

Scaling theory of hydrodynamic dispersion in percolation networks

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Real-space renormalization-group arguments are used to derive scaling relations for the mean displacement $\langle R \rangle$ and the variance $\langle (R - \langle R \rangle)^2 \rangle$ of a tracer particle in a fluid flowing through a heterogeneous material which is near a percolation threshold p_c ; both small- and large-Peclet-number regions are studied. The existence of a noninteger-power-law dependence of $\langle R \rangle$ and $\langle (R - \langle R \rangle)^2 \rangle$ on time and the strength of flow, which cannot be described by a convection-diffusion equation, is revealed. Particularly at large Peclet numbers, the variance exhibits anomalously fast time dependence and an associated divergence near p_c . As $p \rightarrow p_c$, the region dominated by convection extends prominently, while the region controlled by diffusion shrinks.

Dispersion is a very common phenomenon in transport through porous media and appears in many important problems, ranging from chromatography to petroleum engineering.¹ Recently, much interest has been devoted to this subject and it has been shown that the heterogeneity of porous materials has an important influence on dispersion.²⁻⁴ Now, a percolation network is a good model for the heterogeneous geometries of pore structures so it seems reasonable to study dispersion in percolation networks. Many recent papers have revealed various types of anomalous dynamics of percolation systems in the vicinity of a threshold p_c .⁵ The essential feature is that the self-similarity of percolation clusters causes nonanalytic power-law behavior and associated scaling relations near p_c , as in thermal critical phenomena. In the case of dispersion, therefore, we can expect the existence of similar anomalies which cannot be described by a conventional convection-diffusion equation approach.¹ The purpose of this work is to investigate hydrodynamic dispersion in percolation networks and to clarify its scaling behavior near p_c .

We consider the following process. The system is modeled by a regular network of bonds and sites in which a single fluid is flowing. We assume that the background flow in each bond obeys Darcy's law $u = k\alpha/\eta$, where u is the velocity of flow, k the permeability of bonds, η the viscosity of a fluid, and α the pressure gradient. The probability distribution of k is given by a binary distribution $P(k) = p\delta(k - k_0) + (1-p)\delta(k)$, that is, a bond-percolation problem is treated. When p is below p_c , no path of bonds spans a macroscopic system. As p_c is approached from below, the maximum size of regions connected by bonds (clusters) diverges as $\xi \sim |p - p_c|^{-\nu}$, where $\epsilon = p - p_c$, ξ is the correlation length, and ν the correlation length exponent. Just above p_c , macroscopic paths for the fluid do exist, but they are extremely tortuous. This is why a percolation network near p_c is a good prototype for a highly heterogeneous material. Above p_c , ξ still has a physical significance, that of the linear size of tortuous regions. The flow field in our problem is the same as the current field in a corresponding random resis-

tor network. We release a tracer particle in the network and trace the time dependence of its position. Two mechanisms drive dispersion: molecular diffusion and convection caused by the background flow. A dispersion coefficient due to convection is estimated as ul , where l is the lattice constant (length of a bond). Hence, the relative importance of the mechanisms is measured by the Peclet number $P_e = ul/D$, where D is the molecular diffusion coefficient. At small-Peclet numbers, convection is negligible and the dispersion coefficient is determined by D . At large-Peclet numbers, molecular diffusion is negligible and the particle follows the background flow completely. In this work, we discuss both the small- and large-Peclet-number regions.

We will characterize the dispersion by the cumulant averages $C_j(t) = \langle R^j(t) \rangle_c$ of the position of the particle in the flow. So, C_1 gives the mean particle velocity and C_2 is a direct measure of the amount of dispersion. We adopt a real-space renormalization-group (RSRG) method to calculate the C 's. The present scheme is quite similar to that for diffusion on percolation lattices developed before.^{6,7} Four parameters characterize the problem: the lattice constant l , the percolation probability p , the velocity u of the background flow, and the molecular diffusion coefficient D . All physical quantities are functions of these four parameters. Now we assume the existence of a RSRG transformation with a rescaling factor b . One might think that this assumption is justified by the self-similarity of percolation clusters.⁸ Recently, however, multiscaling relations have been identified in some geometrical properties of fractals like the current distribution in percolation networks.⁹ It is, nevertheless, believed that dynamical properties of fractals such as diffusion and sound propagation (or equivalently fraction modes) obey simple scaling.¹⁰ The essential point is that in these time-dependent properties, there exists a characteristic length scale associated with time and the process is dominated by clusters, paths, etc. of this length scale. In case of dispersion, therefore, we consider that moments of the particle is governed by characteristic paths of length of order $\langle R(t) \rangle$ and simple

scaling holds. Dimensional analysis shows that the recursion relations for l , p , u , and D are of the form $l' = bl$, $p' = f(p, P_e) = f(p)$, $u' = g(p, P_e)u$, and $D' = h(p, P_e)D$, where a prime denotes a renormalized quantity. Here we invoke the fact that p is a purely geometrical quantity and independent of u and D .

First, we study the small-Peclet-number (weak-flow) region. Dimensional analysis leads to $C_j(l, p, u, D; t) = l^j C_j^*(p, P_e; t/w_0)$, where $w_0 = l^2/D$. At low P_e , where molecular diffusion is dominant, w_0 is the natural time for the problem; it is via such arguments that we can obtain different results in the different regions. In the same spirit we expand C_j^* in a power series in P_e ,

$$C_j^*(p, P_e; t/w_0) = \sum_{n=0}^{\infty} P_e^n C_{j,n}(p, t/w_0). \tag{1}$$

This expansion is not expected to behave well near p_c . Our approach is to write down the series and then examine its convergence properties. Recursion relations are also expanded as $\epsilon' = \lambda_p \epsilon + O(\epsilon^2)$, $u' = \lambda_{u,0} u + O(\epsilon, P_e)$, and $D' = \lambda_{D,0} D + O(\epsilon, P_e)$, where $\epsilon = p - p_c$, $\lambda_p = df/dp|_{p_c}$, $\lambda_{u,0} = g(p_c, 0)$, and $\lambda_{D,0} = h(p_c, 0)$. These expansions are always assumed to work well in RSRG calculations because of the analyticity of recursion relations. The key to extracting information from the transformation, of course, is the basic RG idea that relevant physical quantities are kept invariant under the transformation; thus $l^j P_e^n C_{j,n}(p', t/w_0) = l^j P_e^n C_{j,n}(p, t/w_0)$. Substituting the recursion relations for l, p, u , and D , and keeping the lowest-order terms in ϵ and P_e , we get a recursion relation for $C_{j,n}$,

$$C_{j,n}(\epsilon, t/w_0) = b^j (\lambda_{u,0} b / \lambda_{D,0})^n C_{j,n}(\lambda_p \epsilon, \lambda_{D,0} t / b^2 w_0). \tag{2}$$

Recursion relations for u and D also yield $u(l') = u(bl) = \lambda_{u,0} u(l)$ and $D(l') = D(bl) = \lambda_{D,0} D(l)$. These lead to $u(\xi) \propto \xi^{-\phi_0}$ and $D(\xi) \propto \xi^{-\theta_0}$ where $\phi_0 = -\ln(\lambda_{u,0})/\ln(b)$ and $\theta_0 = -\ln(\lambda_{D,0})/\ln(b)$. We can relate ϕ_0 and θ_0 to the conductivity exponent μ and other known exponents for percolation.¹¹ To do this, first consider that there are two factors which cause the conduction to vanish near p_c : increase of the effective length $L(\xi)$ of a channel and decrease of the effective number (area) $A(\xi)$ of channels. Here we regard the network as composed of many channels. By definition, $I(\xi)/\xi^{d-1} = \sigma(\xi)V(\xi)/\xi$, where I is the current, V the voltage, and $\sigma(\xi) \propto \xi^{-\mu/\nu}$ the conductivity. Since the current is in proportion to $A(\xi)$ and the conductance of each channel is in inverse proportion to $L(\xi)$, we obtain $I(\xi)/V(\xi) \propto A(\xi)/L(\xi)$. It follows that $A(\xi)/L(\xi) \propto \xi^{d-2-\mu/\nu}$. On the other hand, the mass $M(\xi)$ of the network obeys $M(\xi) \sim A(\xi)L(\xi) \propto \rho(\xi)\xi^d$, where $\rho(\xi) \propto \xi^{-\beta/\nu}$ is the density and β the percolation probability exponent. Then we find $L(\xi) \propto \xi^{1+(\mu-\beta)/2\nu}$. Now, consider the fluid flow velocity $u(\xi)$. Since the time necessary to travel each channel is in proportion to $L(\xi)$, the velocity varies as¹²

$$u(\xi) \sim \xi/L(\xi) \propto \xi^{-(\mu-\beta)/2\nu} \sim [\sigma(\xi)/\rho(\xi)]^{1/2},$$

and thus $\phi_0 = (\mu - \beta)/2\nu$. The situation is the same for all velocities, i.e., the anomalously long length $L(\xi)$ gives anomalously slow velocities with very similar scalings.^{13,14} As for the diffusion coefficient, Einstein's relation tells us that $D(\xi) \sim \sigma(\xi)/\rho(\xi) \propto \xi^{(\mu-\beta)/\nu}$.¹⁵ Consequently, $\theta_0 = (\mu - \beta)/\nu$.

The recursion relation (2) together with expressions for ϕ_0 and θ_0 ($\lambda_{u,0}$ and $\lambda_{D,0}$) show that $C_{j,n}$ is a generalized homogeneous function satisfying the scaling relation¹⁶

$$C_{j,n}(\xi, t/w_0) = (t/w_0)^{(j\chi_0+n)/2} F_{j,n}(t/\tau_0), \tag{3}$$

where $\chi_0 = 1/(1 + \phi_0)$ and $\tau_0 = \xi^{2/\chi_0} w_0$. We further deduce that $F_{j,n}(x) \rightarrow 1$ ($x \rightarrow 0$) and $F_{j,n}(x) \rightarrow x^{(2-j\chi_0-n)/2}$ ($x \rightarrow \infty, j=1,2$), 0 ($x \rightarrow \infty, j \geq 3$). This asymptotic behavior of $F_{j,n}$ comes from the requirement that at $t \ll \tau_0$, $C_{j,n}$ is independent of ξ , and when $t \gg \tau_0$, $C_{j,n}$ becomes Gaussian, i.e., $C_{j,n} \propto t$ ($j=1,2$) and 0 ($j \geq 3$). We first derive the conditions under which molecular diffusion dominates the dispersion. Very simply, this will be the case if the expansion in P_e is rapidly converging, i.e., $P_e^n C_{j,n} / P_e^{n+1} C_{j,n+1} \gg 1$. Inserting Eq. (3) into this condition, we obtain (a),

$$P_e \ll \xi^{-1/\chi_0}$$

or (b),

$$P_e \ll 1, \quad ut/l \ll P_e^{-1}.$$

Thus the Peclet number needed to ensure "weak" flow, or dominance of molecular diffusion, becomes vanishingly small as p_c is approached and ξ diverges. Furthermore, we find that, at larger P_e 's, which are nevertheless small compared to unity, "weak flow" may nevertheless be observed at short times. Under these conditions, $C_1^* \simeq P_e C_{1,1}$ and $C_2^* \simeq C_{2,0}$, because $C_{1,0} = 0$. Equation (3) then directly yields the scaling relations

$$\langle R(t) \rangle / l = P_e (t/w_0)^{(\chi_0+1)/2} F_0(t/\tau_0) \tag{4}$$

$$\propto t^{(\chi_0+1)/2} u \quad (t \ll \tau_0) \tag{5}$$

$$\propto \xi^{(\chi_0-1)/\chi_0} t u \quad (t \gg \tau_0), \tag{6}$$

$$\langle R(t)^2 \rangle_c / l^2 = (t/w_0)^{\chi_0} G_0(t/\tau_0) \tag{7}$$

$$\propto t^{\chi_0} \quad (t \ll \tau_0) \tag{8}$$

$$\propto \xi^{2(\chi_0-1)/\chi_0} t \quad (t \gg \tau_0). \tag{9}$$

Equations (7)–(9) coincide with known results about diffusion on percolation lattices.¹⁵

The mean displacement $\langle R(t) \rangle_B$ of a diffusing particle on percolation lattices in the presence of an external bias γ like a uniform electric field exhibits nonanalytic behavior.⁷ Below a critical bias γ_c (in the weak bias region), $\langle R(t) \rangle_B$ varies as

$$\langle R(t) \rangle_B \propto t^{\chi_0} \gamma \quad (t \ll \tau_0). \tag{10}$$

Comparing with Eq. (5), we find that the critical exponent χ_0 for biased diffusion is different from that, $(\chi_0 + 1)/2$, for dispersive transport, so the two processes

belong to different universality classes. This difference probably stems from the fact that in biased diffusion, the bias is applied uniformly in the Euclidean space, whereas in dispersion, the bias (background flow or pressure gradient) is exerted only along the cluster. The bias of the former type is less effective than the latter and the associated critical exponent χ_0 is smaller than that, $(\chi_0 + 1)/2$, of dispersion. The type of bias turns out to be an important factor in determining the universality class. Above γ_c (strong bias), moreover, trapping of the particle by dead ends or backbends of the backbone plays a crucial role, and definite conclusions on the temporal evolution has not been drawn yet. Such is not the case in dispersion, as we shall see next.

The RSRG technique is applicable to the large-Peclet-number (strong-flow) region in much the same way as it is to the small-Peclet-number region. We start with $C_j(l, p, u, D; t) = l^j C_j^*(p, P_e; t/w_\infty)$, where $w_\infty = l/u$; w_∞ is the natural time for strong flow. Also, C_j^* , g , and h are now expanded as power series in P_e^{-1} , instead of P_e , as

$$C_j^*(p, P_e; t/w_\infty) = \sum_{n=0}^{\infty} P_e^{-n} C_{j,-n}(p, t/w_\infty), \quad (11)$$

$u' = \lambda_{u,\infty} u + O(\epsilon, P_e^{-1})$, and $D' = \lambda_{D,\infty} D + O(\epsilon, P_e^{-1})$, where $\lambda_{u,\infty} = g(p_c, \infty)$ and $\lambda_{D,\infty} = h(p_c, \infty)$. Substitution of these equations into the relation

$$l'^j P_e'^{-n} C_{j,-n}(p', t/w'_\infty) = l^j P_e^{-n} C_{j,-n}(p, t/w_\infty)$$

gives rise to

$$C_{j,-n}(\epsilon, t/w_\infty) = b^j (\lambda_{u,\infty} b / \lambda_{D,\infty})^{-n} \times C_{j,-n}(\lambda_p \epsilon, \lambda_{u,\infty} t / b w_\infty). \quad (12)$$

Critical exponents are expressed similarly. In this case, however, particle motion is almost restricted to the backbone of the percolation cluster. The probability of getting into dead ends is of order P_e^{-1} . The displacement R_{BB} of the particle in the backbone is of order P_e , while that, R_{DE} , on a dead end is of order unity because it is not carried by the fluid. It follows that $\langle R \rangle_{DE} \sim P_e^{-1} R_{DE} \sim P_e^{-1}$ and $\langle R \rangle_{BB} \sim R_{BB} \sim P_e$. To lowest order in P_e^{-1} , $\langle R \rangle_{DE}$ is negligible in comparison with $\langle R \rangle_{BB}$. This situation holds for higher-order cumulants. Then the relevant density is not $\rho(\xi)$ but that of the backbone, $\rho_B(\xi) \propto \xi^{-\beta_B/\nu}$, where β_B is the backbone probability exponent. In place of ϕ_0 and θ_0 , therefore, we obtain $\phi_\infty = -\ln(\lambda_{u,\infty})/\ln(b) = (\mu - \beta_B)/2\nu$ and $\theta_\infty = -\ln(\lambda_{D,\infty})/\ln(b) = (\mu - \beta_B)/\nu$. It should be emphasized that dead ends are irrelevant to cumulants of the displacement but relevant to those of the time. Since the time T_{DE} spent on a dead end is of order unity and that T_{BB} in the backbone is of order P_e^{-1} , $\langle T \rangle_{DE} \sim P_e^{-1} T_{DE} \sim P_e^{-1}$ and $\langle T \rangle_{BB} \sim T_{BB} \sim P_e^{-1}$. We find that $\langle T \rangle_{DE} < \langle T \rangle_{BB}$ and

$$\langle T \rangle \sim \langle 1/R \rangle \neq 1/\langle R \rangle. \quad (13)$$

Equation (12) leads to the scaling relation for $C_{j,-n}$,

$$C_{j,-n}(\xi, t/w_\infty) = (t/w_\infty)^{j\chi_\infty - n} F_{j,-n}(t/\tau_\infty), \quad (14)$$

where $\chi_\infty = 1/(1 + \phi_\infty)$ and $\tau_\infty = \xi^{1/\chi_\infty} w_\infty$. We obtain $F_{j,-n}$ with the arguments used previously, getting $F_{j,-n}(x) \rightarrow 1$ ($x \rightarrow 0$) and $F_{j,-n}(x) \rightarrow x^{1-j\chi_\infty+n}$ ($x \rightarrow \infty$, $j=1,2$), 0 ($x \rightarrow \infty$, $j \geq 3$). As before, we derive the conditions required for strong flow by insisting that the P_e^{-1} series is rapidly converging. We find that dispersion is dominated by convection when [(c)]

$$P_e \gg 1,$$

or (d),

$$P_e \gg \xi^{-1/\chi_\infty}, \quad ut/l \gg P_e^{-1}.$$

Again, just as in the weak-flow case, scaling relations are derived from Eq. (14):

$$\langle R(t) \rangle / l = (t/w_\infty)^{\chi_\infty} F_\infty(t/\tau_\infty) \quad (15)$$

$$\propto t^{\chi_\infty} u^{\chi_\infty} \quad (t \ll \tau_\infty) \quad (16)$$

$$\propto \xi^{(\chi_\infty - 1)/\chi_\infty} t u \quad (t \gg \tau_\infty), \quad (17)$$

$$\langle R(t)^2 \rangle_c / l^2 = (t/w_\infty)^{2\chi_\infty} G_\infty(t/\tau_\infty) \quad (18)$$

$$\propto t^{2\chi_\infty} u^{2\chi_\infty} \quad (t \ll \tau_\infty) \quad (19)$$

$$\propto \xi^{(2\chi_\infty - 1)/\chi_\infty} t u \quad (t \gg \tau_\infty). \quad (20)$$

Let us now discuss our results. First notice that $1 > \chi_\infty > \chi_0$, because $\mu > \beta_B > \beta$. It should be emphasized that the strong-flow (convective) region [conditions (c) and (d)] extends notably near p_c as $\xi \sim (p - p_c)^{-\nu} \gg 1$. Especially in the long-time limit $t \rightarrow \infty$, convection is dominant at $P_e \gg \xi^{-1/\chi_\infty}$ and negligible only when $P_e \ll \xi^{-1/\chi_0}$. It should be noted that the noninteger power-law behavior of $\langle R(t) \rangle$ and $\langle R(t)^2 \rangle_c$ in Eqs. (4)–(9) and (15)–(20) cannot be described by a convection-diffusion equation. Furthermore, we find that anomalously fast time-dependence and an associated divergence of $\langle R(t)^2 \rangle_c$ as $p \rightarrow p_c$ in the strong-flow region because $\chi_\infty > 0.5$ at dimensions less than 6.¹³

Since χ_∞ is less than unity, Eq. (16) represents anomalously slow movement of a particle. These kinds of anomalies are generally observed in particle motion on fractals and reflect their self-similarity. A geometrical interpretation is straightforward.¹⁴ The backbone of a percolation cluster describes not a smooth curve but a singularly irregular curve. The effective contour length S of the backbone is much longer than the distance R in the Euclidean space and expressed as $S \propto R^{1/\chi_\infty}$. On the other hand, there is nothing unusual about particle motion along the backbone and $S \propto tu$. Even if a particle travels S along the structure, however, we measure anomalously slow behavior $R \propto S^{\chi_\infty} \propto t^{\chi_\infty} u^{\chi_\infty}$ in the Euclidean space. This situation is the same as in other unusual dynamics of fractals such as diffusion and wave propagation.¹⁴ Equation (5) also expresses slower-than-normal time dependence because $\chi_0 < 1$. There is a significant qualitative difference, however. In the strong-flow region, $\langle R(t) \rangle$ is a nonanalytic function of the strength of the bias (velocity), whereas in the weak-flow region, $\langle R(t) \rangle$ is just in

proportion to the bias.

In contrast, $\langle R^2(t) \rangle_c$ shows anomalously fast behavior, as mentioned before. In addition, we find from Eqs. (16) and (19) that

$$\langle R^2(t) \rangle_c \sim \langle R(t) \rangle^2 \quad (t \ll \tau_\infty). \quad (21)$$

A geometrical meaning of this relation is obvious. In this time scale, $\langle R(t) \rangle \ll \xi$ and the system is self-similar. The network contains many paths, at all length scales, and a deviation of displacement in different paths of linear size L is of order L . Since a particle travels on each path with a probability of same order, its variance after passing L on average becomes of order L^2 . The anomalously large variance reflects this purely geometrical nature of particle motion. Generally in fractals, the variance of a geometrical quantity is in proportion to the square of its average. This is a significant characteristic of self-similarity and is observed in various quantities.¹⁷ At the same time, this argument suggests that there is no significant difference between longitudinal and transverse variances (dispersion coefficients). In this length scale, the system is isotropic and the particle cannot recognize in which direction the fluid is flowing as a whole. Thus numerical coefficients may be different but scaling relations and associated critical exponents should be equal.

Hitherto, we discussed scaling relations in terms of the flow velocity u . In most measurements, however, the pressure gradient α , and not u , is the control parameter, and under a constant pressure gradient u goes to zero as $p \rightarrow p_c$. In other words u depends on p . Since the fluid is flowing only through the backbone, u is given by

$$u(p) = u(\xi) = \xi^{-\phi_\infty} u_0, \quad (22)$$

where $u_0 = k_0 \alpha / \eta$. Inserting Eq. (22) and rewriting conditions (a)–(d), Eqs. (4)–(9), and Eqs. (15)–(20), we have our various criteria and scaling relations under a given pressure gradient. Note that the meaning of the ξ dependence of u in Eq. (22) is totally different from that used in deriving expressions for ϕ_0 and θ_0 or ϕ_∞ and θ_∞ . Equation (22) represents the dependence of the velocity of microscopic flow at length scale l on a given percolation probability, while those in the latter case stand for the dependence of an effective velocity, which a particle feels, on the length scale ξ .

Dispersion in the weak-flow region and in the strong-flow region is governed by different critical exponents χ_0 and χ_∞ . This difference originates in the existence of dead ends at all length scales up to ξ . In the weak-flow region, a particle is distributed uniformly over the whole cluster, whereas in the strong-flow region a particle is confined only to the backbone. This indicates that to investigate crossover behavior in the intermediate-flow region, careful arguments about exchange dynamics of a particle between the backbone and dead ends are necessary, as suggested by de Gennes.² It is noticed that the scaling theory developed here holds for all self-similar systems such as Sierpinski gaskets and diffusion-limited aggregates, as well as percolation networks. In fractals with no dead ends, however, processes in both the weak- and strong-flow regions are described by the same critical exponents.

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