LETTER TO THE EDITOR

Percolation in multi-layered structures

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Abstract. We study a model for layered porous media. The model is a d-dimensional lattice percolation system with different concentrations given to different layers. We find for the 2D site percolation system, a critical line on which the percolation clusters are anisotropic and characterized by two different correlation length exponents. Our results suggest that due to the introduction of long-range correlations one obtains a new universality class different from isotropic percolation and from directed percolation.

The percolation model has been studied very extensively in recent years [1–4]. In particular, percolation has proved to be a useful model for many realizations of discovered systems. Most of the studies have focused on isotropic percolation systems. However, in general, *disordered systems in nature are anisotropic, such as layered porous rocks* [5]. In this paper we suggest a percolation model for anisotropic multi-layered porous media in which different layers may have different physical properties such as density and porosity. For that model we suggest a general anisotropic scaling theory to describe its critical behaviour. Numerical data from two different simulations support the predictions of the scaling theory.

In order to model a multi-layered percolation system, we assume a d-dimensional lattice constructed of a sequence of many sublattices, each with $d-1$ dimensions along a predefined axis. We shall call such a sublattice, a layer, and choose the predefined axis as the y axis. Each layer is a simple $(d-1)$-dimensional percolation with a unique concentration. This corresponds to the fact that multi-layered systems in nature may have different physical parameters at different layers. For simplicity, we study here the case of $d = 2$, and only two concentration values, p_1 and p_2, are assumed. Let $p(y)$ be the concentration of a layer in the y coordinate, and let $p(y)$ be independent random variables distributed according to: $\Pr(p(y) = p_1) = \Pr(p(y) = p_2) = \frac{1}{2}$. The parameters p_1 and p_2 define a point in a phase space. In this phase space the line $p_1 = p_2$ corresponds to the set of regular isotropic percolations, while the other points describe systems with anisotropic clusters. It is important to note that although the choice of the distribution of $p(y)$ is arbitrary, it is important to have a *random* distribution of $p(y)$ and not a periodic one. If, for example, we choose $p(y) = p_1$ if y is even and $p(y) = p_2$ if y is odd, then one obtains a model with no singular anisotropy and Δ is just a marginal perturbation. We shall call that model the 'alternating' model compared with our 'random' model. The random layer model has also been presented by Obukhov [6].

We find it more convenient to define $\bar{p}$ and Δ by $p_1 = \bar{p} - \Delta$ and $p_2 = \bar{p} + \Delta$ rather than p_1 and p_2. Thus, as we increase Δ from 0, we find that anisotropic clusters become

extended along the x axis and narrowed along the y axis. (This behaviour is common both to the 'alternating' and the 'random' models, but we shall see that only in the latter are the critical exponents affected by the anisotropy.) The parameter Δ may be understood as a measure for the amount of anisotropy in the system. The anisotropic clusters in the 'random' systems are found to be characterized by two different correlation lengths ξ_x and ξ_y, with two different exponents $\nu_x \neq \nu_y$, resulting in self-affine cluster structures [7].

It is well known that for $\Delta = 0$ there is a phase transition along the $\bar{p}$ axis [1]. Exactly at $\bar{p} = p_c$ an infinite cluster appears as $\bar{p}$ increases. We therefore assume that for every value of Δ there exists a unique critical value of $\bar{p}$, $\bar{p}_c(\Delta)$. This assumption is supported numerically and $\bar{p}_c(\Delta)$ is found. It is also well known that all the critical behaviour of an isotropic percolation system can be described by two exponents such as ν and β. All other exponents such as τ, γ, are related to β and ν. In the following we have generalized the relations between these exponents for anisotropic systems.

In the following we present a scaling approach for a general anisotropic d-dimensional percolation system. We assume that every axis has its own unique correlation exponent, i.e. $\xi_i \sim |p - p_c|^{-\nu_i}$. Therefore, the typical 'volume' of a cluster in such a percolation can be expressed as

$$V = \prod_{i=1}^{d} \xi_i = |p - p_c|^{-\sum \nu_i}.$$

Similarly, we define $d_f^{(i)}$ as the exponent characterizing the typical 'mass' of a cluster $s(\bar{\xi}) = \xi_i^{d_f^{(i)}}$ for $1 \le i \le d$. Generalizing from the isotropic case, we assume that the probability of an arbitrary site to belong to a cluster with linear sizes: $\xi_1, \xi_2, \ldots, \xi_d$ is:

$$Q(\bar{\xi}, p) \sim s(\bar{\xi})^{1-\tau} F(-|p - p_c|^{1/\sigma} \cdot s(\bar{\xi})). \tag{1}$$

Then, using scaling arguments (such as conservation of $V \cdot Q$ while changing p to p'), we get the following rule:

$$\nu_i d_f^{(i)} \sigma = 1. \tag{2a}$$

This corresponds to the well known isotropic law: $\nu d_f \sigma = 1$

$$\tau = 1 + \sum_{i=1}^{d} \frac{1}{d_f^{(i)}}. \tag{2b}$$

In the isotropic case we have $\tau = 1 + d/d_f$

$$\beta\sigma = \tau - 2. \tag{2c}$$

This law comes from the definition of $P_\infty \sim |p - p_c|^\beta$. In the isotropic case this law has the common form: $d_f = d - \beta/\nu$. Note that the equivalent formula for the anisotropic case for $d = 2$ is $d_f^x = (1 + \nu_y/\nu_x) - \beta/\nu_x$. Note also that a similar equation to (2b) was found for directed percolation [8].

In this paper we present numerical data to test if the two-dimensional 'random' model behaves according to the general rules resulting from these scaling arguments. The numerical simulations are performed using two methods: the 'gradient percolation' method and the 'cluster perimeter' method. Before describing the methods we define the term 'perimeter' which is necessary to the understanding of both methods, and find its critical behaviour using similar scaling arguments.

The perimeter of a cluster is the continuous path of occupied sites on its boundary. The perimeter can be either internal or external to the cluster. It was shown by Sapoval

et al [9] that such perimeters are useful to study percolation systems. They found that the fractal dimension of the perimeter of an isotropic percolation d_H is related to the correlation length exponent ν by $d_H = 1 + 1/\nu$. It is easy to see that the relations (2) are valid not only for the mass of the clusters but also for their perimeters (replacing d_f by d_H, etc). Thus, in the following we shall use the exponents β_H, τ_H, σ_H and $d_H^{(i)}$, to denote exponents characterizing the perimeter, and the analogue for (2) is

$$\nu_i d_H^{(i)} \sigma_H = 1 \tag{3a}$$

$$\tau_H = 1 + \sum_{i=1}^{d} \frac{1}{d_H^{(i)}} \tag{3b}$$

$$\beta_H \sigma_H = \tau_H - 2. \tag{3c}$$

Note that the exponent ν is the same for the perimeter model as for the mass model. Thus percolation properties can be studied directly from the perimeter. This fact is very important for simulations purposes, since the perimeter can be simulated much easier than the total mass of the cluster.

The 'gradient percolation' method [9] consists of studying the properties of the clusters and the cluster perimeters in the presence of a constant gradient of concentration along one direction z (here x or y). An 'infinite' cluster exists in the high-concentration region $p > p_c$. This infinite cluster has a frontier located in the region of concentration p_c. This frontier has a width ω_f which depends on the gradient of concentration ∇p through a power-law behaviour $\omega_f \sim |\nabla p|^{-\nu(1+\nu)}$.

At distances shorter than ω_f the frontier is similar to the hull of the infinite percolating cluster and has a fractal dimension: $d_H = 1 + 1/\nu$. This approach is used here to determine $p_c(\Delta)$ and $\nu_i(\Delta)$ (or $d_H^{(i)}(\Delta)$). A gradient of concentration, ∇p, is superimposed on the multi-layered structure, along one of the two directions $z = x$ or $z = y$. When z is along x, the width is expected to vary as $\omega_f \sim |\nabla p|^{-\nu_x/(1+\nu_x)}$, while when z is along y the width is expected to vary as $\omega_f \sim |\nabla p|^{-\nu_y/(1+\nu_y)}$. The number of sites N_f on the frontier also presents a power-law behaviour of ω_f (and therefore of the gradient) $N_f \sim \omega_f^{d_H - 1}$. In practice, it is convenient to count the number of occupied sites on the front N_{fA} and the empty sites N_{fB}, which are nearest neighbours of these occupied front sites, in order to get a precise method of calculating p_c: $p_c(\nabla p) = N_{fA}/(N_{fA} + N_{fB})$ and $p_c(0) = p_c$. In the anisotropic case we have to take care that now the critical concentrations have to be calculated separately in both kinds of layers: $p_c^{(i)}(\nabla p) = N_{fA}^{(i)}/(N_{fA}^{(i)} + N_{fB}^{(i)})$ for $i = 1$ or 2. We use this method here to determine $\bar{p}_c$. In the present calculations, the 'gradient percolation' approach is nevertheless limited to values $\Delta < 0.25$. This is due to the fact that for large anisotropy, a too-small value of the gradient is needed to avoid the frontier reaching a concentration of $p = 1$, in which case the method would become inaccurate.

In the 'cluster perimeter' method [10] the algorithm simulates the perimeter of a random cluster as if it was a self-avoiding walk, ignoring all sites that are not adjacent to the perimeter, which saves much computation time. Using a similar algorithm to that developed by Ziff *et al* [11] and a dynamic memory allocation method, one can simulate very large and very anisotropic clusters almost without any finite lattice size effects (which is crucial in the present model).

In order to find the phase diagram $\bar{p}_c(\Delta)$ we have used a modified version of the method used by Ziff [12] to find p_c with high accuracy. The main idea is that at the critical point there is an equal probability for an arbitrary perimeter of a large cluster to be either internal or external. Let $R(\Delta, \bar{p})$ denote the ratio between the number of

external and internal perimeters, at the point $(\Delta, \bar{p})$ in the phase space. For a given fixed value of Δ we have calculated $R(\Delta, \bar{p})$ for several values of $\bar{p}$, and extrapolated (using linear regression) the value of $\bar{p}_c$ such that $R(\Delta, \bar{p}_c) = 1$. The phase diagram $\Delta(\bar{p}_c)$ for anisotropic percolation is shown in figure 1. It is seen that a critical line separates the phase space into two regimes, A and B. Regime A contains systems with only finite clusters while B contains systems with infinite cluster in agreement with what is known about the line $\Delta = 0$ (isotropic percolation). Both the 'gradient percolation' and the 'cluster perimeter' methods gave the same results (for $0 < \Delta < 0.25$) with a precision of three digits (see [11] and [12] for a discussion about the accuracy of the 'cluster perimeter' method).
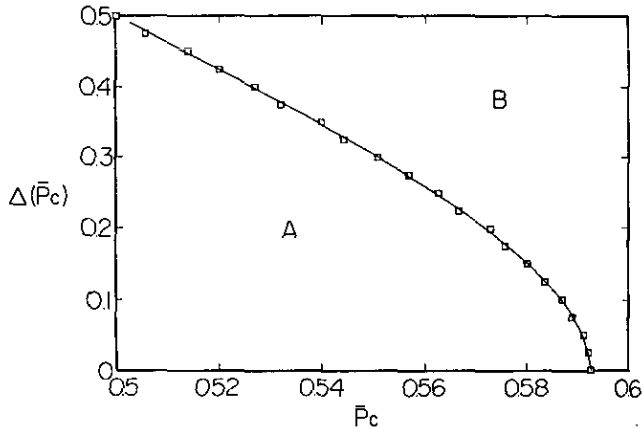

Figure 1. Phase diagram $\Delta(\bar{p}_c)$ for 2D multi-layered percolation.

We have also checked the 'alternating' percolation model and found that $\bar{p}_c(\Delta)$ is smaller than that of the 'random' model. Yet it is clear that two points in the phase space are common to the two models, namely $(\Delta = 0, \bar{p} = p_c)$ and $(\Delta = 0.5, \bar{p} = 0.5)$.

We now present results describing the properties of the anisotropic clusters on the critical line. This is done by estimating the correlation length exponents in the x and y directions independently. Let $\langle X(n) \rangle$ denote the RMS estimation of $X(t + n - 1) - X(t)$ where $X(t)$ is the x coordinate of the perimeter after t steps from its (arbitrary) origin. We expect that $\langle X(n) \rangle \sim n^{\bar{\nu}_x}$ and $\langle Y(n) \rangle \sim n^{\bar{\nu}_y}$, where $\bar{\nu}_i \equiv 1/d_H^{(i)} = \sigma_H \cdot \nu_i$. We have calculated $\bar{\nu}_x$ using a linear regression of $\log(\langle X(n) \rangle)$ against $\log(n)$, where the $\langle X(n) \rangle$ measurements were taken from very large and open (i.e. that have not been closed) perimeters [12]. The same procedure was repeated to estimate $\bar{\nu}_y$. The values of $\bar{\nu}_x$ and $\bar{\nu}_y$ measured on the critical line as a function of Δ, are shown in figure 2. Again, in the range of Δ available in the 'gradient percolation' method, the two methods gave similar results. When these measurements were taken on the 'alternating' model, both $\bar{\nu}_x$ and $\bar{\nu}_y$ were varying in the range 0.572 ± 0.004 for a wide range of Δ $(0.1, \ldots, 0.4)$. This result is in excellent agreement with the isotropic value of $d_f = 1.75 = 1/0.571 \ldots$, and demonstrates the sensitivity of our measurements to the anisotropy in the 'random' model.

Another feature that emerges from figure 2 is the finite-size effect for the size of the perimeter being analysed. As mentioned above, we do not take finite closed perimeters into account but only parts of the perimeter of the infinite cluster. Since
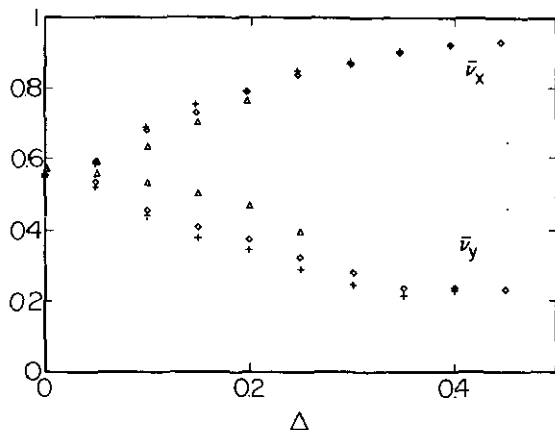
Figure 2. Measured values of the exponents $\bar{\nu}_x$ and $\bar{\nu}_y$ as a function of Δ for different ranges of n: $n \sim 1000$ ($\triangle$), $n \sim 10\,000$ ($\diamond$), $n \sim 100\,000$ (+).

this is impossible, we truncate the perimeter after n (of the order of 10^6) steps. It is seen from figure 2 that for different regimes of n, different values of $\bar{\nu}_x$ and $\bar{\nu}_y$ are estimated, such that $\bar{\nu}_x$ tends to increase and $\bar{\nu}_y$ tends to decrease as n increases. This suggests that for any $\Delta > 0$, $\bar{\nu}_x$ and $\bar{\nu}_y$ will approach asymptotically the values obtained for Δ approaching $\frac{1}{2}$.

Moreover as seen in figure 2, $\bar{\nu}_x$ and $\bar{\nu}_y$ tend to some constant values as Δ increases. Thus we assume that $\bar{\nu}_x(\Delta \to 0, n) = \bar{\nu}$, $\bar{\nu}_x(\Delta \to \frac{1}{2}, n) = \bar{\nu}_\parallel$ and $\bar{\nu}_x(0 < \Delta < \frac{1}{2}, n \to \infty) = \bar{\nu}_\parallel$ and similarly for $\bar{\nu}_y$).

In order to test this assumption we will try to describe $\langle X(\Delta, n) \rangle$ by a single scaling function:

$$\langle X(\Delta, n) \rangle = n^{\bar{\nu}} f_x(\Delta) \cdot n). \tag{4}$$

with $\bar{\nu} = 1/d_f = 4/7$. A similar scaling law will be assumed for $\langle Y(\Delta, n) \rangle$. In order to get the above properties of $\bar{\nu}_x(\Delta, n)$, we expect $f_x(u \to 0) = \text{constant}$, $f_x(u \to \infty) \sim u^{\bar{\nu}_\parallel - \bar{\nu}}$, $g_x(\Delta \to 0) = 0$ and also $g_x(\Delta \to \frac{1}{2}) = \infty$. Similarly, we define f_y and g_y. Using numerical fitting methods, we have found a function $g_x(\Delta)$ such that all the measured points of $\langle X(\Delta, n) \rangle$ collapse to a single line which corresponds to f_x in (4). Figure 3 shows $\log(f)$ as a function of $\log(n \cdot g(\Delta))$. It is interesting to note that all the points of $\langle X(\Delta, n) \rangle$ measured for different values of Δ and n on the critical line, show an excellent agreement with a single scaling function. It can be seen that as $\log(n \cdot g(\Delta)) \to -\infty$ (i.e. as $n \cdot g(\Delta) \to 0$), $\log(f) \to \text{constant}$, while in the other regime, when $\log(n \cdot g(\Delta)) \to \infty$, $\log(f)$ is a linear function. From this linearity one can derive $\bar{\nu}_\parallel$ and $\bar{\nu}_\perp$ using the relations:

$$\log(f_x) \sim (\bar{\nu}_\parallel - \bar{\nu}) \log(n \cdot g(\Delta)) \tag{5a}$$

$$\log(f_y) \sim (\bar{\nu}_\perp - \bar{\nu}) \log(n \cdot g(\Delta)). \tag{5b}$$

From the asymptotic slopes of $\log(f_x)$ and $\log(f_y)$ we obtain $\bar{\nu}_\parallel = 0.94 \pm 0.01$ and $\bar{\nu}_\perp = 0.21 \pm 0.01$. If we assume that the exponent τ_H in the anisotropic system has the same value as the isotropic one, namely $\tau_H = \frac{15}{7}$, then one can see that $(3b)$ is satisfied with 1% accuracy.
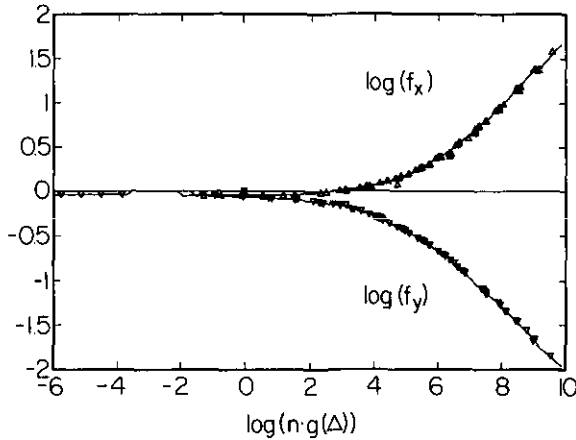
Figure 3. The scaling functions (5a) and (5b). The upper curve ($\triangle$) represents $\log(f_x)$, and the lower curve ($\triangledown$) $\log(f_y)$. The values of $\bar{\nu}_\perp$ and $\bar{\nu}_\parallel$ are obtained from the asymptotic slopes of these curves.

To check this assumption about τ_H, we measure τ_H directly using the cluster size distribution $n_s(p) \sim s^{-\tau_H} \cdot \exp(-|p-p_c|^{1/\sigma_H} \cdot s)$, where s is the cluster size (length of the perimeter) and p is the percolation concentration. We assume that n_s of our anisotropic model behaves as $s^{-\tau_H}$ on the critical line. It has been shown [11] that it is much more accurate to estimate τ_H from the number of clusters that are greater than s:

$$N_s \sim \int_s^\infty x^{-\tau_H}\, \mathrm{d}x \sim s^{1-\tau_H}. \tag{6}$$

From the calculated values of N_s, τ_H is estimated using linear regression, $\log(N_s) = (1-\tau_H)\log(s) + \text{constant}$. Figure 4 shows some values of τ_H that were calculated on the critical line using both equations (6) and (3b). For the former it is seen that the numerical values found for τ_H are within 2% of the isotropic value $\frac{15}{7}$. However, there seems to be a systematic deviation which might be attributed to the fact that we do not measure exactly on the critical line. This needs to be checked in the future.
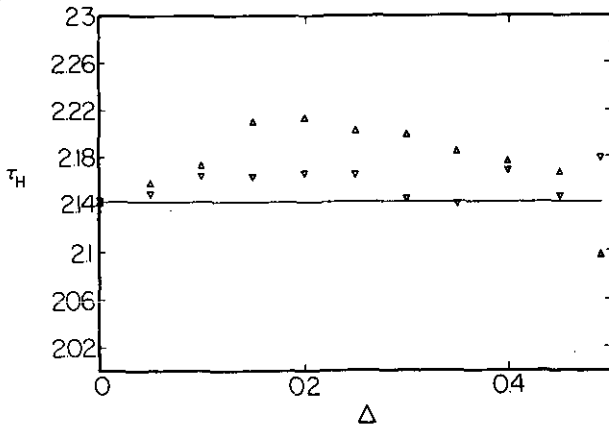
Figure 4. Plot of τ_H as a function of Δ found directly using (6) ($\triangle$) and using (3b) ($\triangledown$). The line represents the isotropic value $\tau_H = \frac{15}{7}$.

In order to estimate the correlation length exponents $\nu_\perp$ and $\nu_\parallel$ we assume that, similar to τ_H, σ_H is also not affected by anisotropy. Substituting $\sigma_H = \frac{3}{7}$ in (3a) we conclude that $\nu_\parallel = 2.19 \pm 0.02$ and $\nu_\perp = 0.49 \pm 0.02$ which suggest that $\nu_\parallel = \frac{13}{6}$ and $\nu_\perp = \frac{1}{2}$. These values differ significantly from the values obtained for directed percolation [8].

It has been shown here that even a small anisotropy introduced by any finite Δ results in long-range correlations. These correlations introduce anisotropic clusters with fractal dimensions: $d_H^\perp (= 1/\bar{\nu}_\perp)$ and $d_H^\parallel (= 1/\bar{\nu}_\parallel)$. The structure of the cluster is therefore self-affine and not self-similar as in isotropic percolation.

However, our numerical data suggests that the exponent τ_H has the same value for the anisotropic percolation as for the ordinary isotropic one. Thus, expressing τ_H as $1 + d/d_H$ and substituting into (3b), it follows that

$$\frac{1}{d_H} = \frac{1}{d} \sum_{i=1}^{d} \frac{1}{d_G^{(i)}}. \tag{7}$$

Our direct measurements of $d_H^\perp$ and $d_H^\parallel$ also support this conjecture.

Assuming the validity of (7), it follows that for $d = 2$, $d_H^\perp$ and $d_H^\parallel$ are not independent and represent only one degree of freedom. Thus, similar to the isotropic case, the anisotropic model is characterized by only two independent critical exponents.

It is interesting to compare our results with those argued by Obukhov [7]. His conclusions for layered percolation are: (a) the critical line $\bar{p}_c(\Delta)$ is linear and (b) the transition is of infinite order. Our numerical results do not support any of these conclusions, as seen from figure 1, $\bar{p}_c(\Delta)$ is not linear and our support of the scaling function (1) and the hyperscaling relation (2) suggest a second-order transition.

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