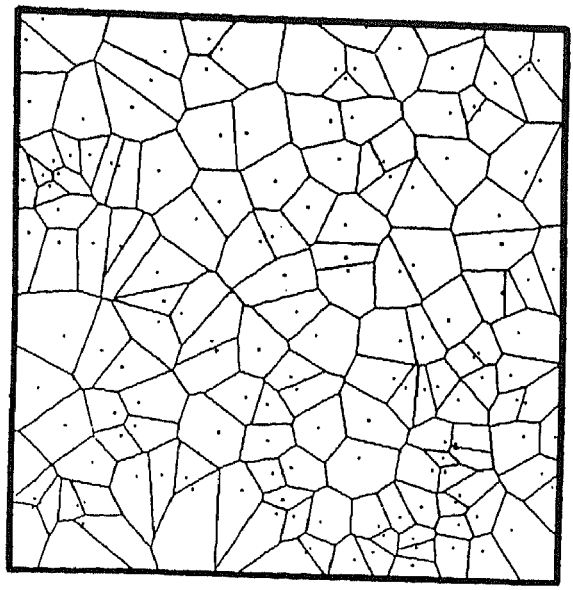

FIGURE 2.2. Two-dimensional Voronoi tessellation. The boundaries of the polygonal grains are formed from points which are equidistant from the dots in two neighboring grains. These boundaries form the channels of easiest flow in consolidated granular media and fractured rocks.

2.3 Critical Path Analysis in Highly Disordered Porous and Conducting Media

We now present a continuum percolation model which closely represents fluid flow in some fractured rocks and consolidated granular media, as well as electrical conduction in some matrix-particle composites [GK]. For simplicity, we first give the formulation for electrical conductivity in $N = 2$, but the model and result can be carried over to $N = 3$ and to fluid flow in a porous medium obeying Darcy's law. Consider the checkerboard of unit white squares in $\mathbb{R}^2$, with centers of the squares being the points of the lattice $\mathbb{Z}^2$. Randomly color the squares red with probability p, where the probability of coloring one square is independent from any other. Then for $x \in \mathbb{R}^2$, 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

$$\sigma(x) = e^{\lambda S(x)}. \tag{3.1}$$

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 (3.1) 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

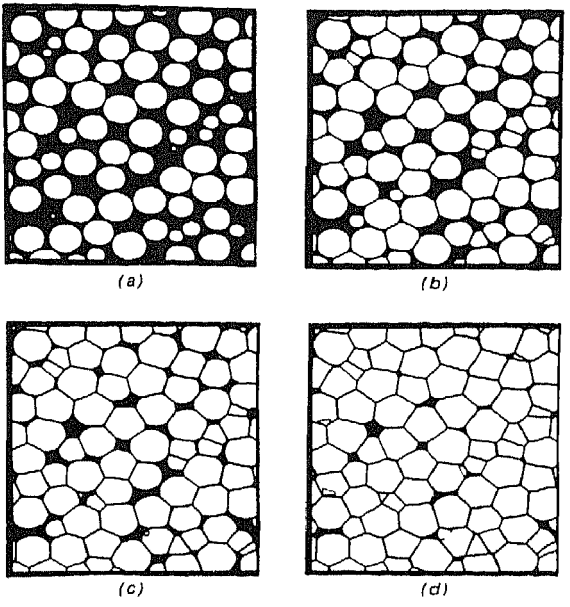

FIGURE 2.3. Computer simulation of the consolidation of spherical grains, showing decreasing porosity ϕ, with (a) $\phi = 0.364$, (b) $\phi = 0.200$, (c) $\phi = 0.100$, and (d) $\phi = 0.030$.

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. 2.2 (from [Sah95]). 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 (3.1), which is defined as follows (e.g., [Koz78, GP83]). 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 $\langle E(x) \rangle = e_k$, where e_k is a unit vector in the kth direction, and $\langle \cdot \rangle$ denotes ensemble or infinite volume average. Then, the effective conductivity σ^* is defined via

$$\langle J \rangle = \sigma^* \langle E \rangle. \tag{3.2}$$

For fluid flow (with unit viscosity) in porous media [Sah95] obeying Darcy's law, $v = -\kappa(x)\nabla P$, where $\kappa(x)$ is the local permeability corresponding to (3.1), v is the fluid velocity satisfying $\nabla \cdot v = 0$, and P is the pressure (including gravity), one is interested in the effective permeability κ^*, defined analogously to (3.2),

$$\langle v \rangle = -\kappa^* \langle \nabla P \rangle \tag{3.3}$$

As briefly mentioned earlier, if $\kappa(x)$ has the form (3.1), 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.

The degree of penetration into the grains decreases as λ increases. Fig. 2.3 (from [RS85]) shows a computer simulation of a grain consolidation process [RS85]. 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 [RS85].

To state the results for the asymptotics and to more fully describe our model, we first consider standard, nearest neighbor-site percolation on $\mathbb{Z}^2$ [SA92, Kes82, Gri89], 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 $\mathbb{Z}^2$, 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, so 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, $j \leq 1$, 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 $(m, n) \in \mathbb{Z}^2$ 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 (because the distance between connected squares is 0 in both cases), so that $p_c(1) = 1 - p_c^s \simeq 1 - 0.59 = 0.41$ [Koz89, BG94, Gol94b]. Furthermore $p_c(j+1) < p_c(j)$, which can be obtained from [AG91]. From analysis of a Poisson distribution of discs in the plane (see below), one can also find that

$$p_c(j) \sim \frac{\pi \mu_c}{j}, \qquad j \to \infty, \tag{3.4}$$

where μ_c is the critical percolation intensity for unit discs [Gri89].

The last ingredient needed 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 $R_j = \{x \in \mathbb{R}^2 : S(x) \leq S_c(j)\}$ percolates in $\mathbb{R}^2$, where, for j associated with $(m, n) \in \mathbb{Z}^2$ as above,

$$S_c(j) = r_j/2 = \sqrt{(1 - \delta_{m0})(m-1)^2 + (n-1)^2}\Big/2, \tag{3.5}$$

where δ_{m0} is the Kroneker delta. By percolating in $\mathbb{R}^2$, 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

$$S_c(p) = S_c(j), \quad p_c(j) < p < p_c(j-1), \quad j \geq 1, \tag{3.6}$$

where $p_c(0) = 1$. These critical distances $S_c(j)$ in our model correspond, via (3.1), 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 $\sigma(x) = e^{\lambda S(x)}$, for $p \neq p_c(j)$,

$$\frac{1}{\lambda} \log \sigma^*(p) \sim S_c(p), \qquad \lambda \to \infty, \tag{3.7}$$

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

$$S_c(p) \sim \sqrt{\frac{\mu_c}{p}}, \qquad p \to 0, \tag{3.8}$$

where p is the density of red squares (i.e., with units of inverse square length). Via (3.7) and (3.8), 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 set $\lambda = 1$, so that $\sigma(x) = e^{S(x)}$ and consider the asymptotics of $\sigma^*(p)$ as $p \to 0$, we also find that

$$\sqrt{p} \log \sigma^*(p) \sim \sqrt{\mu_c}, \qquad p \to 0. \tag{3.9}$$

Now, we give the analysis which leads to these results. The idea is to exploit the variational definition of σ^* equivalent to (3.2) and its dual to obtain upper and lower bounds on σ^*. Let $\Lambda_L = [0, L] \times [0, L] \subset \mathbb{R}^2$. Then the variational form of (3.2) is given by

$$\sigma^* = \lim_{L \to \infty} \frac{1}{L^2} \inf_{u \in \mathcal{P}} \int_{\Lambda_L} \sigma(x) \, |\nabla u|^2 dx, \tag{3.10}$$

where $\mathcal{P} = \{\text{continuous potentials } u \text{ on } \Lambda_L : u(0, x_2) = 0, \ u(L, x_2) = L, \forall x_2 \in [0, L]\}$. We obtain bounds by inserting trial u in (3.10). To describe the construction, we recall certain properties of standard-site percolation. It has been shown [GK84, CC86] that, for $p > p_c^s$, the number α_L of disjoint crossings of Λ_L by nearest neighbor sites satisfies (roughly speaking) $\alpha_L = O(L)$ as $L \to \infty$. In our generalized model, recall that, for $p > p_c(j)$, $R_j = \{x \in \mathbb{R}^2 : S(x) \leq S_c(j)\}$ percolates in $\mathbb{R}^2$. In this case, the number $\alpha_L(j)$ of disjoint crossings of Λ_L by j-connected red squares also satisfies $\alpha_L(j) = O(L)$ as $L \to \infty$. We call the associated disjoint subsets of R_j that cross Λ_L pink "j-chains" (we say "pink" because such sets contain both red squares and parts of white ones). For our purposes, it will be necessary to consider only those chains which cross vertically. Now the trial u is constructed roughly as follows. In the regions between the j-chains, u is flat, so that $\nabla u = 0$. However, on the j-chains, u increases linearly across the chain, so that the total contribution to (3.10) of $|\nabla u|^2$ on the pink j-chains is $O(L^2)$. (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 [Koz89] for the so-called Laplace–Dirichlet integral.) For such u, then, the integrand in (3.10) is zero off the j-chains, and, for x in the pink j-chains,

$$\sigma(x) \leq e^{\lambda S_c(j)}, \tag{3.11}$$

which leads to the inequality,

$$\sigma^*(p) \leq C_1 e^{\lambda S_c(j)}, \qquad p > p_c(j), \tag{3.12}$$

for some $C_1 > 0$ (depending on p), with $j \geq 1$.

To get the lower bound, we first note that the dual to (3.10) is 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 $p_c(0) = 1$), $W_j = \{x \in \mathbb{R}^2 : S(x) \geq S_c(j)\}$ percolates in $\mathbb{R}^2$, which can be seen as follows. When $p < p_c(j-1)$, R_{j-1} cannot percolate, $j \geq 2$. In this case, easy geometrical reasoning shows that infinite chains of white cells must exist, so 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, $Wj = \{x \in \mathbb{R}^2 : S(x) \geq r_j/2 = S_c(j)\}$, which contains this set of white chains, percolates in $\mathbb{R}^2$. Now, constructing u similar to that above, one obtains

$$[\sigma^*(p)]^{-1} \leq C_2 e^{-\lambda S_c(j)}, \qquad p < p_c(j-1), \tag{3.13}$$

for some $C_2 > 0$ (depending on p) , with $j \geq 1$. Combining (3.12) and (3.13) yields (3.7).

To obtain the asymptotic behavior of the thresholds in (3.4) and the exponents in (3.8), we connect our work to the analogous problem for a Poisson distribution of discs in $\mathbb{R}^2$. 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 (3.1), $S_\mu(x) = \text{dist}\{x, \text{ nearest } x_k\}$ and $\sigma_\mu(x) = e^{\lambda S_\mu(x)}$. Let S_μ^c be the smallest h for which $\{x \in \mathbb{R}^2 : S_\mu(x) \leq h\}$ percolates. Then, S_μ^c coincides with r_μ^c, the minimum radius so that the discs of radius r_μ^c, centered at the x_k, percolate. For the effective conductivity σ_μ^* in this case, the above arguments used for the long-range checkerboard yield

$$\frac{1}{\lambda} \log \sigma_\mu^* \sim S_\mu^c = r_\mu^c, \qquad \lambda \to \infty. \tag{3.14}$$

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 (3.14).

It is 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 $h\mathbb{Z}^2$, $h > 0$, 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, $\hat{S}_c(p) \to S_\mu^c$ as $p \to 0$. Then, with $\hat{S}_c = hS_c$ and $h = \sqrt{p/m}$, setting $\mu = 1$ yields $\sqrt{p} S_c(p) \to r_1^c$ as $p \to 0$, which is equivalent to (3.8), because $\mu_c = (r_1^c)^2$.

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

$$\mu_c = \lim_{j\to\infty} p_c(j) r_j^2 \,. \tag{3.15}$$

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

$$j \sim \pi r_j^2 \,, \quad j \to \infty \,, \tag{3.16}$$

which, combined with (3.15), yields (3.4).

In closing, we wish to make a few remarks. Presumably, an effective medium approach as in [SK82] could account for 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 calulations 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 (3.12) diverges like $\xi(p)$, as $p \to p_c(j)^+$, where $\xi(p)$ is the correlation length for $j-$percolation.

Ulrich Hornung

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