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ADDENDUM

Diffusion and trapping of excitations in disordered systems

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Abstract. We investigate diffusion and trapping of excitations in percolation systems by an effective medium approximation. We calculate $P_0(t)$, the probability of being at the origin at time t, on the percolation cluster at the percolation threshold p_c, $S_N(t)$, the mean number of distinct sites visited after time t, at and above p_c and $N(t)$, the survival probability after time t. All of our findings are completely consistent with the previously proposed relations by Rammal and Toulouse. We also consider a more general problem in which the waiting time distribution of an associated continuous-time random walk is long-tailed and propose generalisations of the Rammal–Toulouse conjectures. Our results indicate that the scaling properties of these quantities are sensitive to the details of the system.

Transport processes in disordered systems have attracted much attention in recent years. For example, conduction processes in amorphous materials have been much studied (see e.g. Pfister and Scher 1978 and references therein). In such processes, basic quantities such as AC and DC conductivities are influenced by the nature of the disordered system in which transport processes take place. Most of the theoretical studies have concentrated on models based on regular lattices. However, there has recently been a good deal of interest in fractal structures, chiefly due to their scale-invariance property in contrast to translationally invariant systems such as Bravais lattices. The largest percolation cluster at the percolation threshold p_c, linear and branched polymers and epoxy resins (Alexander *et al* 1983) are but a few examples. Thus fractal structures are expected to fill the gap between periodic structures and completely disordered systems. Therefore it is of considerable interest to study dynamical structures on fractals.

Rammal and Toulouse (1983) recently studied many physical problems on fractal structures. In particular, they studied random walks on such structures and proposed several relations concerning the statistics of random walks. The probability $P_0(t)$ that a random walker will find itself at the origin of the random walk at time t was proposed to be given by (see also Alexander and Orbach 1982)

$$P_0(t) \sim t^{-\frac{1}{2}d_s}, \tag{1}$$

where d_s is the fracton or spectral dimension of the fractal (Alexander and Orbach 1982). The spectral dimension is given by $d_s = 2d_f/d_w$, where d_f is the fractal dimension of the fractal and d_w is the fractal dimension of the random walk on the fractal.

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Another quantity of interest is the mean number of distinct sites visited, $S_N(t)$ at time t (after N steps) which, for a fractal, was conjectured to be given by (Rammal and Toulouse 1983)

$$S_N(t) \sim t^{\frac{1}{2}d_s}. \tag{2}$$

Numerical simulations of random walks on fractals appear to confirm these relations; see Blumen *et al* (1983) and Angles d'Auriac *et al* (1983). Scaling concepts have also been used to support equations (1) and (2) (Pandey and Stauffer 1983, Webman 1984). If a fraction c of sites of the fractal are traps, one is also interested in quantities such as $N(t)$, the probability that a random walker is not yet trapped at time t.

The most prominent and physically appealing fractal system is perhaps the largest percolation cluster at p_c. In this paper we study random walks on the percolation clusters at p_c by an effective medium approximation (EMA). We calculate quantities such as $P_0(t)$ and $S_N(t)$ by the EMA and find that the EMA predictions are consistent with equations (1) and (2) and thus provide relatively firm theoretical support for equations (1) and (2). The governing equation for the random walks is the linear master equation

$$\partial P_i(t)/\partial t = \sum_{j\in\{i\}} W_{ij}[P_j(t) - P_i(t)], \tag{3}$$

where $P_i(t)$ is the probability of being at site i at time t and W_{ij} the transition rate between sites i and j; $\{i\}$ denotes the set of nearest-neighbour sites of i. An EMA for random walks governed by the linear master equation (3) has been developed by several authors (Webman 1981, Haus *et al* 1982, Sahimi *et al* 1983). We follow these authors and develop an EMA which enables us to calculate the quantities of interest.

In the simplest EMA (the single-bond EMA) one replaces the random transition rates of all bonds but one with an effective transition rate W_m. It turns out that it is more convenient to work in the Laplace transform space. The random transition rate of the single bond causes a perturbation in the probability gradient across the single bond. One insists that the average value of this perturbation vanishes, the average being taken with respect to the single-bond transition rate probability density function $f(W)$. The governing equation for W_m is given by

$$\int_0^\infty \frac{f(W)\,\mathrm{d}W}{1-\gamma(\varepsilon)(W/W_m - 1)} = 1, \tag{4}$$

where λ is the Laplace transform variable conjugate to t and $\varepsilon = \lambda/W_m(\lambda)$. Here γ is a Green function given by $\gamma = 2/Z - \lambda G(\lambda)/W_m$, where Z is the coordination number of the lattice. We restrict our attention to a d-dimensional simple cubic lattice for which

$$G(\lambda) = -\frac{1}{2}\frac{1}{(2\pi)^d}\int_{-\pi}^{\pi}\cdots\int \frac{\mathrm{d}\theta_1\ldots\mathrm{d}\theta_d}{d+\frac{1}{2}\varepsilon - (\cos\theta_1 + \cdots + \cos\theta_d)}. \tag{5}$$

We first consider the simplest case in which the random transition rates have the probability density

$$f(W) = (1-p)\delta_+(W) + p\delta(W-1). \tag{6}$$

Near the percolation threshold p_c one has $\varepsilon = \lambda/W_m \ll 1$. We thus expand $G(\lambda)$ in powers of ε; we obtain

$$G(\lambda) = -\tfrac{1}{2}I_W(d) + (A_1/\lambda)(\lambda/W_m(\lambda))^{\frac{1}{2}d} + \ldots, \tag{7}$$

where A_1 is a constant given by

$$A_1 = \operatorname{cosec} \tfrac{1}{2}\pi(d-2)/[2^d \Gamma(d/2)\pi^{d/2-1}], \tag{8}$$

and $I_W(d)$ is a generalised Watson integral

$$I_W(d) = \frac{1}{(2\pi)^d} \int_{-\pi}^{\pi} \cdots \int \frac{d\theta_1 \ldots d\theta_d}{d - (\cos\theta_1 + \ldots + \cos\theta_d)}. \tag{9}$$

Equation (7) is valid for $2<d<4$. We now substitute equations (6) and (7) into (4) to find $W_m(\lambda)$. The result is given by

$$W_m(\lambda) = [d/2(d-1)]\{p - p_c + [(p-p_c)^2 + 2\lambda(d-1)I_W(d)/d^2]^{1/2}\} + \ldots. \tag{10}$$

The Laplace transform $\hat{P}_0(\lambda)$ of $P_0(t)$ is given by $\hat{P}_0(\lambda) = -G(\lambda)/W_m$. We thus obtain

$$\hat{P}_0(\lambda) = \frac{I_W(d)}{2W_m}\left[1 + \frac{A_1}{d}\left(\frac{\lambda}{W_m}\right)^{d/2} [(p-p_c)^2 + \lambda I_W(d)/2dW_m]^{-1/2}\right] + \ldots. \tag{11}$$

Therefore at $p = p_c$ and to leading order in λ we have $\hat{P}_0(\lambda) \sim \lambda^{-1/2}$, which means that

$$P_0(t) \sim t^{-1/2}, \qquad 2<d<4. \tag{12}$$

By a similar method we find that

$$G(\lambda) = -A_2(\lambda/W_m)^{(d/2)-1}, \qquad 1<d<2, \tag{13}$$

where A_2 is a constant. By following the same line of derivation as above we find that $\hat{P}_0(\lambda) \sim \lambda^{-2/(d+2)}$, which means that

$$P_0(t) \sim t^{-d/(d+2)}, \qquad 1<d<2. \tag{14}$$

We now calculate the mean number of distinct sites visited $S_N(t)$ at time t. For a translationally invariant lattice $\hat{S}_N(\lambda)$, the Laplace transform of $S_N(t)$, is related to $\hat{P}_0(\lambda)$ by (Montroll and Weiss 1965)

$$\hat{S}_N(\lambda) = [\lambda^2 \hat{P}_0(\lambda)]^{-1}. \tag{15}$$

The largest percolation cluster at p_c is a fractal object and lacks translational invariance. In this case equation (15) does not hold for large λ (short times). Hughes *et al* (1983) have derived a general expression for $\hat{S}_N(\lambda)$ for such a case (see their equation (21)). However, we may expect that as $t \to \infty$ the probability of being at the origin at time t will no longer depend on the origin of the walk, in which case equation (15) should hold as $\lambda \to 0$ ($t \to \infty$). We thus obtain

$$S_N(t) \sim \begin{cases} t^{d/(d+2)}, & 1<d<2, \quad (16) \\ t^{1/2}, & 2<d<4. \quad (17) \end{cases}$$

Above the percolation threshold p_c one has to distinguish between two regimes. At short times, i.e. when the span of the walk $R_s \equiv (\langle R^2(t)\rangle)^{1/2}$ is smaller than the percolation correlation length ξ_p, where $\langle R^2(t)\rangle$ is the mean-squared displacement of the walk, the random walk is not diffusive and is characterised by the fractal dimension d_w. In this regime we expect that equations (12), (14), (16) and (17) hold. However, at much longer times when $R_s \gg \xi_p$, the random walk is diffusive and we recover the results for periodic lattices, e.g. $P_0(t) \sim at^{-d/2}$ at all d and $S_N(t) \sim bt$ for $d \geqslant 3$. A characteristic time τ can be defined such that for $t \gg \tau$ the random walk is diffusive, while for $t \ll \tau$ the random walk is characterised by the fractal dimension d_w. It can be shown that the EMA predicts that

$$\tau \sim \begin{cases} (p-p_c)^{-(1+2/d)}, & 1<d<2, \quad (18) \\ (p-p_c)^{-2}\ln(p-p_c), & d=2, \quad (19) \\ (p-p_c)^{-2}, & d>2. \quad (20) \end{cases}$$

The prefactors a and b do depend on p. For example, it is straightforward to show that for $t \gg \tau$ and $p > p_c$ the EMA yields

$$S_N(t) \sim (p-p_c)t, \qquad 2<d<4. \tag{21}$$

The spectral dimension d_s of percolation networks has recently been calculated by an EMA. This was done by calculating the density of states $N(\omega)$ for percolation networks at p_c. One should have $N(\omega) \sim \omega^{d_s-1}$, where ω is the frequency. For $1<d<2$ one has (Derrida *et al* 1984)

$$d_s = 2d/(d+2), \qquad 1<d<2, \tag{22}$$

whereas for $2<d<4$ one obtains (Sahimi 1984, Derrida *et al* 1984)

$$d_s = 1, \qquad 2<d<4. \tag{23}$$

Equations (12), (14), (16) and (17) together with (22) and (23) are completely consistent with equations (1) and (2) that have been proposed by Rammal and Toulouse (1983) for random walks on fractals. For $p > p_c$ and $t \gg \tau$, Webman (1984) has suggested that

$$S_N(t) \sim \tau^{\frac{1}{2}d_s-1}t, \qquad d \geqslant 3, \tag{24}$$

which is again consistent with our result, equation (21). On the other hand our results can also be interpreted as an EMA prediction of d_s if one accepts the Rammal–Toulouse relations.

If a concentration c of the sites of a lattice are trapping sites, then the probability $N(t)$ that a random walker is not yet trapped at time t is given by (Weiss and Rubin 1983)

$$N(t) \sim 1 - cS_N(t) \simeq \exp[-cS_N(t)], \tag{25}$$

where it has been assumed that $cS_N(t) \ll 1$. Equation (25) together with (16) and (17) provide means of calculating $N(t)$ by an EMA. In particular, for three-dimensional systems the EMA predicts that $N(t) \sim \exp(-B_1 t^{1/2})$ where B_1 is a constant. Klafter *et al* (1984) have shown that for a fractal structure

$$N(t) \sim \exp(-B_2 t^{\alpha}), \tag{26}$$

where $\alpha = d_s/(d_s+2)$ and B_2 is a constant. For regular lattices one may use (26) but with (Kayser and Hubbard 1983) $\alpha = d/(d+2)$ and another constant B_3. Equation (26) predicts that $N(t) \sim \exp(-B_2 t^{0.4})$, if we take $d_s \simeq \frac{4}{3}$ for percolation clusters. For three-dimensional regular lattices one obtains $N(t) \sim \exp(-B_3 t^{0.6})$. The EMA prediction lies just in between these two results which is a quite satisfactory result. Moreover, Klafter *et al* (1984) have shown, by numerical simulation, that the exponent $\alpha = d_s/(d_s+2)$ cannot be observed experimentally and one should observe larger values of α if $d_s \neq 1$. These observations indicate that the EMA should provide a highly accurate expression for $N(t)$ at $d=3$. We remark that equations (1) and (2) have been proposed for random walks that take place on the largest percolation cluster. Angles d'Auriac and Rammal (1983) have shown that if the random walk takes place on *all* clusters, the resulting equations for $P_0(t)$ and $S_N(t)$ will be different. However, the EMA cannot explicitly distinguish between clusters of different sizes and thus it cannot be used for this case.

Other quantities of interest can be calculated by the above technique. For example, it can be shown that $M_0(t)$, the mean number of visits to the origin after time t, is given by

$$M_0(t) \sim t^{1-\frac{1}{2}d_s}, \tag{27}$$

so that if we take $d_s \simeq \frac{4}{3}$ for percolation clusters, we find $M_0(t) \sim t^{1/3}$. This result is valid if the random walk takes place on the largest percolation cluster at p_c or any other fractal. If the random walk is performed over all clusters, equation (27) must be modified, a matter to be discussed elsewhere.

We now turn to a more general situation in which the distribution of transition rates is given by

$$f(W) = (1-p)\delta_+(W) + ph(W), \tag{28}$$

where $h(W)$ has no generalised function component at the origin. It can be shown that (Sahimi *et al* 1983) if

$$h_{-1} = \int_0^\infty \frac{1}{W} h(W)\,\mathrm{d}W \tag{29}$$

is finite, the predictions of the EMA do not change qualitatively. However, if $h_{-1} = \infty$, the critical exponents depend heavily on the behaviour of $h(W)$ as $W \to 0$. For example, if we take $h(W) \sim CW^{-\beta}$, as $W \to 0$ and $0 < \beta < 1$, the results will be completely different from the results obtained so far. By using the exact correspondence between master equations, generalised master equations (Klafter and Silbey 1980) and continuous-time random walks (CTRW) (Kenkre *et al* 1973) one can express the results in terms of $\psi(t)$, the waiting time distribution of the associated CTRW. For $d > 2$ the EMA predicts that $\psi(t)$ is given by

$$\psi(t) \sim t^{(2\beta-3)/(2-\beta)}, \tag{30}$$

which is a long-tailed distribution, i.e. the mean waiting time $\langle t \rangle$ is infinite. By using the same analysis as before we obtain the following result (within the EMA)

$$S_N(t) \sim t^{(1-\beta)/2(2-\beta)}, \qquad 2 < d < 4. \tag{31}$$

If we compare equations (17), (30) and (31), we see that if in general $\psi(t) \sim t^{-1-\delta}$, then

$$S_N(t) \sim t^{\delta d_s/2}, \tag{32}$$

where $0 < \delta < 1$. Equation (32) can be viewed as a generalisation of the conjecture of Rammal and Toulouse (1983). Equation (32) also means that $\langle R^2(t) \rangle$ is given by

$$\langle R^2(t) \rangle \sim t^{2\delta/d_w}, \tag{33}$$

so that the random walk process is sensitive to the parameter δ.

In summary, we have used the effective medium approximation to investigate diffusion and trapping of excitations on percolation clusters. We have calculated $P_0(t)$, the probability of being at the origin at time t, at the percolation threshold p_c, $S_N(t)$, the mean number of distinct sites visited after time t, both at and above p_c, and $N(t)$, the survival probability if a fraction c of sites are traps. Our results are completely consistent with the previously proposed relations by Rammal and Toulouse (1983).

We have also investigated a more general random walk problem in which the waiting time distribution of the associated continuous-time random walk is long-tailed. We have proposed a generalisation of the Rammal and Toulouse (1983) relations for this case.

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