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## LETTER TO THE EDITOR

# Concentration gradient approach to continuum percolation in two dimensions

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Abstract. We have investigated percolation properties of a system of overlapping discs randomly distributed with a gradient of concentration. Fractal properties and critical exponents of this system appear to be identical to their counterparts on a lattice. This is in agreement with the universality of critical exponents of percolation. The concentration gradient approach permits a precise calculation of the percolation threshold, corresponding to a critical area fraction of  $0.6766 \pm 0.0005$ . We have investigated the part of the external perimeter of the percolation cluster which is accessible to discs of various sizes. The fractal dimension of this 'accessible' perimeter is found to decrease abruptly as a function of the radius of invading discs, from  $1.75 \pm 0.02$  to  $1.35 \pm 0.02$ .

Percolation in a gradient of concentration in 2D and 3D lattices has provided a new insight into the usual percolation problem and a precise determination of percolation parameters [1-3]. Gradient percolation has also been shown to provide a useful approach to different physical problems where concentration gradients are present, such as diffusion fronts or invasion of porous media under gravity [4-6]. Obviously, generalisation to the continuum is an important consideration.

Several authors have verified that percolation on a continuum belongs to the same universality class as usual percolation on a lattice [7-9]. A first goal of this study is to confirm this universality in the case of a gradient, allowing applicability of the gradient percolation approach to a wide class of experimental situations. Besides, a very accurate calculation of percolation parameters is possible, as in the case of lattice percolation.

We also investigate a question recently addressed by Grossman and Aharony [10, 11], concerning the fractal dimension of the 'accessible' perimeter of the percolating cluster. Looking for sites of the percolating cluster, accessible for particles with different sizes coming from the 'outside' (or infinity), Grossman and Aharony found that they constitute subsets of the hull with dimension  $D_h$ , decreasing from 1.75 to 1.35 when the particle size increases. Aharony [12], and Saleur and Duplantier [13] have recently predicted that the dimension should in fact vary as a step function of the particle size—as soon as some apertures in the hull are closed the dimension is reduced to the constant value 1.33. Recent simulations [14] support this prediction. However, on a lattice one can only investigate a discrete set of particle sizes. The present study of this problem in the continuum permits us to consider particles with size ranging continuously from zero to infinity.

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Gradient percolation on a lattice is very simply derived from the usual percolation problem [1, 4]. One considers a lattice where the concentration p of occupied sites may vary in a range  $p_0 . If the percolation threshold <math>p_c$  for this lattice lies within this range, one finds an 'infinite' or percolating cluster, which extends in the whole region  $p_c . We have shown that, for two-dimensional (2D) lattice systems$ [4], the front or external perimeter of this cluster is a fractal object [15], similar to thehull of the usual percolating cluster [16]. This front is situated in the region where $the concentration of the diffusing particles is very close to <math>p_c$ . It is restricted to a concentration region with a spatial width  $\sigma_f$  depending on the local concentration gradient  $\nabla p$  as

$$\sigma_{\rm f} \propto |\nabla p|^{-\alpha_{\sigma}} \tag{1}$$

where

$$\alpha_{\sigma} = \nu/(1+\nu) \tag{2}$$

with  $\nu$  being the critical exponent for the 2D percolation coherence length  $(\nu = \frac{4}{3})$ . Because the front is fractal the number of particles  $N_f$  in the front varies with  $\sigma_f$  as

$$N_{\rm f} \propto L \sigma_{\rm f}^{D_{\rm f}-1} \tag{3}$$

where  $D_{\rm f}$  is the fractal dimension of the front, and L measures the spatial extent of the front perpendicular to the concentration gradient  $(L \gg \sigma_{\rm f})$ . Hence  $N_{\rm f}$  is also a power-law function of the gradient

$$N_{\rm f} \propto |\nabla p|^{-\alpha_{\rm N}} \tag{4}$$

with

$$\alpha_N = (D_f - 1)\alpha_\sigma. \tag{5}$$

The fractal dimension  $D_f$  is thought to be given by  $D_f = (\nu + 1)/\nu = \frac{7}{4}$  in 2D systems [13, 17], a conjecture originally based on the observation that  $\alpha_N + \alpha_\sigma = 1$  [4].

Generalisation to the continuum is straightforward. We consider a random distribution of overlapping discs, with radius  $R_0 = 1$  in a rectangular box with dimensions  $L \times L'$  and a constant gradient of concentration along the x direction, corresponding to side L'. This distribution is such that the concentration  $\rho$  is maximum for x = 0and  $\rho = 0$  for x = L'. The maximum value for  $\rho$  is taken equal to  $2.25/\pi R_0^2$ , approximately equal to  $2 \rho_c$  where  $\rho_c$  is the percolation threshold. In consequence, in the high-concentration region almost all discs belong to the 'infinite cluster' whose extension is obviously limited in the low-concentration region. The determination of the external boundary of the infinite cluster is as follows (see figure 1). We start from the disc belonging to the infinite cluster such that the concentration at the position of the disc is minimum (this is the hatched disc in figure 1)—this disc obviously belongs to the front. Then, the next disc on the front (shown with dotted shading in figure 1) is that which first intersects the origin disc in a given direction (clockwise in this case). The front is constructed, step by step, following the same rule, with periodic boundary conditions in the direction perpendicular to the concentration gradient.

Simulations are systematically performed for a series of samples with decreasing concentration gradients  $\nabla p \propto R_0/L'$ . As for the lattice case, we calculate  $N_f$  and  $\sigma_f$ , from which we can deduce the exponents  $\alpha_{\sigma}$  and  $\alpha_N$  and the dimension  $D_f$ , from (1), (3) and (4), respectively. Variations of  $N_f$  and  $\sigma_f$  with gradient  $\nabla p$  give  $\alpha_N = 0.42$  and  $\alpha_{\sigma} = 0.56$  (see figure 2). The fractal dimension of the front is found to be  $D_f = 1.75 \pm 0.02$ . All these numbers are very close to their counterparts in the lattice problem [4], confirming the universality of these exponents.



Figure 1. Schematic illustration of the determination of the front (shown as shaded discs; see text for explanation).



Figure 2. Variation of the number  $N_f$  of points of the front per unit length (**I**) and of the width  $\sigma_f$  ( $\blacklozenge$ ) with the inverse gradient  $\nabla p^{-1} \propto L'/R_0$ .

Gradient percolation was shown to enable a very accurate determination of  $p_c$  in the lattice percolation problem [1, 2]. This determination is based on the observation that the front tends asymptotically to  $p_c$  when the gradient  $\nabla p$  tends to zero. This may be generalised to the continuum percolation problem. In this case, the percolation threshold may be determined for different quantities, such as critical area fraction  $\Phi_c$ , critical concentration  $\rho_{c'}$  or critical coordination number  $B_c$ . All these quantities may be simply deduced from one another [9, 18]. Basically, the critical value of a parameter is its value at the average position  $\langle x_f \rangle$  of the front:  $\langle x_f \rangle = (\Sigma_i x_i)/N_f$ , the sum being over all discs belonging to the front. We also consider the quantity  $\langle x_{fr} \rangle = (\Sigma_i x_i/\rho_i)/N_f$ , where  $\rho_i$  is the concentration at abscissa  $x_i$ . In the lattice case, Gouyet *et al* [19] have shown that  $p(\langle x_f \rangle)$  and  $p(\langle x_{fr} \rangle)$  converge to  $p_c$ , the percolation threshold, according to

$$p(\langle x\mathbf{r}_{\mathbf{f}}\rangle) - p_{c} \propto |\nabla p| \tag{6a}$$

$$p(\langle x_{\rm f} \rangle) - p(\langle x_{\rm fr} \rangle) \propto |\nabla p|^{6/7}.$$
(6b)

Figures 3(a) and 3(b) show that (6a) and (6b) are well verified, for a series of samples with  $R_0/L'$  ranging from  $\frac{1}{20}$  to  $\frac{1}{3000}$ , and total numbers of discs on the front being approximately equal to  $10^5$  for each gradient. Extrapolation of these data to zero gradient gives  $B_c = 4.515 \pm 0.006$ , corresponding to  $\Phi_c = 0.6766 \pm 0.0005$ . This is in agreement with, although more precise than, a previous determination by Gawlinski and Stanley [9].

In addition, when the front is defined we investigate its accessible part (Grossman-Aharony behaviour [10]) with the following method. Each disc on the front has its radius increased by a factor  $R/R_0$  (in the present calculation,  $R/R_0$  is increased from



Figure 3. (a) Variation of the average coordination number B per disc, at the location of the front  $\langle x_{fr} \rangle$  as a function of the concentration gradient  $\nabla p \propto R_0/L'$ . A linear variation is observed, from which we deduce  $B_c = 4.515$ . (b) Scaling behaviour of the quantity  $B(\langle x_f \rangle) - B(\langle x_{fr} \rangle)$  as a function of the concentration gradient.

1 to 2). Increasing R obviously closes apertures in the front, with approximate widths  $\Delta R = 2(R - R_0)$ . We then obtain a modified front, using the same rule as above for discs with radius R. Correspondingly, this modified front is defined by parameters  $N_f(R)$ ,  $x_f(R)$  and  $\sigma_f(R)$ . As above, using (1), (3) and (4) we obtain an R-dependent dimension  $D_f(R)$  and exponents  $\alpha_{\sigma}(R)$  and  $\alpha_N(R)$  (see figure 4). We observe a very abrupt decrease of  $D_f(R)$  as a function of R, from  $D_f = 1.75 \pm 0.02$  for  $R/R_0 = 1$ , to  $D_{f\infty} = 1.35 \pm 0.02$  for  $R/R_0 \rightarrow \infty$ . The value  $D_{f\infty} = 1.35$  is in agreement with the prediction of Saleur and Duplantier [13]. Figure 4 shows that  $\alpha_{\sigma}(R)$  is almost constant while  $\alpha_N(R)$  has a behaviour comparable to  $D_f(R)$  as a function of R, with a limiting value of  $\alpha_{N\infty} = 0.20 \pm 0.02$  for  $R/R_0 \rightarrow \infty$ .



**Figure 4.** Variation of exponents  $\alpha_{\sigma}(R)$  ( $\bullet$ ) and  $\alpha_{N}(R)$  ( $\blacksquare$ ) and of the fractal dimension  $D_{\rm f}(R)$  ( $\Box$ ) as a function of the ratio  $R/R_0$ .  $\alpha_{\sigma}(R)$  is almost constant, whereas  $\alpha_{N}(R)$  and  $D_{\rm f}(R)$  decrease very rapidly as functions of  $R/R_0$ .

The steep decrease observed for low values of  $R/R_0$  is in qualitative agreement with the prediction that only two values,  $\frac{7}{4}$  and  $\frac{4}{3}$  should be obtained [12, 13]. In particular, we cannot exclude here the possibility that the continuous decrease may be due to a finite gradient effect. Such an effect is visible on the  $\alpha_{\sigma}(R)$  against  $R/R_0$ curve (figure 4). It is responsible for the slight decrease of  $\alpha_{\sigma}(R)$  observed at small  $R/R_0$ . This decrease is associated with a different behaviour of the width  $\sigma_f(R)$  at large and small gradients. For large gradients,  $\sigma_f(R)$  decreases slowly as a function of  $R/R_0$ , whereas for small gradients,  $\sigma_f(R)$  follows rather a step-like function as a function of  $R/R_0$ .

One should note however, that this  $\sigma_f(R)$  effect only slightly alters the variation of  $D_f(R)$  as a function of  $R/R_0$ .

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