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LETTER TO THE EDITOR

Diffusion of interacting particles on fractal aggregates

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Abstract. We study the effect of excluded volume interactions on diffusion in random diffusion-limited aggregates. Performing Monte Carlo simulations we calculate the mean square displacement $\langle r^2(t)\rangle$ of a tracer particle as a function of the concentration c of the diffusing particles. We find that the long time behaviour of $\langle r^2(t)\rangle$ can be described by $\langle r^2(t)\rangle \sim [(1-c)f_T(c)t]^{2/d_w}$ where d_w is independent of c and $f_T(c)$ describes correlations between consecutive jumps of the tracer particle. Close to $c=1$ we find that $\langle r^2(t)\rangle$ scales as $\langle r^2(t)\rangle = g(t/t_x)$ where $t_x = c^2/(1-c)^2$ and $g(x) \sim x^{1/2}$ for $x \ll 1$ and $g(x) \sim x^{2/d_w}$ for $x \gg 1$.

How are the laws of diffusion and transport on fractal lattices changed when particles interact with each other? This question has been of immense recent interest (see, e.g., Stanley and Ostrowsky 1985), especially since Laibowitz and Gefen (1984) found experimentally that the transport properties of real materials with a fractal structure cannot be explained simply in terms of random walk models where the walkers do not interact with each other.

In real systems, the interaction between diffusing particles may become very complicated. The interaction always consists of a short-range part due to the excluded volume of a particle and may also involve a long-range part, which in the case of charged particles is governed by the Coulomb interaction. Their influence on polarisation has been studied by Gefen and Halley (1984). The influence of the Coulomb interaction on the *diffusion properties* is difficult to investigate, even for Euclidean lattices (Bunde and Dieterich 1984). Therefore, we will concentrate on short-range interactions and will restrict ourselves to the simplest case, the hard-core interactions, where double occupancy of a given site is forbidden. While the influence of the hard-core interaction on the diffusion process has been investigated in detail in Euclidean lattices (see Kehr and Binder 1984, Bunde *et al* 1985, and references therein) the problem to our knowledge has not yet been studied for fractal geometries.

In this letter we present the first study on a fractal lattice. We have chosen diffusion-limited aggregates (DLA) (see, e.g., Witten and Sander 1983) for which the non-interacting limit has been extensively studied (Meakin and Stanley 1983). Using the Monte Carlo technique we have investigated how the mean square displacement $\langle r^2(t)\rangle$ of a tracer particle is changed by the hard-core interaction between the particles. We find the remarkable result that the dimensionality of the walk is not changed by the interaction and thus does not depend on the concentration c of the diffusing particles. In contrast, the amplitude of $\langle r^2(t)\rangle$ depends crucially on the interaction, since the hard-core interaction creates strong correlations between consecutive jumps

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of the tracer particle. Close to $c=1$ we find the intriguing result that $\langle r^2(t)\rangle = g(t/t_x)$ with a crossover time t_x which depends on the concentration in the same way it does in one-dimensional systems, i.e. $t_x = [(1-c)/c]^{-2}$ (Richards 1977).

For our numerical study we have generated large DLA clusters in the way described by Witten and Sander (1983). For high concentrations of diffusing particles, we expect that the boundary of the cluster might play a role. To allow for larger concentrations, we used the following method: we grow a cluster of a given number of sites (usually 5000) and we keep only that portion of it that fits into a box of given length (usually $L=100$) containing about 2000 cluster sites. Then we use mirror boundary conditions to replicate the box. Due to the fractal structure of the DLA, the conventional periodic boundary conditions are not useful since the end points at opposite boundary sites only very rarely will fit to each other. We have also studied diffusion on a single large cluster of 20 000 sites (kindly supplied by P Meakin). Our results were not affected, so we conclude that the mirror boundary conditions do not matter.

In order to simulate the diffusion, first a random configuration of particles with probability p, i.e. concentration c, has to be generated on the DLA. Next, a particle is selected at random and an attempt is made to move to a randomly selected adjacent site. If the adjacent site belongs to the cluster and is empty the particle moves, otherwise it stays. After each trial the time is increased by $1/N$, where N is the total number of particles, so that after one unit of time has passed, every particle (on average) had the possibility of moving once. We have measured the mean square displacement of the tracer particle for times up to 10^5 time steps, and averaged over up to 200 samples. For $c \to 0$ the hard-core interaction should not matter and we expect that $\langle r^2(t)\rangle$ becomes identical to $\langle r^2(t)\rangle$ for the non-interacting limit, which for large times is given by

$$\langle r^2(t)\rangle = (\alpha t)^{2/d_w} \tag{1}$$

where α is proportional to the jump frequency of the particle and d_w is the dimension of the walk. For DLA $d_w = 2.57 \pm 0.1$ (Meakin and Stanley 1983, Havlin 1984).

First we investigated if d_w is affected by the hard-core interaction. Figure 1 shows representative results of $\langle r^2(t)\rangle$ for four concentrations of diffusing particles, $c=0.2$, 0.5, 0.8 and 0.9. We see clearly that for large times the slopes ($=2/d_w$) are independent of c and approach the value $2/d_w = 0.78 \pm 0.03$. We therefore obtain the same value for the dimension of the walk, $d_w = 2.57 \pm 0.1$, found by Meakin and Stanley in the non-interacting limit. For comparison, in two- and three-dimensional Euclidean lattices, $d_w = 2$ and is not changed by switching on a hard-core interaction between the random walkers, while in one dimension the hard-core interaction changes d_w from 2 into 4 for all concentrations (Richards 1977).

In contrast to the exponent d_w the jump frequency α is strongly concentration dependent and decreases drastically as c tends to 1 (see figure 1). How can we understand this?

Obviously, by considering many hard-core particles, the average jump rate of the tracer particle is changed from α to $(1-c)\,\alpha$ since now the number of available sites for diffusion is diminished by the factor $c_v = 1-c$, which represents the concentration c_v of vacancies. On average, a particle has to wait $1/[(1-c)\alpha]$ time steps before it can make a jump. Therefore, in the spirit of a mean-field approach we might consider consecutive jumps of a tracer particle as being independent of each other. Then the mean square displacement would be

$$\langle r^2(t)\rangle_{\mathrm{MFA}} = (\alpha c_v t)^{2/d_w} \tag{2}$$

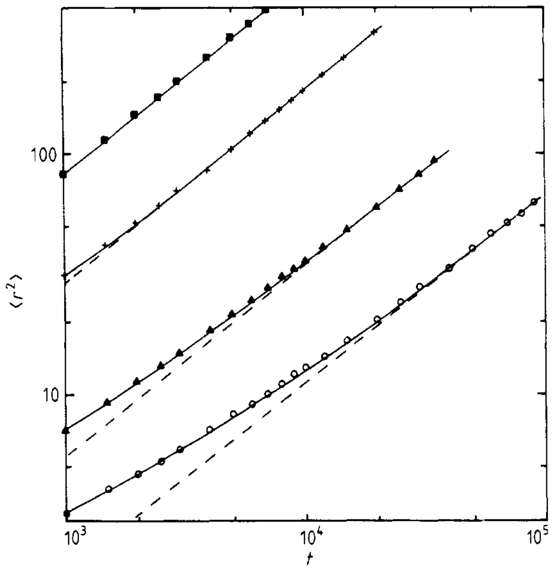

Figure 1. Mean square displacement $\langle r^2(t)\rangle$ against time t of a tracer particle for the concentrations $c = 0.2$ (■), 0.5 (+), 0.8 (▲) and 0.9 (○) of hard-core particles on the DLA. The broken lines describe the asymptotic behaviour of $\langle r^2(t)\rangle$.

for large t. However, consecutive jumps are not independent of each other, as can be most easily seen for $c_v \to 0$. In this limit, vacant sites are rare and the probability of a particle jumping back to a site it just left is much higher than that of its jumping to any other neighbouring site. Clearly this correlation diminishes $\langle r^2(t)\rangle$ further. Deviations of $\langle r^2(t)\rangle$ from $\langle r^2(t)\rangle_{MFA}$ are an indication of the presence of these dynamical correlations. In order to incorporate them, we introduce a tracer correlation factor $f_T(c)$ via

$$\langle r^2(t)\rangle = [\alpha(1-c)f_T(c)t]^{2/d_w} \tag{3}$$

which generalises the conventional definition for two- and three-dimensional Euclidean lattices (see, e.g., Kehr and Binder 1984) to fractals.

The result for $f_T(c)$ in the DLA fractal is shown in figure 2. For comparison we have also shown the exact results for $f_T(1)$ in the square lattice and in the simple cubic lattice (Nakazato and Kitahara 1980, Tahir-Kheli and Elliott 1983). By definition, $f_T(0) = 1$. Above $c = 0.1$, f_T shows a sharp decrease. For $c_v < 0.1$, $f_T(c)$ approaches zero as

$$f_T(c) \sim c_v. \tag{4}$$

This result shows that in the DLA cluster close to $c_v = 0$ consecutive jumps of the tracer particle are strongly correlated. These dynamical correlations lead to a breakdown of the mean-field approach close to the 'critical point' $c_v = 0$, reminiscent of the breakdown of the mean-field approach in critical phenomena close to the critical point. In two- and three-dimensional Euclidean lattices the dynamic correlations are considerably weaker and $f_T(c)$ stays non-zero even in the vicinity of $c_v = 0$. The reason is connected with the linear structure of the arms of the DLA.

Next we consider how the short time behaviour of $\langle r^2(t)\rangle$ depends on the concentration of the hard-core particles. Figure 3 shows $\langle r^2(t)\rangle$ for the same four concentrations

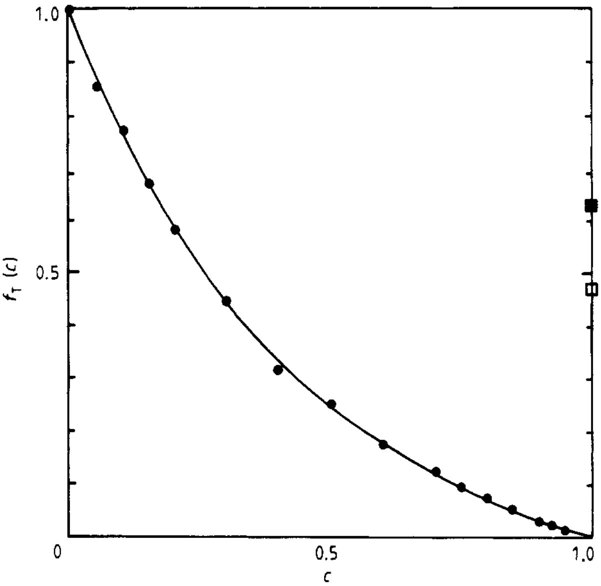

Figure 2. Tracer correlation factor $f_T(c)$ against concentration c of hard-core particles on the DLA. The sumbols (□) and (■) denote the values of $f_T(1)$ for the square lattice and the simple cubic lattice, respectively.

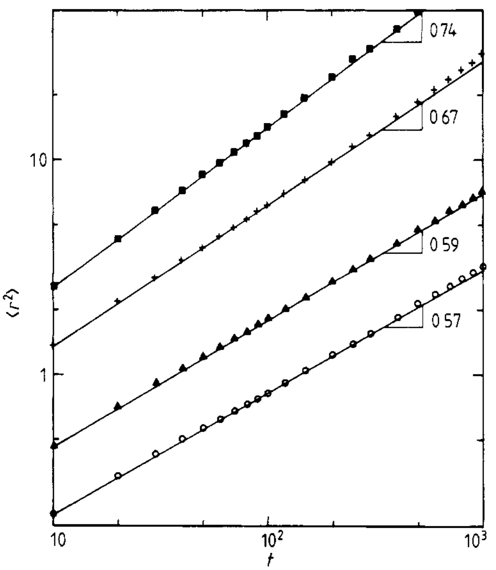

Figure 3. The same as figure 1, but for times $t \leq 1000$.

$c = 0.2$, 0.5, 0.8 and 0.9 as in figure 1 for time steps t between 10 and 1000. In the log-log plot, the four curves represent nearly straight lines but, unlike figure 1, the slope changes continuously. We could say that, at small time, the walk is described by an effective d_w, which increases gradually when c is increased. In figure 4 we have plotted this effective small time d_w^{eff} as a function of c. d_w^{eff} has the value of independent

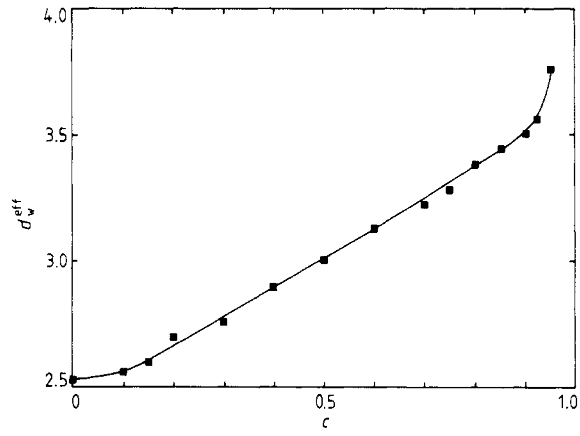

Figure 4. Effective short time dimensionality of the walk, d_w, as a function of concentration.

particles for $c<0.1$ and then increases gradually. For $c_v \rightarrow 0$ d_w^{eff} approaches 4, the result for interacting particles on a one-dimensional chain.

This can be understood as follows. In the limit of $c_v \rightarrow 0$ only a few vacancies are available and therefore the diffusion is very slow. It takes a particle a long time to leave a branch of the DLA and to discover the fractal structure of the cluster. Therefore, at short times the motion of the tracer should be governed by the one-dimensional character of the DLA branches. Hence, in the limit of $c_v \rightarrow 0$ we expect $d_w^{eff}=4$ at short times, which explains our result.

As we know from figure 1, d_w tends asymptotically to its value for the non-interacting limit. The crossover time t_x increases strongly when c increases (see figure 1). For $t \ll t_x$ the motion of the tracer is governed by the one-dimensional branches of the DLA. For one-dimensional systems we know (Richards 1977) that, in the limit of $c \rightarrow 1$,

$$\begin{aligned}\langle r^2(t)\rangle &\sim \{[(1-c)/c]^2 t\}^{1/2} \\ &= [(c_v/c)^2 t]^{1/2}.\end{aligned} \tag{5a}$$

Equivalently

$$\langle r^2(t)\rangle \sim (t/t_0)^{1/2} \tag{5b}$$

where

$$t_0 = (c/c_v)^2 \tag{5c}$$

determines the timescale of the one-dimensional diffusion. We assume now that in the limit of high concentration the mean square displacement of interacting particles in DLA also depends only on the ratio $t/t_0(c)$, i.e. $t_x \sim t_0$:

$$\langle r^2(t)\rangle = g(t/t_x(c)) \tag{6}$$

where $g(x)$ is more general than (5b). For small times, the diffusing particles feel only single topologically one-dimensional branches, so we expect

$$g(x) \sim x^{1/2} \qquad (x \ll 1). \tag{7a}$$

For large times, the particles feel the fractal and we expect

$$g(x) \sim x^{2/d_w} \qquad (x \gg 1) \tag{7b}$$

in analogy with the zero concentration limit of (2).

We have checked this scaling ansatz for various concentrations in the vicinity of $c = 1$ (see figure 5) and the result confirms the scaling.

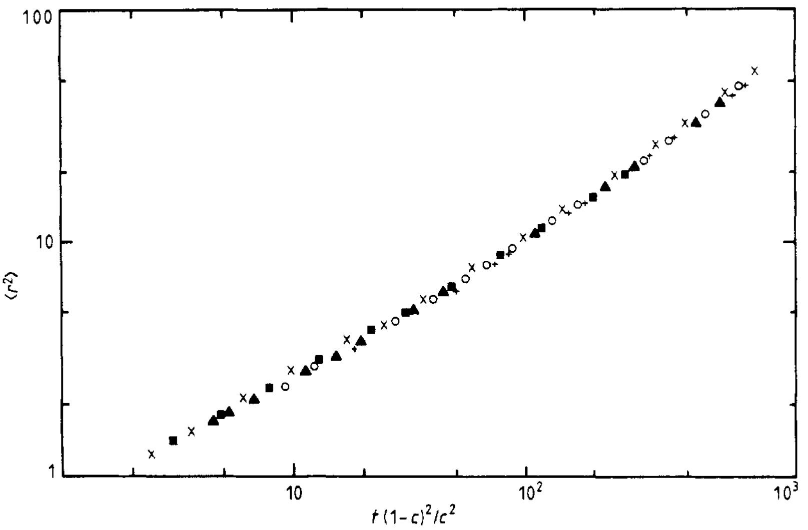

Figure 5. $\langle r^2(t) \rangle$ against $t/t_x \equiv t(1-c)^2/c^2$ for various concentrations of hard-core particles close to $c = 1$: $c = 0.8$ (+), 0.85 (○), 0.9 (×), 0.92 (▲) and 0.95 (■) of concentration of hard-core particles on DLA.

When we compare (7*a*), (7*b*) and (5*c*) with (3) we get a more accurate description of $f_T(c)$ close to $c = 1$. We find

$$f_T(c) \sim c_v/c^2 \qquad (c_v \to 0). \tag{8}$$

In conclusion we have studied how short-range hard-core interactions affect diffusion in DLA clusters. We have considered the mean square displacement of a tracer particle and have found that the dimension of the walk is not affected by the interaction, but the interaction induces strong correlations between consecutive jumps of the tracer, which drastically change the amplitude of $\langle r^2(t) \rangle$. Close to $c_v = 0$, i.e. $c = 1$, we have found that $\langle r^2(t) \rangle$ shows scaling behaviour with a crossover time that is typical for one-dimensional systems.

In a separate work (Amitrano *et al* 1985), we have also studied diffusion of hard-core particles on the incipient percolation cluster in $d = 2$. We have found that in this case also the interaction did not affect the dimension of the walk. After submitting this letter we learnt that this observation has also been confirmed by Heupel (1985) for percolation in $d = 3$.

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References

Amitrano C, Bunde A and Stanley H E 1985 *Preprint* Boston University

Bunde A and Dieterich W 1984 *Phys. Rev.* B **31** 6012

Bunde A, Havlin S, Nossal R and Stanley H E 1985 *Phys. Rev.* B **32** 3374

Gefen Y and Halley J F 1984 in *Kinetics of Aggregation and Gelation* ed F Family and D P Landau (Amsterdam: North-Holland) p 161

Havlin S 1984 in *Kinetics of Aggregation and Gelation* ed F Family and D P Landau (Amsterdam: North-Holland) p 145

Heupel L 1985 *Preprint*

Kehr K W and Binder K 1984 *Applications of the Monte Carlo Methods in Statistical Physics* ed K Binder (Berlin: Springer)

Laibowitz R B and Gefen Y 1984 *Phys. Rev. Lett.* **53** 380

Meakin P and Stanley H E 1983 *Phys. Rev. Lett.* **51** 1457

Nakazato K and Kitahara K 1980 *Prog. Theor. Phys.* **64** 2261

Richards P M 1977 *Phys. Rev.* B **16** 1363

Stanley H E and Ostrowsky M 1985 *On Growth and Form: Fractal and Non-fractal Patterns in Physics*, (Dordrecht: Martinus Nijhoff)

Tahir-Kheli R A and Elliott R J 1983 *Phys. Rev.* B **27** 884

Witten T A and Sander L M 1983 *Phys. Rev.* B **27** 5686