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LETTER TO THE EDITOR

Properties of the density relaxation function in classical diffusion models with percolation transition

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Abstract. The relation between the density relaxation function Φ and the pair connectedness is shown. Static and dynamical scaling for Φ and quantities related to it are derived from percolation scaling theory. Due to finite clusters Φ contains a non-ergodic singularity even in the conducting phase, whence a Green–Kubo identity does not hold. The form factor of this singularity is discussed. For $d \geqslant 3$ the static polarisability can be related to a diverging characteristic length also above the threshold. Contributions come from confinement in finite clusters and from the structure of the infinite cluster.

Classical diffusion in disordered media is of much practical and theoretical interest. The conductor insulator transition in such systems is closely related to percolation phenomena. (For reviews on percolation theory see Deutscher *et al* (1983), Stauffer (1979) and Essam (1980).) The diffusion models can be of hopping character, like 'the ant in the labyrinth', i.e. a random walker on percolation clusters (de Gennes 1976), or they can be continuous, like the Lorentz model (see e.g. Hauge 1974). In these models the system becomes insulating if the paths of the diffusing particle are blocked at the percolation threshold p_c. Since theoretical approaches (Götze *et al* 1981, Odagaki and Lax 1981, Webman 1981) heavily use the concept of the density relaxation function, it seems to be worth investigating what can be said about this function from the point of view of percolation theory.

Let us briefly list some well known results. We give here the formulae for d-dimensional lattice site and bond percolation. The transformation to continuum problems is trivial if universality is supposed to be true (see e.g. Kertész and Vicsek 1982): instead of site occupation probability one has to take the allowed volume fraction, and instead of discrete sums, integrals have to be performed.

The pair connectedness $C(\boldsymbol{r}, p, h)$ (see e.g. Essam 1980) can be written as a sum (Stauffer 1978):

$$C(\boldsymbol{r}, p, h) = \sum_s{}' s n_s(p, h) \langle \eta(\boldsymbol{r}) | C_s \rangle \tag{1}$$

where $n_s(p, h) = n_s(p) \exp(-hs)$, $n_s(p)$ is the number of s-clusters per site at an occupation probability p, $\eta(\boldsymbol{r})$ is 1 if the site at $\boldsymbol{r}$ belongs to the same cluster as the origin and 0 otherwise. $\langle \ldots | C_s \rangle$ denotes the configurational average with the condition that the origin belongs to an s-cluster. The prime on the summation symbol indicates that only finite cluster contributions are to be taken. For convenience a ghost field h

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has been introduced. (For a physical interpretation of the ghost field see Reynolds *et al* (1977). In diffusion models $h \neq 0$ could mean allowed hops via the ghost site.)

Near the critical point, $p = p_c$, $h = 0$, the pair connectedness has a scaling form (Essam 1980):

$$C(\boldsymbol{r}, \varepsilon, h) = L^{-2\beta/\nu} C(\boldsymbol{r}/L, \varepsilon L^{1/\nu}, hL^{-\beta/\nu+d}) \tag{2}$$

where $\varepsilon = p - p_c$, ($|\varepsilon| \ll 1$, $h \ll 1$ is implied here and in the following), ν is the critical exponent of the coherence length $\xi(p) \propto |\varepsilon|^{-\nu}$ and β is the critical exponent of the percolation probability $P_\infty(p) \propto \varepsilon^\beta$, $p > p_c$. The definition $P_\infty(p, h) = 1 - p_F(p, h)/p_s$; $p_F(p, h) = \Sigma' sn_s(p, h)$, where $p_s = 1$ or p for bond and site percolation, respectively, expresses for $h = 0$ the conservation of probability.

We now turn to describe the properties of the classical density relaxation function $\Phi(\boldsymbol{r}, t)$ of a tagged particle moving in a random environment, which describes the probability of the particle to be at site $\boldsymbol{r}$ at time t, if it has started ($t = 0$) at the allowed site $\boldsymbol{0}$. $\Phi(\boldsymbol{r}, t)$ is a function which can be measured in a molecular dynamics experiment. Assuming that the distribution of the particle in the allowed region is uniform at $t \to \infty$ (Vicsek 1983), one has immediately the representation

$$\Phi(\boldsymbol{r}, t \to \infty | p, h) = (1/p_s)\{\Sigma' sn_s(p, h)\langle \eta(\boldsymbol{r})/s | C_s\rangle + P_\infty(p, h)/N\} \tag{3}$$

where a formal extension to the $h \neq 0$ case has been included and the dependence on p and h has been indicated. N is the total number of lattice sites.

One quantity of interest is the probability of finding the particle for $t \to \infty$ at the same place where it has started from (see e.g. Odagaki and Lax 1982, Haus *et al* 1983). Equation (3) allows us to establish a nice relation between this quantity and the generating function of percolation theory:

$$\Phi(\boldsymbol{0}, t \to \infty | p, 0) = (1/p_s)\,\Sigma' n_s(p) \tag{4}$$

where we have used that in the thermodynamic limit ($N \to \infty$) the last term of the RHS of (3) vanishes, and that $\langle \eta(\boldsymbol{0}) | C_s \rangle = 1$. The RHS of (4) is known to exhibit a singularity $\propto |\varepsilon|^{2-\alpha}$ for $p \to p_c$ (Stauffer 1979). α is the exponent of the 'specific heat'. Equation (4) can be confirmed by an exact calculation on the Bethe lattice (Odagaki and Lax 1982, Fisher and Essam 1961).

Generalising the representation (3) to arbitrary times one can write

$$\Phi(\boldsymbol{r}, t | p, h) = \Phi_F(\boldsymbol{r}, t | p, h) + \Phi_\infty(\boldsymbol{r}, t | p, h) \tag{5}$$

where Φ_F and Φ_∞ are the two distinct contributions according to a starting point on a finite cluster or on the infinite cluster. $\Phi_\infty(\boldsymbol{r}, t \to \infty | p, h)$ vanishes in the thermodynamic limit, expressing the obvious fact that the particle diffuses away on the infinite cluster, when it started on it. Once started on a finite cluster, the particle can never escape and thus the correlations described by Φ_F do not decay. Correspondingly the Fourier and Laplace transform $\Phi(\boldsymbol{q}, z) = \Sigma_r \exp(-i\boldsymbol{qr})\, i\int_0^\infty dt \exp(izt)\Phi(\boldsymbol{r}, t)$ contains a singularity $-f(\boldsymbol{q}, p)/z$, or equivalently a $\delta(\omega)$ singularity in the Fourier spectrum, reflecting the non-ergodic behaviour of the system (Kubo 1957, Götze 1978, 1981, 1982). Representations (5) and (3) show that even for $p \geq p_c$ there is a non-ergodic contribution to $\Phi(\boldsymbol{r}, t)$. This has the following consequence: the representation for small wavenumbers, known as the Green–Kubo identity

$$\lim_{q \to 0} \Phi(\boldsymbol{q}, z) = -1/(z + q^2 K(z)),$$

with $K(z)$ being the frequency dependent diffusion coefficient, is here an approximation only. The non-decaying correlations stemming from finite clusters are neglected thereby.

The situation is different if the particle has to be described by quantum mechanics and if the generally accepted view, that extended and localised states do not coexist, is true. Then it follows that Kubo's (1957) density relaxation function, which is the analogue of our Φ in quantum systems (Götze 1982), does not exhibit a non-ergodic singularity in the conducting phase. Some clarifying remarks may be helpful here. Our density relaxation function is not identical with the density–density correlation function $S(\boldsymbol{q}, \omega)$ of n particles, measurable in a scattering experiment. The inhomogeneous particle density, arising from static disorder (e.g. non-allowed volumina) and acting as scattering potential, yields an elastic $\delta(\omega)$ contribution to $S(\boldsymbol{q}, \omega)$. This $\delta(\omega)$ singularity is in full analogy to the one calculated for a quantum system by Belitz *et al* (1983). In the thermodynamic limit $\Phi(\boldsymbol{r}, t \to \infty) = 0$, if the particle has always the possibility to diffuse away. Thus the $\delta(\omega)$ singularity in Φ is indeed the characteristic of non-ergodicity (Kubo 1957, Götze 1978, 1981).

Comparing (1) and (3) we arrive at the relation

$$-\partial\Phi(\boldsymbol{r}, t \to \infty | p, h)/\partial h = C(\boldsymbol{r}, p, h) \qquad (6)$$

and from the scaling form (2) for C near to the critical point

$$\Phi(\boldsymbol{r}, t \to \infty | \varepsilon, h) = L^{-\beta/\nu - d}\Phi(\boldsymbol{r}/L, t \to \infty | \varepsilon L^{1/\nu}, hL^{-\beta/\nu + d}). \qquad (7)$$

Hence one concludes that the prefactor of the non-ergodic singularity has the following properties:

$$f(\boldsymbol{q}, p = p_c) \propto q^{\beta/\nu}, \qquad q \to 0, \qquad (8a)$$

$$f(\boldsymbol{q}, p) \propto |\varepsilon|^{\beta} + \text{constant}, \qquad q \gg \xi^{-1}. \qquad (8b)$$

Using further that $f(\boldsymbol{q} = \boldsymbol{0}, p) = p_F(p)/p_s$, which is easily seen from (3), and the obvious properties $f(\boldsymbol{q}, p = 0) = 1$, $f(\boldsymbol{q}, p = 1) = 0$, one can draw the qualitative behaviour of $f(\boldsymbol{q}, p)$ (figure 1).

Turning back to equation (7), we assume a dynamical scaling form in analogy to earlier ones, introduced to describe the critical time dependence of the mean square distance of the particle from the starting point $\langle(\Delta\boldsymbol{r}(t))^2\rangle$ (Straley 1980, Gefen *et al*

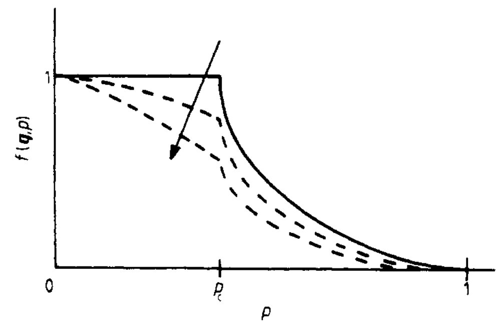

Figure 1. The prefactor $f(\boldsymbol{q}, p)$ of the non-ergodic contributions in $\Phi(\boldsymbol{q}, z)$ The full line represents $f(\boldsymbol{0}, p)$. q is growing along the arrow. At p_c there is a non-analyticity (8b).

1983), which were tested by huge Monte Carlo simulations (Pandey and Stauffer 1983):

$$\Phi(\boldsymbol{r}, t|\varepsilon, h) = L^{-\beta/\nu-d}\Phi(\boldsymbol{r}/L, t/L^{\bar{z}}|\varepsilon L^{1/\nu}, hL^{-\beta/\nu+d}) \tag{9}$$

where $\bar{z}$ is the dynamical exponent. The scaling form (9) should be valid for both Φ_{F} and Φ_∞. Using

$$\langle(\Delta\boldsymbol{r}(t))^2\rangle = \sum_{\boldsymbol{r}} r^2\Phi(\boldsymbol{r}, t)$$

then (5) implies that the mean square displacement can also be decomposed into two parts $R_{\mathrm{F}}^2(t)$ and $R_\infty^2(t)$, according to the starting position. With the help of (9) one concludes

$$R_{\mathrm{F}}^2(t) \propto |\varepsilon|^{-2\nu+\beta}, \qquad p \neq p_{\mathrm{c}}, t \to \infty, \tag{10a}$$

$$R_\infty^2(t) \propto \varepsilon^{\mu}t + \text{constant } \varepsilon^{-2\nu+\beta} + l(t), \qquad p > p_{\mathrm{c}}, t \to \infty, \tag{10b}$$

$$\langle(\Delta\boldsymbol{r}(t))^2\rangle \propto t^{2/(2+\theta)}, \qquad p = p_{\mathrm{c}}, t \to \infty. \tag{10c}$$

Here μ is the critical exponent of the diffusion constant, and $\bar{z} = (1/\nu)\ (2\nu - \beta + \mu)$, and therefore from (9) and (10*c*) $2/(2+\theta) = (2\nu-\beta)/(2\nu-\beta+\mu)$ (Gefen *et al* 1983). The origin of the function $l(t)$ in (10*b*) can be understood if one considers the exact relation

$$\langle(\Delta\boldsymbol{r}(t))^2\rangle = 2d\int_0^t \mathrm{d}\tau\,\{t-\tau\}K(\tau), \tag{11}$$

$K(t)$ being the time dependent diffusion coefficient. In the low scatterer density limit of the Lorentz model it is known that $K(t) \propto t^{-(d/2+1)}$ for $t \to \infty$ (Ernst and Weyland 1971). Accepting universality arguments, analogous to the case of liquids (Forster *et al* 1977), one expects the same behaviour for all $p_{\mathrm{c}} < p < 1$. Then $l(t) \propto \ln(t)$ in $d = 2$ and $l(t \to \infty) = 0$ for $d \geq 3$. In hopping models of diffusion the same long time tails are predicted (Odagaki and Lax 1981, Haus *et al* 1983). For $d \geq 3$ one can therefore define a diverging length r_0 by means of

$$r_0^2 = -\int_0^\infty \mathrm{d}\tau\,\tau K(\tau) \tag{12}$$

even for $p > p_{\mathrm{c}}$, which is the analogue of the localisation length introduced for $p < p_{\mathrm{c}}$ (Götze 1978).

For $p < p_{\mathrm{c}}$ (10*a*) is a rederivation of a known result (Stauffer 1979), where here only a scaling assumption for the pair connectedness (2) was used. As can be seen from (10), $r_0^2 \propto \varepsilon^{-2\nu+\beta}$ for $p > p_{\mathrm{c}}$ too. r_0 consists of two parts: the finite cluster contributions (10*a*) are similar to the $p < p_{\mathrm{c}}$ case (Kertész 1983), and an infinite cluster contribution comes from the fact that the coherence length ξ is present in the structure of the infinite cluster (Stanley and Coniglio 1983).

The quantity r_0^2 is measurable for instance via the static polarisability χ

$$\chi = r_0^2 = \lim_{\omega\to\infty} \mathrm{Re}\, K(\omega + \mathrm{i}0)/\omega. \tag{13}$$

In two dimensions it is physically clear that a diverging length analogous to r_0^2 must exist (see (5) and (10*a*)). However, long time tail effects are so prominent that it cannot be expressed by (12) and (13) and the static polarisability becomes infinite for $p_{\mathrm{c}} < p < 1$.

In conclusion, we have shown that the density relaxation function is related to the pair connectedness and consequently a scaling assumption is plausible near p_c. The probability of finding the particle at the site where it has started from for $t \to \infty$ could be identified with the generating function of percolation theory. Due to finite clusters there is even in the conducting phase $(p > p_c)$ a non-ergodic singularity, whence a Green–Kubo identity cannot hold. The prefactor of this $\delta(\omega)$ contribution was shown to exhibit a non-analyticity $\propto |\varepsilon|^{\beta}$ at p_c. Using the scaling assumption for Φ we discussed the analogue of the localisation length for $p > p_c$.

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