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CRITICAL DIMENSIONALITY FOR A SPECIAL PERCOLATION PROBLEM

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Résumé. — Nous considérons un système de chaînes idéales, à N maillons ($N \gg 1$) inscrites sur un réseau périodique à d dimensions. Il n'y a pas de corrélations entre chaînes différentes : chaque site peut être occupé par plusieurs maillons. Deux chaînes sont dites *connectées* si elles ont au moins un site en commun. Ceci définit un problème de percolation (où la variable est la fraction c de sites occupés) relié physiquement à certains mécanismes de *gélification* des polymères. Pour $d > 4$ le seuil de percolation c_0 est proportionnel à N^{-1} . Nous donnons un argument qui suggère que le comportement près de c_0 est alors du type champ moyen. Cette simplification est due au fait que $c_0 \gg c^*$ où c^* ($\sim N^{1-d/2}$) est la concentration à laquelle les chaînes commencent à s'interpénétrer. Pour $d < 4$ on prévoit $c_0 \sim c^*$ et des exposants critiques qui sont ceux de la percolation usuelle. La même dimensionalité critique ($d_c = 4$) devrait se retrouver pour le problème plus réaliste des chaînes avec interactions répulsives.

Abstract. — We consider a set of ideal chains, each with N beads ($N \gg 1$) inscribed on a d -dimensional periodic lattice. Different chains are uncorrelated : thus any lattice site may belong to more than one chain ; two chains are said to be connected if they have at least one site in common. This defines a percolation problem (where the variable is the fraction c of occupied sites) physically related to a gelation process in polymers. For $d > 4$ the critical fraction c_0 is proportional to N^{-1} and the behaviour near c_0 is of the mean field type. This simplification is due to the fact that $c_0 \gg c^*$, where c^* ($\sim N^{1-d/2}$) is the concentration at which the chains begin to overlap. For $d < 4$ we expect $c_0 \sim c^*$ and critical exponents not different from those of a site (or bond) percolation problem. We expect the same critical dimensionality to be maintained for the (more realistic) case of chains coupled by a repulsive interaction.

1. Introduction. — The percolation problems, introduced long ago by Broadbent and Hammersley [1], have many important applications in solid state physics for alloys, amorphous systems, etc. [2]. For instance, below a certain critical concentration c_0 , a disordered substitutional alloy $A_c B_{1-c}$ will show only finite clusters of A atoms. But, for $c > c_0$, a finite fraction of the A atoms will belong to a cluster of infinite size. The changes of behaviour occurring at $c = c_0$ have been studied by refined numerical methods [3]. As pointed out by various authors there is a certain qualitative similarity between these changes and a second order phase transition. In particular, a relation between percolation and the *Potts model* has been established [4a]; through this relation, and also through direct scaling arguments, it is possible to see that percolation follows a simple mean field behaviour only when the dimensionality d is larger than $d_c = 6$ [4b, 5].

The present note describes a different percolation problem, where the individual units are not atoms occupying one single lattice site, but *chains*, as represented on figure 1. We say that two chains are connect-

FIG. 1. — Chains inscribed on a square lattice ($d = 2$). The present example shows a cluster of two chains (A) and (B) with one intersection I. Each chain contains N beads ($N = 24$ on this example). Physically, point I might be associated with a chemical link established between the two chains e.g. a sulfur bridge in the vulcanization of rubbers.

ed if they have at least one site in common. Physically, this connection could correspond to the *vulcanization* process of rubbers, or to certain gelation processes, for which there exists a vast theoretical literature [6-11]. Our model, however, contains some extra, unphysical simplifications. Each chain is described by a simple

random walk of N steps (N is fixed and large). Different chains are uncorrelated and may intersect each other freely. The chain distribution is entirely characterized by a single parameter : the average number of beads per site c . This model is highly idealized, but may retain some interest for reasons to be explained below. (More general cases are briefly considered in section 4.)

A set of n connected chains which are not connected to any other chain will be called a *cluster* of rank n . In section 3 we analyse the distribution of clusters by a series expansion which follows the approach introduced in reference [12] for the simple site problem. For the moment, however, we shall present some qualitative remarks, which may help to understand why a critical dimensionality shows up in this problem. Let us start with a dimensionality d larger than 4. In this case, the following sequence of situations is found when c increases from 0 :

1.1 AT VERY SMALL c we have a dilute collection of coils, each with a radius $R_0 \sim N^{1/2}$. The coils are non overlapping provided that $c < c^*$ where

$$c^* \cong N/R_0^d \cong N^{1-d/2}. \quad (1.1)$$

In the regime $c < c^*$ there is clearly no percolation.

1.2 AT HIGHER CONCENTRATIONS $c_0 > c > c^*$ the coils overlap strongly, but the contacts between them are not numerous : the clusters are still finite. The concentration c_0 will turn out to be of order $1/N$, and the ratio

$$\frac{c^*}{c_0} = N^{2-d/2}$$

is much smaller than unity (for $d > 4$) : thus, domain (b) has a large span when N is large

1.3 FOR $c > c_0$ percolation sets in.

Note incidentally that all the concentrations of interest (c^* , c_0 , etc.) are small compared with unity when N is large : all the systems of interest here are dilute. (This is to be contrasted, for instance, with the practical vulcanization of rubbers, which takes place in a molten polymer with $c \sim 1$.)

The value of c_0 and the special role of $d = 4$ will now be explained by a qualitative argument : consider one chain (A) among the others. The probability that one particular site on (A) belongs also to another chain is proportional to c . The average number Γ of contacts between A and other chains is thus of order Nc . We may write Γ as a product

$$W\delta = \Gamma \sim Nc \quad (1.2)$$

where W is the average number of chains (B) in direct contact with (A), and $\delta (\geq 1)$ is the number of contact points for one (AB) pair. Let us estimate δ for $d > 4$. We consider two chains (A) and (B) which are known to have at least one site I in common. How many others than S do they have one the ave-

rage ? The two coils occupy volumes Ω_A , Ω_B of order R_0^d and the intersection $\Omega_A \cap \Omega_B = \Omega$, is also of order R_0^d . The density of beads from chain one is of order N/R_0^d and the long range contributions to $\delta - 1$ are

of order $N \frac{N}{R_0^d} = N^{2-d/2} \ll 1$. Thus, for $d > 4$, the

contributions to $\delta - 1$ come only from the near vicinity of I ; they are of order unity. Since $\delta \sim 1$, eq. (1.2) gives $W \sim Nc$. The percolation threshold is expected to occur when $W \sim 1$ [6]. This implies

$$c_0 \cong N^{-1} \quad (d > 4). \quad (1.3)$$

Note that for $d < 4$, the same argument leads us to

$$\delta \sim N^{2-d/2} \gg 1.$$

Again setting $W \sim 1$ we obtain from (1.2)

$$c_0 \sim N^{-1} \delta \sim N^{1-d/2} \sim c^* \quad (d < 4). \quad (1.4)$$

We have not been able to compute exactly the coefficient in equation (1.3), but, in section 3, we shall give some arguments concerning the behaviour for $c \rightarrow c_0$. They tend to prove that, for $d > 4$, the chain percolation problem is comparatively simple, and of the *mean field* type. On the other hand, for $d < 4$, the chain problem resembles percolation for a random set of (d dimensional) spheres-where two overlapping spheres are automatically connected. Thus, the critical behaviour for $d < 4$ must be essentially identical to what is known from the conventional percolation problem [3]. The main interest of our model is to exhibit this critical dimensionality effect.

In section 2 we consider the regime (b) with $c^* < c < c_0$ and $d > 4$. We analyze the probability for one chain to be disconnected from all other chains. In section 3 we generalize the discussion to clusters of n chains, and derive a generating function for clusters. In section 4 we extrapolate our discussion to the more difficult case of chains with repulsive interactions.

2. Statistical weight for one disconnected chain. —

We choose one chain (A) and put it on the lattice with a given conformation defined by lattice points $A_1 \dots A_N$. Keeping these points fixed, we now distribute all the other chains (B chains) at random on the lattice, and count how many conformations are allowed for them.

2.1 Without any restriction : the number is

$$\Xi_0 = \frac{1}{v!} K^v Z^N \quad (2.1)$$

where v is the number of B chains, K the number of sites on the lattice (proportional to the volume of the system) and Z the coordination number of each

site ⁽¹⁾. We have $N^v = cK$ where c is the bead concentration.

2.2 When the chains B are not allowed to intersect chain A. The number of possibilities is now reduced, and can be written as :

$$\Xi = \Xi_0 e^{-\Lambda v} \quad (2.2)$$

The exponential dependence on v expresses that different B chains are uncorrelated.

To compute the factor Λ we construct a perturbation series in the following way : we define a reduced statistical weight $G_N(12)$ for a (B) chain extending from site (1) to site (2), such that $Z^N G_N(12)$ gives the number of B chain conformations linking sites (1) and (2) in N steps. The effect of the (A) chain will be described by a repulsive potential $V(r)$ which is equal to $+V$ on the sites $A_1 \dots A_N$ and vanishes elsewhere. (Ultimately we shall let $V \rightarrow +\infty$.)

The equation for G is of the Schrödinger form [13]

$$\frac{\partial G}{\partial N}(12) = Z^{-1} \sum'_{3(1)} (G(13) - G(12)) - V(1) G(1) \quad (2.3)$$

where N has been treated as continuous, and the sum $\sum'_{3(1)}$ is limited to the nearest neighbours of site (1).

The boundary condition is

$$G_{N=0}(12) = \delta(12). \quad (2.4)$$

The perturbation series for G has the form

$$\begin{aligned} G_N(12) &= G_N^0(12) - \sum_{N_1} \sum_3 G_{N_1}^0(13) V(3) G_{N-N_1}^0(32) \\ &+ \sum_{N_1 N_2} \sum_{34} G_{N_1}^0(13) V(3) G_{N_2}^0(34) V(4) G_{N-N_1-N_2}^0(42) \\ &+ \dots \end{aligned} \quad (2.5)$$

where G^0 corresponds to an ideal chain, and is normalized according to

$$\sum_2 G_N^0(12) = 1. \quad (2.6)$$

The reduction factor Λ is related to this expansion

$$e^{-\Lambda} = \frac{\sum_{12} G_N(12)}{\sum_{12} G_N^0(12)} = K^{-1} \sum_{12} G_N(12). \quad (2.7)$$

We shall see that Λ is of order N^2/K , and thus infinitesimal

simally small in the limit of a large volume ($K \rightarrow \infty$) : this allows to rewrite eq. (2.7) and eq. (2.5) as

$$\begin{aligned} \Lambda &\cong 1 - e^{-\Lambda} \\ &= K^{-1} \sum_{N_1} \sum_{123} G_{N_1}^0(13) V(3) G_{N-N_1}^0(32) - \\ &\quad - K^{-1} \sum_{N_1 N_2} \sum_{1234} G_{N_1}^0(13) V(3) \\ &\quad \times G_{N_2}^0(34) V(4) G_{N-N_1-N_2}^0(42) \\ &\quad + \dots \end{aligned} \quad (2.8)$$

The potential V is a sum of terms corresponding to all the sites of chain A. We shall rearrange eq. (2.9) using the method of Watson [14] : successive scatterings by one same site are added : the result is a certain T matrix, which in the N representation appears as a retarded operator $T_{NM}(R) = T_{N-M}(R)$. However, in the limit of strong repulsive V which is of interest here, retardation becomes negligible and we can put $T_{NM} \rightarrow T \delta_{NM}$ as T remains finite and of order unity in this limit [12]. The rule is now to replace V by T in eq. (2.8) and to retain only the terms where the same site never appears in two consecutive positions.

Having an explicit (although complex) expression for the reduction factor Λ we shall now average it over all conformations of chain A, and investigate the series (2.8). The first term gives simply

$$\Lambda_1 = K^{-1} T \tilde{N} N \quad (2.9)$$

where $\tilde{N}$ is the average number of *distinct* lattice sites occupied by chain A. The calculation of $\tilde{N}$ is described (for instance) in the appendix of reference [12]. $\tilde{N}$ is proportional to N ; the ratio $\tilde{N}/N$ is a numerical constant depending on the lattice symmetry and on the dimensionality d . Thus, we may write

$$\Lambda_1 = K^{-1} \alpha_1 N^2 \quad (2.10)$$

where α_1 is a factor of order unity.

The corrections of order T^2 are less trivial : integrating over (1) and (2) in eq. (2.8) and making use of (2.6) one arrives at

$$\Lambda_2 = -K^{-1} N \sum_{N_2} \sum_{34} \langle T(3) T(4) \rangle G_{N_2}^0(34). \quad (2.11)$$

The potential correlation function $\langle T(3) T(4) \rangle$ is simply proportional to the probability $\tilde{p}(4-3)$ of finding one monomer of chain A at point 4 when one monomer is known to be at point 3. Neglecting end effects :

$$\tilde{p}(4-3) = 2 \sum_{M=1}^{\infty} G_M^0(34) \quad (2.12)$$

$$\langle T(3) T(4) \rangle = T^2 N K^{-1} \tilde{p}(4-3). \quad (2.13)$$

Returning to (2.11) we then see that the sum $\sum_{N_2}$ also gives a factor $\tilde{p}$

$$\Lambda_2 = \text{const. } K^{-1} N^2 T^2 \sum_{\mathbf{R}} \tilde{p}^2(\mathbf{R}). \quad (2.14)$$

⁽¹⁾ For simplicity, our counting is done assuming that the *head* and the *tail* of each chain are distinguishable.

The function $\tilde{p}(\mathbf{R})$ has been discussed by many authors, and in particular by Edwards [15]. It is easy to see from (2.12) and from the equation ruling G^0 that (in the continuous limit) :

$$\nabla^2 \tilde{p}(\mathbf{R}) = 2 \delta(\mathbf{R}) \quad (2.15)$$

and thus

$$\tilde{p}(\mathbf{R}) = \text{const.} \frac{1}{R^{d-2}} \quad (R \rightarrow \infty), (d > 2). \quad (2.16)$$

Inserting this into (2.14) we see that the sum $\sum_R$ is of the form

$$\int_a^\infty R^{d-1} dR \frac{1}{R^{2d-4}} = \int_a^\infty \frac{dR}{R^{d-3}} \quad (2.17)$$

(where a is a cut off comparable to the lattice distance). This converges provided that $d > 4$: the correction A_2 is then finite and comparable to A_1 :

$$A_2 = K^{-1} \alpha_2 N^2 \cong A_1. \quad (2.18)$$

Proceeding to the higher order terms, we find that A_3 contains contributions from three distinct sites which involve the square of (2.17) and thus have the same convergence properties, plus terms involving only two sites which are then connected by two propagators G^0 , and ultimately lead to an integral over p^3 . The latter converges more easily than (2.17). On the whole, we arrive at

$$A = \alpha K^{-1} N^2 \quad (d > 4) \quad (2.19)$$

where α is some unknown numerical constant. We may say that the probability for one chain to be disconnected from all others is thus

$$e^{-A} = e^{-\alpha N c}. \quad (2.20)$$

We shall often refer to αN as the effective number of excluded sites.

3. The percolation series. — Let us now return to the chain percolation problem : we choose one lattice site O and calculate the probability $P_n(c)$ for this site to belong to a cluster of n chains [12]. The sum of these probabilities will be written in the form :

$$\sum_{n=1}^{\infty} P_n(c) = c \Sigma(c). \quad (3.1)$$

Below the percolation threshold ($c < c_0$) the sum is absolutely convergent and $\Sigma(c) \equiv 1$, but for $c > c_0$ the probability of belonging to an infinite cluster is different from zero : since this is not counted in eq. (3.1) we then have $\Sigma(c) < 1$.

An important practical simplification comes from the fact that $c_0 \ll 1$. Thus, for all c values of interest, factors like e^{-c} can be replaced by unity (but $e^{-Nc} < 1$).

Let us start by a discussion of $P_1(c)$. This corresponds to one disconnected chain going through O . The probability of finding the site occupied is c . The probability for the corresponding chain to be isolated is given by eq. (2.20). Thus

$$P_1(c) = c e^{-\alpha N c} \quad (d > 4). \quad (3.2)$$

Consider now a cluster of two chains : one of them (A) includes the site O , while the other chain (B) crosses A at (at least) one point I . Let us first determine the probability for an (AB) system like this to be disconnected from all other chains. We can repeat the argument of section 2. The main point is that because integrals like (2.17) converge, the reduction factor A is again a sum of local contributions all along the cluster of interest : the number of effective sites is an extensive function. Thus, neglecting only end effects (of relative importance $1/N$), we may say that the probability of isolation is $e^{-2N\alpha c}$. To obtain $P_2(c)$ from this, we must count all possible locations of the intersection point I among the effective sites ; Their number is $N\alpha$. We must also insert one factor c for each chain. The result is

$$P_2(c) = \alpha c^2 N e^{-2\alpha N c}. \quad (3.3)$$

In fact we do not need to do the counting explicitly, because of a sum rule : for $cN \rightarrow 0$, the series $c\Sigma(c)$ must be identically equal to c (or Σ identical to one) as explained earlier in this section. Thus we must have

$$P_1(c) + P_2(c) = c - O(c^3). \quad (3.4)$$

Expanding the exponential in P_1 we see that

$$c - \alpha N c^2 + \alpha N c^2 = c - O(c^3) \quad (3.5)$$

as required by the sum rule.

The argument is easily generalized to higher orders : the crucial point is that for an n -cluster (with n finite), the number of effective sites is still extensive, and that the probability of isolation is $e^{-nN\alpha c}$. This leads to a series for the function Σ of eq. (3.1) :

$$\begin{aligned} \Sigma(\mu) &= e^{-\mu} + \mu e^{-2\mu} + \frac{3}{2} \mu^2 e^{-3\mu} + \frac{8}{3} \mu^3 e^{-4\mu} + \dots \\ &= \sum_{n=1}^{\infty} a_n \mu^n e^{-n\mu} \\ \mu &= \alpha N c \end{aligned} \quad (3.6)$$

where the successive coefficients a_n are again imposed by the sum rule. We shall now see that this series is uniformly convergent only when $\mu < 1$. In fact, an analytic continuation of (3.6) is given by the solution of the implicit equation

$$\tilde{\Sigma} = e^{\mu(\tilde{\Sigma}-1)}. \quad (3.7)$$

To see this, write eq. (3.7) in the form

$$\tilde{\Sigma} = e^{-\mu} \left[1 + \mu \tilde{\Sigma} + \frac{\mu^2 \tilde{\Sigma}^2}{2!} + \dots \right] \quad (3.8)$$

and solve by iteration, starting with $\tilde{\Sigma} = e^{-\mu}$ as the 0 order approximation, and inserting this value on the right hand side of (3.8).

a) It is easily checked that the first few terms of the series for Σ and $\tilde{\Sigma}$ coincide.

b) The coefficients in both series obey the same sum rule since $\tilde{\Sigma} = 1$ is one solution to eq. (3.7).

c) Since the algorithm for the construction of successive terms is the same, and the first term is also the same, for both series, we must have $\Sigma = \tilde{\Sigma}$ for all μ where (3.6) is uniformly convergent.

The function $\tilde{\Sigma}(\mu)$ is easily constructed by inversion of eq. (3.7)

$$\mu = \frac{\ln \tilde{\Sigma}}{\tilde{\Sigma} - 1}. \quad (3.9)$$

The general aspect of $\tilde{\Sigma}(\mu)$ is shown on figure 2. The change in behaviour occurs at $\mu = 1$. Thus the critical concentration is

$$c_0 = \frac{1}{\alpha N}. \quad (3.10)$$

FIG. 2. — Probability η for one bead to belong to an infinite cluster, as a function of the reduced bead concentration $c/c_0 = \mu$. This curve is expected to hold only when the dimensionality d is larger than 4.

For c slightly larger than c_0 we may write

$$\tilde{\Sigma} = 1 - \eta \quad (3.11)$$

and we have :

$$\mu = \frac{1}{\eta} \left[\eta + \frac{\eta^2}{2} + \frac{\eta^3}{3} + \dots \right] \quad (\mu > 1)$$

$$\mu - 1 = \frac{1}{2} \eta + \frac{1}{3}.$$

The quantity η measures the fraction of atoms which belong to an infinite cluster, and we seen that for μ slightly larger than 1, η is a linear function of μ

$$\eta \sim 2(\mu - 1). \quad (3.12)$$

This behaviour characterizes all mean field approximations in the percolation problem [12]. The same

structure for $\Sigma(\mu)$ is also well known for lattices which are not periodic, but which have a special tree structure (Bethe lattices) [16]. It is also of interest to investigate the region where c is slightly smaller than c_0 . Consider for instance the average number of chains in a cluster

$$m = \sum_{n=1}^{\infty} n a_n \mu^n e^{-n\mu} \quad (\mu < 1) \quad (3.13)$$

$$= \frac{1}{1 - \mu} \frac{\partial}{\partial \mu} (\mu \Sigma). \quad (3.14)$$

In the region of $\mu < 1$, Σ is constantly equal to unity and

$$m = \frac{1}{1 - \mu} \quad (3.15)$$

again characteristic of a mean field behaviour.

One remark should be added at this point : our discussion of successive terms $a_n \mu^n e^{-n\mu}$ in the series $\Sigma(\mu)$ assumed long chains (N large) and n finite. Very close to $\mu = 1$ we shall need large values of n , and our arguments break down. There will be a *crossover* of the following type

$$\left. \begin{array}{l} |\mu - 1| > N^{-\Phi} \text{ mean field behaviour} \\ |\mu - 1| < N^{-\Phi} \text{ critical behaviour} \end{array} \right\}$$

Here Φ is an (unknown) crossover exponent ($\Phi > 0$), but for all practical purposes with N large, the simple mean field behaviour should apply.

4. Concluding remarks. — **4.1 CHAINS WITH REPULSIVE INTERACTIONS.** — A discussion of the chain percolation problem with $d = 3$, and chains which are coupled by a finite repulsive interaction would be of some practical interest for gelation problems in polymer solutions [6-11]. Here we have discussed only $d > 4$ and ideal chains. However, we wish to point out that repulsive interactions between chains do not change the picture very drastically.

4.1.1 For $d > 4$ the propagator $\tilde{G}$ for a self avoiding chain differs from the propagator G_0 for an ideal chain only by relatively trivial renormalization factors [17]. All the changes can be incorporated in a correction to the numerical factor α .

4.1.2 For $d < 4$ the qualitative arguments of section 1 can be adapted as follows. To estimate the number δ of contacts between two chains which have one contact (as in the text after eq. (1.2)) we write

$$\delta \sim \frac{N^2}{R^d} \quad (4.1)$$

where R is now the radius of a swollen coil. Anticipating the result that c will not be much larger than the overlap concentration c^* , we may write $R \sim N^\nu$ where ν ($> \frac{1}{2}$) is the critical exponent associated with

a single coil. An excellent value for ν is given by Flory [6] :

$$\nu = \frac{3}{d+2} \quad (d < 4). \quad (4.2)$$

The overlap concentration is now

$$c^* \sim \frac{N}{R^d} \sim N^{1-\nu d} \quad (4.3)$$

and the number of contact points δ from eq. (4.1)

$$\delta \sim N^{2-\nu d}. \quad (4.4)$$

Finally, the equation replacing (1.1) for the percolation threshold is

$$c_0 \sim N^{-1} \delta \sim N^{1-\nu d} \sim c^*.$$

Thus, even when the chains repel each other, there is a change in behaviour at $d = 4$ from a regime $c_0 \sim N^{-1}$ ($d > 4$) to a regime $c_0 \sim c^*$ ($d < 4$).

4.2 POSSIBLE APPLICATIONS OF RENORMALIZATION GROUPS. — It might be feasible to compute the critical exponents for the chain percolation problem (with, or without, repulsive interactions) in three space dimensions by an expansion in powers of $\varepsilon = 4 - d$, similar in spirit to the Wilson calculation for the n -vector model of phase transitions [1]. This would be of direct interest for various gelation problems.

However, it could happen that the ε expansion does not exist for our problem, and that the critical exponents switch abruptly at $d = 4$, returning for $d < 4$ to the values which are numerically known for site or bond percolation [3]. Even in this case, a study of the crossover between chain behaviour and site behaviour would be of some interest.

On the whole, the present paper suffers from two opposite defects : it completely lacks mathematical rigor, and at the other end it is still very far from any practical polymer situation, but there is a slight hope that it may help to bridge the gap between these two points of view.

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