The fractal nature of a diffusion front and the relation to percolation

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The fractal nature of a diffusion front and the relation to percolation

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Résumé. — On montre sur une simulation à deux dimensions qu'un front de diffusion possède une géométrie fractale sur une largeur dépendant de la longueur de diffusion. Le nombre de particules sur le front, et la largeur mesurant son étalement, suivent des lois de puissance en fonction de la longueur de diffusion. Les exposants de ces lois et la dimension fractale peuvent être écrits simplement en fonction des exposants critiques de la percolation à deux dimensions.

Abstract. — Using a two dimensional simulation, a diffusion front is shown to have a fractal geometry in a range increasing with the diffusion length. The number of particles on the front, and the width measuring its spread, follow power laws as a function of the diffusion length. The associated exponents and the fractal dimension can be expressed as simple functions of the critical exponents of the two dimensional percolation problem.

In the simplest picture, diffusion in solids is the consequence of Brownian motion of atoms on a lattice. Brownian motion is known to create fractal patterns [1, 2]. The purpose of this paper is to show, through a numerical simulation on a 2D lattice, that the diffusion front is a fractal object [3] with dimension $D_f = 1.76 \pm 0.02$. Moreover we put forward the idea that there is a close similarity between this line and the external frontier of a percolating cluster. These considerations may have practical importance since, for example, atomic diffusion plays a role in many ways in realizing electrical contacts : a good contact between two bodies can often be realized by heating the system so enabling diffusion to occur in a reasonably short time.

We consider the simple case of the diffusion of particles A on a 2D lattice B with the diffusion source being a line of A atoms kept at a constant concentration equal to unity. We restrict the analysis to nearest neighbour site hopping without double occupancy. Under these circumstances the concentration varies as a function of time and space as [4]

$$p(x) = \operatorname{erfc}(x/l_D) = 1 - (2/\pi^{1/2}) \int_0^{x/l_D} \mathrm{d}u \exp(-u^2) \tag{1}$$

where $l_D = 2(Dt)^{1/2}$ is the diffusion length at time t with diffusion coefficient D. The average

concentration is a function of time and space but in the following we will only consider the situations obtained at fixed successive times. Both square and triangular lattices will be investigated.

We define the microscopic physical interface in a simple manner which is illustrated by the following electrical picture. The diffusing A atoms are supposed to be metallic, and « conducting » only if they are nearest neighbours, the B lattice being otherwise insulating. In this picture if a potential is applied between materials A and B all the occupied sites which are connected with A are at the same electrical potential. We investigate the frontier of this region. In the case of a square lattice the frontier is made of those A atoms which are connected through nearest neighbours with the diffusion source and have an empty first or second neighbour which itself is connected through empty first or second neighbours to the region far in B where no A atom are present. This is shown in figure 1 where we see that occupied sites build a land connected to the diffusion source, and also islands. The empty sites create an « open sea » and lakes. The object we are looking at is the thick line in figure 1 : it is the « seashore ». Correlations between particles may play a role in the structure of this seashore. In this simplified model we neglect such effects : a given diffused state is simply obtained by a random distribution of particles obeying equation (1). This procedure provides for a considerable gain in computing time compared to the complete simulation of diffusion.

The simulation is made by choosing for each site in a row of atoms at constant x a random number between 0 and 1. If this random number is smaller than $p(x)$ as given by equation (1), we put an atom in that site. In this way we constitute one detailed diffusion picture on which we look for the seashore as defined above. Periodic boundary conditions are used to define this seashore. We name $p_f(x)$ the probability of finding one point of the frontier at x. It is the density of seashore at distance x. This quantity is obtained from the simulation calculation. It is found to behave very nearly as a Gaussian function of x. These density profiles are then averaged over 100 different diffusion pictures of lateral size $L = 512$ or $L = 1\ 024$ (see Fig. 2a)
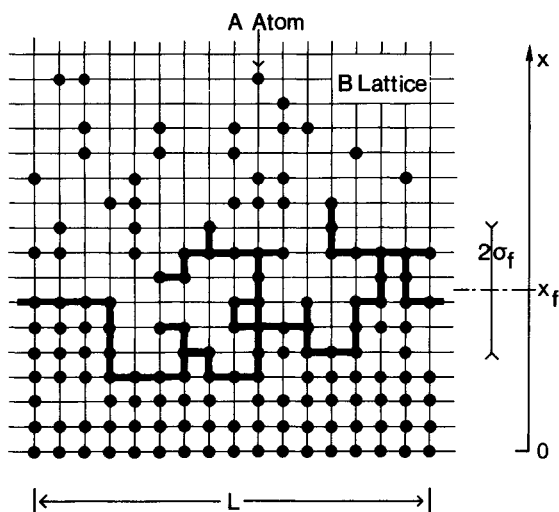

Fig. 1. — Schematic picture of the simulated diffused system at a given time. The source of A atoms at the bottom is kept at unit concentration. The concentration decreases with increasing x. The thick line represents the diffusion front that we are studying. Its mean position is x_f and it is spread on a width σ_f.
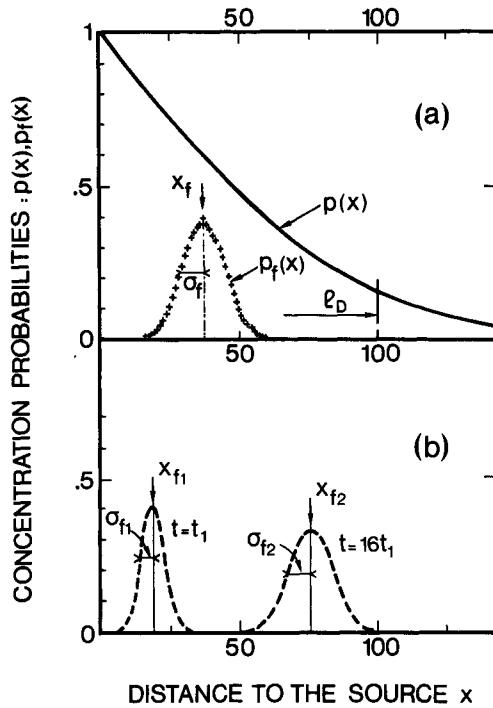
Fig. 2. — Statistical distribution of the simulated frontier defined in figure 1 in the case of a square lattice. (a) Comparison of the frontier density (crosses) with the overall atom density (continuous line) at a given time. (b) Frontiers for $l_{D,1} = 50$ and $l_{D,2} = 200$.

and we calculate the mean position x_f and the width σ_f using

$$x_f = \int_0^\infty x p_f(x)\, dx \Big/ \int_0^\infty p_f(x)\, dx \tag{2}$$

and

$$\sigma_f^2 = \int_0^\infty (x - x_f)^2\, p_f(x)\, dx \Big/ \int_0^\infty p_f(x)\, dx. \tag{3}$$

The results are qualitatively represented in figure 2b. One can see two different profiles of the frontier for two different times corresponding to $l_{D,1} = 50$ and $l_{D,2} = 200$. The mean position x_f varies like l_D. Figure 3 gives the quantitative results for the width σ_f and the number of points on the frontier N_f. We observe that these quantities follow power laws to within a very good approximation. This is discussed below.

The geometry of the frontier has been determined by the mass to radius relation [1] :

$$M \sim R^{D_f}. \tag{4}$$

In our case we take as the mass the number of sites of the frontier in a square of side R. Figure 4 shows the log-log plot in two cases with two very different values of the diffusion length. We observe that the fractal geometry exists only up to a scale of the order of σ_f. Because of the dependence of σ_f on l_D the fractal geometry exists on a scale which increases with the diffusion length.
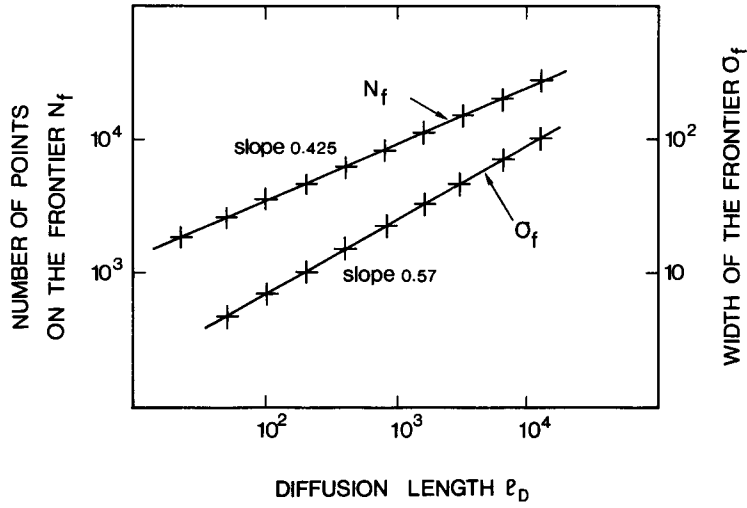
Fig. 3. — Variation of the number of points of the frontier N_f and of the width σ_f as a function of the diffusion length l_D. The above results for N_f have been obtained for, or rescaled to, a sample lateral size of $L = 512$ in the case of the square lattice.

Fig. 4. — Determination of the fractal dimension of the diffusion front from the mass-to-radius relation (square lattice). The diffusion front is a fractal only up to a scale of the order of the width σ_f. The value 1.62 quoted for $l_D = 50$ comes from a finite size effect which disappears for $l_D \geqslant 1\ 600$.

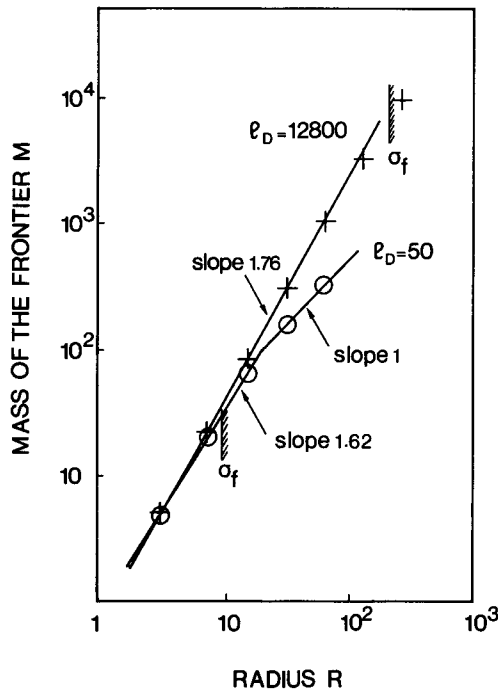The dimension is one when R is larger than a few σ_f. This only tells us that the length of the frontier is proportional to the lateral size L of the sample. (L is always taken larger than σ_f.) We obtain in the fractal region :

$$D_f = 1.76 \pm 0.02 . \tag{5}$$

We want now to show that the above physical situation is closely related to the usual percolation problem. The percolation problem is defined with a constant concentration probability over the lattice. One generally thinks that because percolation is a critical phenomenon the probability of occupation should be kept rigorously constant, otherwise the critical effects are expected to be smeared out. In fact this is not true and interesting new features appear in the present situation.

Consider the diffusion system defined above at a given time : it is a random distribution but with non-uniform probability. There is obviously an infinite cluster which links both lateral sides of the sample because the concentration near the source is close to unity. Consider now the geometry of this cluster and its penetration depth. Here again it is obvious that far from the source the infinite cluster has no branches because in these regions the concentration is too small. Hence the frontier of the infinite cluster should be in a region where the concentration is around the critical concentration p_c for the considered lattice. The frontier has a mean distance to the source x_f and thus we expect $p(x_f)$ to be near p_c. The result of the simulation is shown in table I for the two cases of the square and triangular lattices. A striking result is obtained : the simulated frontier is situated at p_c. More precisely it appears that the concentration at x_f tends asymptotically towards p_c in the infinite diffusion limit. But already for $l_D = 100$ the deviation is only 1 %, both for square and triangular lattices. For $l_D = 6\,400$ this deviation is as small as 0.2 %.

This explains a result previously mentioned : because x_f appears to correspond to a constant concentration it varies proportionally to the diffusion length l_D. This is a consequence of equation (1) which indicates that a given p is obtained for a constant value of x_f/l_D. Values of x_f/l_D are shown in table I for various choices of l_D for square and triangular lattices. If we postulate that

$$p(x_f) = p_c \tag{6}$$

we expect $x_f = 0.4769\, l_D$ on the triangular lattice where $p_c = 0.5$ and $x_f = 0.378\, l_D$ on the square lattice using $p_c = 0.593$ [5]. One sees in table I that the results of the simulation on finite samples are very close to these values.

Table I. — *Mean position of the frontier and concentration of diffused atoms at this position. These results are obtained from the* 2D *simulation for selected values of the diffusion length, both for square and triangular lattices. The values of* p_c *for these two lattices are respectively 0.593 and 0.5* [7].

	l_D	100	800	6 400
Square lattice	x_f/l_D	0.3736 ± 0.0008	0.3771 ± 0.0006	0.3781 ± 0.0003
	$p(x_f)$	0.5973 ± 0.0008	0.5938 ± 0.0006	0.5929 ± 0.0003
Triangular lattice	x_f/l_D	0.4699 ± 0.0008	0.4755 ± 0.0006	0.4757 ± 0.0003
	$p(x_f)$	0.506 ± 0.0007	0.5013 ± 0.0006	0.5011 ± 0.0003

Moreover the external frontier of the percolating cluster, the so-called « hull », has in fact the same definition as our seashore [6]. Thus the above ideas suggest that the diffusion front should have the same geometric properties as the percolation hull. The fractal dimension of the percolation hull has been determined for the square lattice to be $D_h = 1.74 \pm 0.02$ or $D_h = 1.76 \pm 0.01$ [6]. Comparison with the value $D_f = 1.76 \pm 0.02$ that we have determined supports the idea of a direct similarity between the diffusion front and the percolation hull.

We then start from the same consideration to predict theoretically the exponents for the dependence on l_D of the width σ_f and the length N_f of the diffusion front. From the data shown in figure 4 we obtain

$$\sigma_f = 0.46(l_D)^{\alpha_\sigma} \quad \text{with} \quad \alpha_\sigma = 0.57 \pm 0.01 \tag{7}$$

$$N_f = L \int_0^\infty p_f(x) \, dx$$

$$= 0.96 \, L(l_D)^{\alpha_N} \quad \text{with} \quad \alpha_N = 0.424 . \tag{8}$$

When x is larger or smaller than x_f the concentration deviates from p_c due to the gradient of the particle concentration. There are lakes and islands in these regions. The characteristic size of these objects should be of the order of the correlation length [7] corresponding to the value of the concentration at that coordinate :

$$\xi(x) = \xi_0 \, | p(x) - p_c |^{-\nu} \tag{9}$$

where ν is the usual critical exponent and ξ_0 is of the order of the lattice parameter [7].

If the distance to x_f of a cluster (lake or island) of characteristic size $\xi(x)$ is equal to ξ this cluster has obviously a finite probability of touching the frontier and thus to be within the frontier width. In other words

$$\sigma_f = K\xi(x_f \pm \sigma_f) \tag{10}$$

where K should be of the order of unity.

Using equations (1), (6), (7) and (9) we derive from (10) by a Taylor expansion

$$\sigma_f = K\xi_0 \left| \sigma_f \left(\frac{\partial p}{\partial x} \right)_{x=x_f} \right|^{-\nu}$$

$$= K\xi_0 \left(0.977 \, \frac{\sigma_f}{l_D} \right)^{-\nu}$$

thus

$$\sigma_f = K' \, \xi_0^{\frac{1}{1+\nu}} \, l_D^{\frac{\nu}{1+\nu}} \tag{11}$$

where K' is also of the order of one. Then we find

$$\alpha_\sigma = \frac{\nu}{1 + \nu} . \tag{12}$$

If we use for ν the value [8] $\nu = 4/3$ we predict $\alpha_\sigma = 4/7 = 0.5714$, while the simulation gives $\alpha_\sigma = 0.57 \pm 0.01$. The value of the prefactor obtained from the simulation (relation (7)) indicates that equation (10) is well verified.

We want now to interpret the exponent α_N that we observe for the dependence of N_f on l_D (Fig. 4). In fact N_f is not an intrinsic quantity because it is proportional to the lateral size L of the sample for L larger than σ_f. We propose to consider a more intrinsic quantity $N_f \, \sigma_f/L$ which

is the number of points on the frontier on a lateral scale σ_f. From (8) and (11) one can write

$$\frac{N_f \, \sigma_f}{L} \sim l_D^{\alpha_N} \, l_D^{\frac{\nu}{1+\nu}}. \tag{13}$$

But as shown above the frontier is a fractal on this scale so that from equations (4) and (11)

$$\frac{N_f \, \sigma_f}{L} \sim \sigma_f^{D_f} \sim l_D^{\frac{\nu}{1+\nu} D_f}. \tag{14}$$

By comparing (13) and (14) one finds

$$\alpha_N = \frac{\nu}{1+\nu} (D_f - 1). \tag{15}$$

Using $\nu = 4/3$ and our measured value $D_f = 1.76 \pm 0.02$ we predict $\alpha_N = 0.43 \pm 0.01$ while the simulation gives $\alpha_N = 0.425 \pm 0.005$.

A remarkable feature arises from table II where the values of $(N_f \, \sigma_f)/(Ll_D)$ are shown. This last
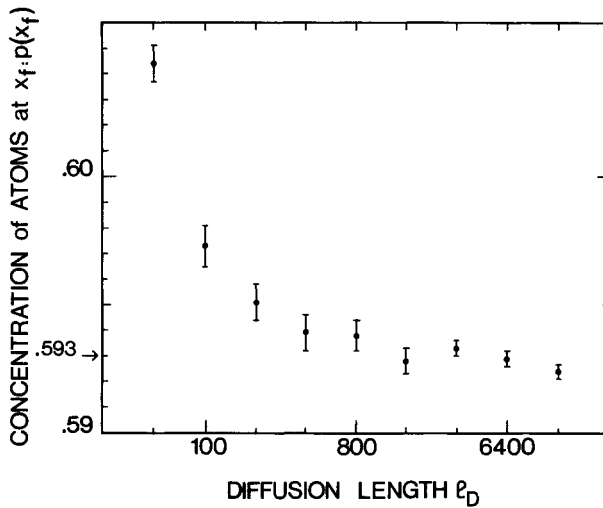

Fig. 5. — Diffused atom concentration at the mean front position x_f as a function of the diffusion length in the case of a square lattice. The approximate value $p_c = 0.593$ has been represented. The error bars represent statistical uncertainties.

Table II. — *Values of* $N_f \, \sigma_f/Ll_D$. *The result of the simulation suggests that this quantity tends to a constant value when* l_D *goes to infinity.*

l_D	200	800	1 600	6 400
$\dfrac{N_f \, \sigma_f}{Ll_D}$	0.464	0.447	0.442	0.441

quantity should vary as

$$\frac{N_f\,\sigma_f}{L l_D} \sim l_D^{(\alpha_N + \alpha_\sigma - 1)}.\tag{16}$$

In table II we see that this quantity is almost constant indicating that the number of points of the frontier contained in a square of side equal to the width of the frontier is proportional to the diffusion length, hence to the total number of diffused atoms. The exponent appearing in (16) should be zero, thus $\alpha_N + \alpha_\sigma - 1 = 0$ and

$$\alpha_N = \frac{1}{1+\nu} = 3/7 = 0.4286.\tag{17}$$

Moreover from (12) and (15) we conjecture that

$$D_f = \frac{1+\nu}{\nu} = 7/4 = 1.75\tag{18}$$

may hold in two dimensions. The same relation should hold for D_h the fractal dimension of the percolation hull.

In conclusion we have shown that the diffusion front is a fractal line with the same dimension as the « hull » of the infinite cluster in the percolation problem on a 2D lattice. The observation that the mean frontier occurs in the region where the concentration of diffused atoms is equal to the percolation value p_c indicates the close connection between this problem and the percolation problem. Moreover the width and the number of points on the frontier depends on the diffusion length through power laws, whose exponents are obtained with a very good precision. These exponents are not critical exponents but appear to be simply related to the critical exponents of percolation. This result suggests that a fruitful approach to percolation problems can be obtained in systems with a gradient of concentration. The case of a linear gradient will be discussed in detail in a forthcoming paper [9].

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