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LETTER TO THE EDITOR

# Novel dimension-independent behaviour for diffusive annihilation on percolation fractals

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**Abstract.** We report the first studies of diffusive annihilation on fractal structures. We find super-universal ($d$-independent) behaviour for the time decay of the particle density; specifically, for the reaction $A+A\rightarrow 0$ we find $\rho_A \sim t^{-2/3}$, while for the reaction $A+B\rightarrow 0$ we find $\rho_A \sim t^{-1/3}$. A scaling theory for diffusive annihilation is developed that predicts that the first exponent is $\frac{1}{2}d_s$ and the second is $\frac{1}{4}d_s$, where $d_s = 2d_f/d_w$ is the spectral dimension. Thus our findings support the Alexander–Orbach conjecture that $d_s$ is independent of $d$.

A classic problem in chemical kinetics is the effects of diffusion rates on chemical kinetics (see, e.g., the classic review of Noyes 1961). In diffusion-controlled reactions, the reaction rate is determined by the slow diffusive motion required for the reacting species to reach each other. The reaction itself is considered to be instantaneous and irreversible. The classic Smoluchowski theory of such diffusion-controlled reactions has been used to describe the growth of colloidal or aerosol particles, a subject of considerable current interest (the review Chandrasekhar 1943 contains a short exposition of the Smoluchowski theory of coagulation of colloids).

Initial work on this topic focussed on the dilute limit (Montroll 1946) and many treatments assume immobile sinks (Felderhof and Deutch 1976). Recently interest arose in the general case of *mobile* reactants in which all particles diffuse randomly with the same diffusion constant $D$. When two particles collide, they are assumed to produce an inert species irreversibly. Torney and McConnell (1983a,b) recently obtained exact results for the reaction

$$A+A\rightarrow 0, \tag{1a}$$

for one dimension (here O denotes the inert species), while Toussaint and Wilczek (1983) developed a general theory for $d$ dimensions for the 'one-kind' annihilation (1$a$) as well as for the 'two-kind' annihilation

$$A+B\rightarrow 0. \tag{1b}$$

Specifically, Toussaint and Wilczek find that for a Euclidean lattice, the density of surviving A particles decays slowly in time with a power-law tail; for the one-kind

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annihilation (1*a*),

$$\rho_A \sim t^{-d/2} \qquad (d < d_c = 2), \tag{2a}$$

and for the two-kind annihilation (1*b*),

$$\rho_A \sim t^{-d/4} \qquad (d < d_c = 4). \tag{2b}$$

The predictions (2*a*) and (2*b*) were confirmed by computer simulations for $d = 1, 2$ (Toussaint and Wilczek 1983) and we have extended their work to $d = 3$ (figure 1) as well as repeating the $d = 1$ and $d = 2$ calculations for large lattice sizes.

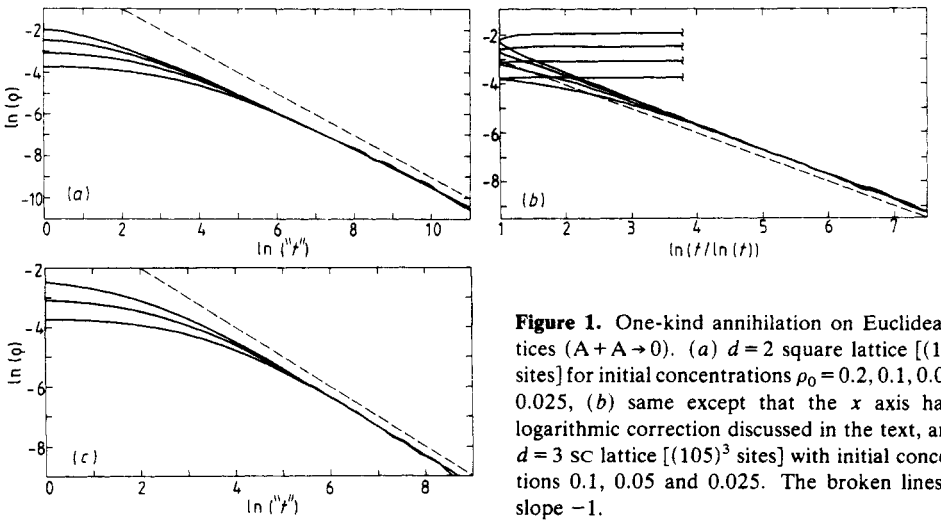


**Figure 1.** One-kind annihilation on Euclidean lattices (A + A → 0). (*a*) $d = 2$ square lattice [$(1024)^2$ sites] for initial concentrations $\rho_0 = 0.2, 0.1, 0.05$ and 0.025, (*b*) same except that the $x$ axis has the logarithmic correction discussed in the text, and (c) $d = 3$ SC lattice [$(105)^3$ sites] with initial concentrations 0.1, 0.05 and 0.025. The broken lines have slope −1.

The main purpose of this work is to generalise the concept of diffusion-controlled annihilation to self-similar 'fractal' structures. To simulate diffusive annihilation on 1*d*, 2*d* or 3*d* Euclidean lattices, sites on the lattice are picked at random and 'occupied' with 'particles' (avoiding multiple occupancy) until a desired initial particle concentration has been reached. In the case where two kinds of particles (A and B) are present, equal numbers of A and B particles are deleted (first one A particle and then one B particle, etc.). To simulate the diffusive annihilation process, particles are selected at random and moved to a randomly selected nearest-neighbour site. In the case of the A + A → 0 reaction both particles are removed if they come to occupy the same lattice site. After each move the 'time' is incremented by $1/N$, where $N$ is the number of surviving particles in the system. In the A + B → 0 case A and B particles are removed if they both occupy the same lattice site. If a randomly selected move would cause a lattice site to be occupied by more than one A particle or more than one B particle the move is rejected and a new particle is picked at random. Periodic boundary conditions were used in all of our simulations.

To simulate diffusive annihilation in a percolation cluster a pseudo random ($x$) number evenly distributed between 0 and 1 ($0 < x < 1$) is generated for each lattice site and that site is occupied if $x$ is less than the percolation threshold probability ($p_c$) which we assumed to have a value of 0.5927 for $d = 2$ (Gebele 1984) and 0.3117 for $d = 3$ (Stauffer 1979). The largest cluster of sites joined via nearest-neighbour

occupancy is then formed (sites joined across the periodic boundaries are considered to belong to the same cluster) and our annihilation simulations are carried out using the largest percolation cluster as the substrate. Now particles are picked at random and an attempt is made to move them to a nearest-neighbour site. The move is rejected if the randomly chosen nearest-neighbour site is not on the percolation cluster and a new particle is picked at random. In the case of diffusive annihilation on percolation clusters the particle concentrations are measured with respect to the total number of sites in the percolation cluster.

Our $d=1$ simulations were carried out on a lattice $10^5$ sites long. The $d=2$ stimulations were carried out using $512\times512$ and $1024\times1024$ lattices, while the $d=3$ simulations were for $105\times105\times105$ lattices. In the case of diffusive annihilation on percolation clusters the percolation clusters were generated on lattices of these sizes also.

We next obtain the analogs of $(2a)$ and $(2b)$ for an *arbitrary* fractal structure and then we describe the results of our computer simulations designed to test the predictions. The most straightforward approach is dimensional analysis or 'scaling'. We begin with the one-kind process, $A+A\rightarrow0$. A fractal is characterised by a volume V that scales asymptotically with the characteristic radius $\xi$ as $V\sim\xi^{d_f}$, where $d_f$ is termed the fractal dimension. Hence one expects that the density of A particles should scale as

$$\rho_A\sim\xi^{-d_f}. \tag{3a}$$

The particles move about on the fractal by a random walk, and the characteristic range of the random walk scales with the time as

$$\xi\sim t^{1/d_w}, \tag{3b}$$

where $d_w$ is termed the fractal dimension of the random walk; for most fractals, $d_w$ is a strong function of $d_f$. Substituting $(3b)$ into $(3a)$, we find that $\rho_A$ scales with time according to

$$\rho_A\sim t^{-d_f/d_w} \qquad \text{[one-kind process]}. \tag{4}$$

For a Euclidean lattice, $d_f=d$ and $d_w=2$ for all dimensions, so that (4) reduces to $(2a)$.

Simulations for $d=2$ and 3 Euclidean lattices are shown in figure 1. From figures $1(a)$ and $1(b)$, we see that the $d=2$ data are more consistent with $(2a)$ if one takes into account a logarithmic correction (since $d_c=2$ for random walks)†. Data on $d=2$ and 3 percolation fractals are shown in figure 2, and we see that the results are consistent with a 'superuniversal' ($d$-independent) value of the spectral dimension $d_s$,

$$d_s=2d_f/d_w=\tfrac{4}{3}. \tag{5}$$

The result (5) was first conjectured by Alexander and Orbach (1982), and later supported by heuristic arguments (Rammal and Toulouse 1983, Leyvraz and Stanley 1938) and numerical calculations for the de Gennes ant on percolation fractals (Pandey

† An independent way of understanding (4) is to recognise that $\rho$ is just inversely proportional to the number of visited sites for a random walk, and this quantity scales as in (4) (see, e.g., Stanley *et al* 1983 and references therein). Hence a logarithmic correction for $d=2$ is expected. Since the number of visited sites in $d=2$ increases with time as $t/\ln t$ (see, e.g., Rammal and Toulouse 1983), we anticipate that the data for $d=2$ may be better fitted by replacing $t$ by $t/\ln t$; this is confirmed by figure $1(b)$. However for the $A+B\rightarrow O$ case we find that this simple replacement does not improve the fit and at present we have no explanation for this fact. Note that for random walks on percolation fractals there is *no* logarithmic correction, and our data also does not support one (Rammal and Toulouse 1983).

and Stauffer 1983, Havlin and Ben-Avraham 1983), lattice animals (Wilke *et al* 1984, Sahimi and Jerauld 1984) and Witten–Sander clusters (Meakin and Stanley 1983).

The argument for the two-kind annihilation $A+B\rightarrow O$ proceeds in the same fashion, except that now an $A$ particle can annihilate only if it finds a B particle. Hence the interaction varies as the 'square' of the density and (4) is replaced by

$$\rho_A \sim t^{-d_f/2d_w} \qquad \text{[two-kind process]}. \tag{6}$$

For a Euclidean lattice, (6) reduces to ($2b$). Toussaint and Wilczek (1983) interpret the two-kind process in terms of the fluctuation in the number of particles in volume $V$; this is also the starting point in the recent scaling theory of Kang and Redner (1983), which has been used to treat diffusive annihilation on regular and fractal lattices. Simulations for Euclidean lattices with $d=2$ and 3 are shown in figure 3†, while the results for percolation fractals are given in figure 4. Again, the percolation, fractals are consistent with the superuniversal value of (5).

In summary, we have studied a lattice version of diffusion annihilation for the one-kind process $A+A\rightarrow 0$ and the two-kind process $A+B\rightarrow 0$. For Euclidean lattices with $d=2$ and 3, our results are consistent with the predictions ($2a$) and ($2b$) of

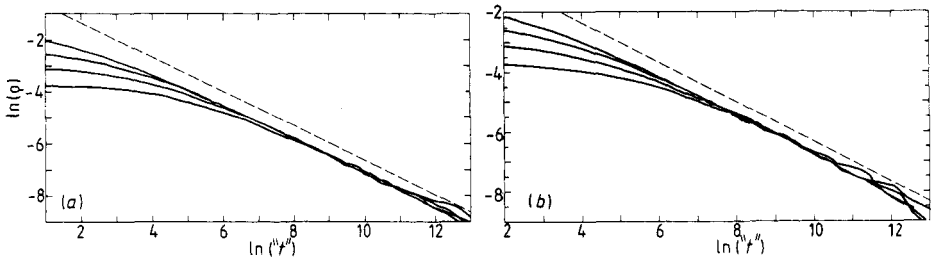


**Figure 2.** One-kind annihilation on percolation fractals ($A+A\rightarrow O$). ($a$) $d=2$ square lattice [$(1024)^2$ sites] at the percolation threshold $p_c=0.5927$, for initial concentrations 0.2, 0.1, 0.05 and 0.025. ($b$) $d=3$ SC lattice [$(105)^3$ sites] at the percolation threshold $p_c=0.3117$, for the same initial concentrations as in ($a$). Note that the limiting slope is independent of $d$ ('superuniversal'). The broken lines have slope $-\frac{2}{3}$.

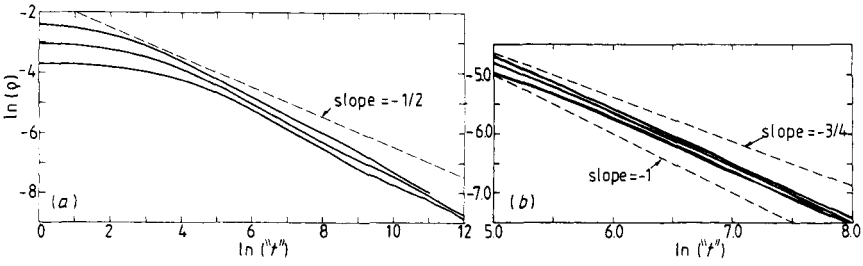


**Figure 3.** $A+B\rightarrow O$ annihilation on Euclidean lattices. ($a$) $d=2$ square lattice [$(1024)^2$ sites] for initial concentrations $\rho_0=0.1$, 0.05 and 0.025. ($b$) $d=3$ SC lattice [$(105)^3$ sites] for initial concentrations $\rho_0=1.0$, 0.05 and 0.025.

† Note that the data for $d=3$ agree as well with a line of slope 1 as with a line of slope $\frac{3}{4}$. Whether this discrepancy is due to an inadequacy of the theory (cf remarks in Toussaint and Wilczek 1983) or whether the discrepancy would vanish with longer simulations is unclear at present. Further work is underway on this point.

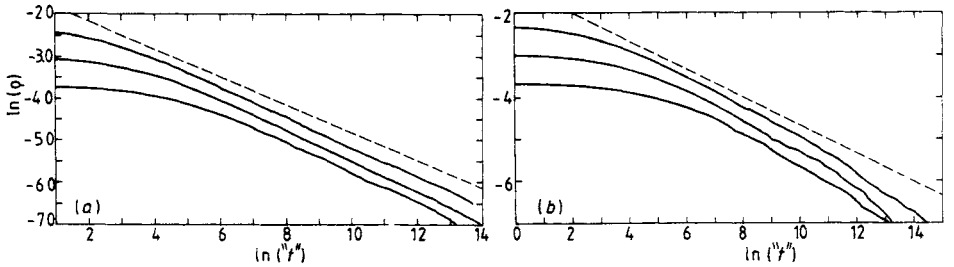


**Figure 4.** A + B → O annihilation on percolation fractals. (*a*) $d = 2$ square lattice [$(1024)^2$ sites] for initial concentrations $\rho_0 = 0.1$, 0.05 and 0.025 at the percolation threshold $p_c = 0.5927$. (*b*) $d = 3$ SC lattice [$(105)^3$ sites] for initial concentrations $\rho_0 = 0.1$, 0.05 and 0.02 at the percolation threshold $p_c = 0.3117$. The broken lines indicate the lines of slope $-\frac{1}{3}$.

Toussaint and Wilczek (1983). For percolation fractals, we find $d$-independent exponents, thereby supporting the Alexander–Orbach conjecture that the ratio $d_f/d_w$ is superuniversal.

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