# Fereydoon Family<sup>1</sup>

Conformation of branched random fractals formed in equilibrium processes is discussed using a Flory-type theory. Within this approach we find only three distinct types or classes of random fractals. We call these the *extended*, the *compensated*, and the *collapsed* states. In particular, the critical clusters in thermal phase transitions are found to be of the compensated type and have approximately the same value of the fractal dimension. The Flory theory predicts the upper critical dimension for these clusters to be 6 instead of 4. This result and the apparent "grand" universality of the fractal geometry of the clusters in critical phenomena are discussed.

**KEY WORDS:** Fractals; critical phenomena; percolation; polymers; Flory theory; universality.

# 1. INTRODUCTION

Fractals<sup>(1)</sup> and the concept of fractal dimensionality have recently received considerable attention<sup>(1-13)</sup> owing to their applicability to a wide range of physical phenomena, including percolation,<sup>(1-5)</sup> polymers,<sup>(6-8)</sup> aggregation,<sup>(9-12)</sup> and gelation.<sup>(12,13)</sup> The fractal dimension D of an object embedded in a d-dimensional space has a value between 1 and d. Therefore, depending on the details of the process used to form the fractal, an object can have a value of D different from that of any other fractal. Thus, a natural question to ask is: How fundamental are these fractal dimensions? Are all fractals different, or, are some of them of the same basic type? The situation here is very much analogous to particle physics, where a wide variety of so called "elementary" particles are found in nature. What one would like to do is to find a classification scheme for these particles. In such

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a scheme, either all particles are found to be equally "fundamental," or, as the case appears to be, they can be cataloged into a finite number of classes of particles.

Fractals can be divided into two basic types: (a) deterministic fractals and (b) statistical (or random) fractals. Deterministic fractals are self-similar objects that are precisely constructed from some basic rules.<sup>(1)</sup> The best examples of these fractals are the-Koch curve,<sup>(1)</sup> Sierpenski gaskets,<sup>(1)</sup> Havlin carpets,<sup>(14)</sup> and Vicsek snowflakes<sup>(15)</sup> (see Fig. 2a). The most important property of deterministic fractals is that their fractal dimension is exactly known. Since a given line, plane, or volume can be divided up in an infinite number of different ways, it is possible to construct infinitely many different deterministic fractals with different fractal dimensions. Therefore, deterministic fractals cannot be classified into a finite number of classes without introducing, in addition to their fractal dimension, other parameters to characterize them.

In contrast to deterministic fractals, statistical fractals are constructed by random processes. The element of randomness makes them a more realistic representation of physical phenomena. The fact that randomness alone, i.e., without any spatial correlations, is sufficient to produce fractals was first pointed out by Mandelbrot.<sup>(1)</sup> The best example of such a fractal is the path of a random walker. However, purely random models are inadequate for most applications to real physical systems.<sup>(8)</sup> The reason is that the excluded volume effect, which is the geometrical constraint that prohibits two different elements from occupying the same spatial point, is not taken into account in these models. For this reason, a wide variety of random models with excluded volume have been introduced and studied in the past several years. Perhaps the best-known examples of these models are selfwalks,<sup>(16)</sup> animals, (17-19) avoiding random lattice and random percolation. (4,20)

Fractals have distinct topological structures depending on the maximum number of elements that can be joined to a given element of the system. If each element can be connected to at most two other elements, the resulting structure has no branches. In analogy with linear polymers, we call these types of structures "linear" fractals. In contrast, when branching can occur, i.e., three or more elements can be joined to a given element, the resulting fractal has a networklike structure. We call these types of objects "branched" fractals. Figure 1 shows two examples of "linear" fractals, (a) a Koch curve and (b) a self-avoiding walk, representing, respectively, an example of a deterministic fractal and a random fractal. Figure 2 shows two examples of "branched" fractals, (a) a Vicsek snowflake<sup>(15)</sup> and (b) a percolation cluster, representing, respectively, an example of a deterministic fractal and a random fractal. For the sake of clarity we confine our discussion and calculations to



Fig. 1. Two examples of linear fractals: (a) a Koch curve, and (b) a self-avoiding walk, representing a deterministic fractal and a random fractal, respectively.

branched random fractals.<sup>2</sup> The case of linear random fractals follows similarly.

How many different types of random fractals exist in equilibrium clustering phenomena? This is the main question of interest in this paper. We begin with some preliminaries in Sections 2 and 3. In Section 2 we discuss the definition of a cluster and point out the importance of properly defining a cluster for interacting systems. In Section 3 we define a fractal and show how its fractal dimension D is determined. We discuss the known scaling relation between D and the critical exponents for percolation and show that in thermal critical phenomena a similiar relation can be defined between the fractal dimension of critical clusters and the critical exponents. Using this relation we determine the fractal dimension of a wide variety of models of critical phenomena. The results are listed in Table I, where it is seen that the critical clusters appear to have approximately the same value of D. This result was first recognized by Suzuki,<sup>(21)</sup> in the context of critical

<sup>2</sup> We only consider systems with isotropic, short-range interactions. We also do not consider conformation of other fractals on random fractals, e.g., backbone, etc.



Fig. 2. Two examples of branched fractals: (a) a Vicsek showflake, and (b) a percolation cluster, representing a deterministic fractal and a random fractal, respectively.

d	Model	D
2	q-state Potts model <sup>(41)</sup>	
	q = 1 (percolation)	91/48 = 1.896
	q = 2 (Ising)	15/8 = 1.875
	q = 3 (hard-Hexagon)	28/15 = 1.867
	q = 4 (Baxter-Wu)	15/8 = 1.875
	Baxter-Wu model with impurities <sup>(42)</sup>	$1.9 \pm 0.1$
3	q-state Potts model:	
	q = 1 (percolation) <sup>(52)</sup>	$2.48 \pm 0.09$
	$q = 2 \; (\text{Ising})^{(53)}$	$2.484 \pm 0.004$
	<i>n</i> -vector model <sup>(53)</sup> ;	
	n = 1 (Ising)	$2.484 \pm 0.004$
	n=2(XY)	$2.483 \pm 0.004$
	n = 3 (Heisenberg)	$2.483 \pm 0.004$
	$n = \infty$ (spherical)	2.5
	Ising magnet with impurities <sup>(54)</sup>	$2.48 \pm 0.08$
	"Nonuniversal" microemulsion	$2.4 \pm 0.1^{(43)}$
	systems	$2.5 \pm 0.1^{(44)}$
	-	$2.4 \pm 0.1^{(45)}$
		$2.5 \pm 0.07^{(46)}$
		$2.5 \pm 0.1^{(46)}$
		$2.6 \pm 0.2^{(47)}$

Table I. Fractal Dimension of the Critical Clusters in Critical Phenomena Determined from the Scaling Relation (3)

phenomena, who proposed that all critical systems have the same value of the magnetic scaling power (this is often called the extended universality hypothesis). In order to classify random fractals into different classes, we begin Section 4 by determining the values of D for random fractals using the Flory approach for polymers.<sup>(22-27)</sup> We find that within this approximation, random fractals have only three possible conformations depending on the statistics of the system. We call these three conformations the *extended state*, the *compensated state*, and the *collapsed state* of a random fractal. In particular, critical clusters, i.e., large clusters at the critical points, have the conformation of a cluster in the compensated state. This implies that from the point of view of fractal geometry, the compensated state is like a "grand" universality class to which all critical clusters belong. This result provides a clear interpretation of the Suzuki universality.<sup>(21)</sup> In addition, we find that the upper critical dimension for the fractal dimension of critical clusters is  $d_c = 6$ , instead of the usual  $d_c = 4$  for critical phenomena.

# 2. PROPER DEFINITION OF A CLUSTER

The basic objects whose fractal properties we would like to investigate here are the clusters formed in a random process. Therefore it is important to have a precise definition of a cluster. For the sake of clarity, consider lattice models in which the basic elements of the system, e.g., spins, monomers, particles, atoms, etc., are placed at lattice sites or bonds. Usually a cluster is defined as a collection of elements connected by nearest-neighbor distances. This definition is known to be suitable for describing clusters in geometrical models like percolation,<sup>(4,20)</sup> and lattice animals.<sup>(8,18)</sup> However, a purely geometrical definition is inadequate for systems where thermal interactions are present.<sup>(28-30)</sup> The reason is that when thermal interactions are present the equilibrium conformation of a cluster is determined by both *geometrical* and *thermal* correlations.

In thermal critical phenomena, the main problem is a proper definition of the cluster whose size diverges at the critical point of the system with the correct exponents.<sup>(28-30)</sup> It is well known that the usual definition of a cluster as a collection of nearest-neighbor elements does not describe a critical system properly. For example, in the Ising model, clusters made of nearestneighbor "down" spins diverge below the critical temperature in three dimensions.<sup>(31)</sup> In two dimensions, these clusters are topologically constrained to diverge at the critical point, (32) but the mean cluster size diverges with an exponent larger than the susceptibility exponent.<sup>(33)</sup> One possible definition has been proposed by Coniglio and Klein<sup>(28)</sup> for the Ising model, and has been extended to the q-state Potts model by Coniglio and Peruggi.<sup>(30)</sup> In these models<sup>(28,30)</sup> a cluster is made of nearest-neighbor elements connected by thermally active bonds, the probability of a bond being active is p and inactive 1 - p. The elements interact according to the system Hamiltonian and the bonds are only introduced to define the connectivity between two nearest-neighbor elements. Thus, the presence or absence of a bond does not affect the thermal interaction energy between the elements. Using this definition, large clusters made of, say, nearest-neighbor down spins in the Ising model are broken up into smaller clusters by the introduction of the bond probability p. Renormalization group calculations<sup>(28,29)</sup> and Monte Carlo simulations<sup>(34)</sup> have shown that these clusters diverge at the critical point with the correct exponents. Therefore in the study of thermal critical phenomena, whenever we refer to a cluster we mean a propertly defined cluster similar to the ones discussed above.

## 3. FRACTAL DIMENSION AND SCALING

The fractal dimension D is a quantitative measure of the degree of ramification or stringiness of an object and is defined in the following way.<sup>(1)</sup>

First, the size of the object is determined by covering it with *d*-dimensional spheres of diameter *r*. Let us assume that N is the number of spheres needed to cover it. If in the limit as  $r \rightarrow 0$ , N varies as

$$N \sim r^{-D} \qquad (r \to 0) \tag{1}$$

then D is the fractal dimension of the object. In this definition the object is assumed to be scale invariant at all length scales smaller than its overall size.

In a real object there exists a minimum microscopic length scale of the order of the diameter of a typical element of the system beyond which scaleinvariance property breaks down. This implies that the limit  $r \rightarrow 0$  cannot be taken in a real object and relation (1) cannot be used to define D. However, D can be determined if the length of the object, e.g., its average radius, tends to infinity as the number of elements N in the object tend to infinity. In order to define D in this case, we note that if the number of spheres of radius r needed to cover a fractal varies as  $r^{-D}$ , then the number of elements within a sphere of radius R must scale as  $R^{D}$ . Therefore, in the limit  $N \rightarrow \infty$ , D is defined by

$$N \sim R^D \qquad (R \to \infty) \tag{2}$$

In polymer literature<sup>(8,24)</sup> this relation is generally written as  $R \sim N^{\nu_F}$  where the Flory exponent  $\nu_F = 1/D$ .

The fractal dimension of purely random models, i.e., models without the excluded volume effect, can be determined exactly. For linear fractals this corresponds to the path of a random walker.<sup>(1)</sup> Since the mean-square end-to-end distance of a random walker is proportional to the number of steps N, then relation (2) implies that D = 2, independent of the dimension.<sup>(1)</sup> For branched random fractals, the corresponding model is a randomly branching cluster whose branches obey the random walk statistics. These types of structures have been studied in a number of different contexts,<sup>(35-37)</sup> where it is found that D = 4, independent of the dimension. In contrast to these "ideal" random models, models with excluded volume, i.e., with correlations, cannot be solved exactly in general. The values of D for these types of systems are usually obtained from either numerical methods, such as Monte Carlo simulations<sup>(3,4,9,10,38)</sup> and exact enumerations,<sup>(39,19)</sup> or from position space renormalization group calculations.<sup>(8,11)</sup>

## Scalling Relation for D

A scaling relation between the fractal dimension D and critical exponents was first obtained for percolation.<sup>(2-5)</sup> The probability  $P_{\infty}(p)$  that at concentration p an element of the system belongs to the infinite cluster

varies as  $P_{\infty} \sim (p - p_c)^{\beta}$ , where  $\beta$  is the order parameter exponent and  $p_c$  is the percolation threshold.<sup>(4,20)</sup> At the critical point, according to finite size scaling,  $P_{\infty} \sim L^{-\beta/\nu}$ , where L is the length of the system and  $\nu$  is the correlation length exponent. Therefore, in a d-dimensional finite system of side L, the number of elements belonging to the infinite cluster varies as  $L^{d}L^{-\beta/\nu}$ , implying that  $D = d - \beta/\nu$ . Using the relations  $\nu^{-1} = y_T$  and  $\beta = (d - y_h)/y_T$  between the critical exponents and the thermal scaling power  $y_T$ and the magnetic scaling power  $y_h$ , we find the well-known relation

$$D = d - \beta / v = y_h \tag{3}$$

This scaling relation for D has been extensively tested for percolation.<sup>(2-5)</sup>

Following similar arguments as for the infinite cluster in percolation, a scaling relation can be obtained between D and the exponents in thermal critical phenomena. Consider the largest cluster in a system at the critical point. The fraction of the elements of the system belonging to this cluster is proportional to the order parameter which varies near  $T_c$  as  $\varepsilon^{\beta}$ , where  $\varepsilon = (T - T_c)/T_c$ . Again, at the critical point, finite size scaling asserts that the number of elements in the critical cluster varies as  $L^{d-\beta/\nu}$ . This implies that for critical phenomena, D is given by relation (3), but with the exponents  $\beta$  and  $\nu$  of thermal critical phenomena. Recently,<sup>(40)</sup> the scaling relation (3) has been verified in d=2 for the q-state Potts model by a direct measurement of the fractal dimension of Potts clusters.

With the use of relation (3), the fractal dimension of critical clusters in a wide variety of systems can be determined. Table I contains a list of these values for the most commonly studied models of critical phenomena. It is not difficult to see from this table that in contrast to what might have been expected—that the value of D are randomly distributed between 1 and d—these systems have values of D that are much closer to each other than to either 1 or d. This behavior is considerably different from the behavior of the thermal critical exponents. For example, the exponent  $\alpha$  for the pure Baxter–Wu<sup>(41)</sup> model is 2/3, whereas its value for the Baxter–Wu model with impurities<sup>(42)</sup> is  $\leq 0$ . In contrast, both models have  $D \simeq 1.9$ . In order to emphasize the generality of this observation, we have listed the values of D, obtained from relation (3), for a number of microemulsion systems<sup>(43-47)</sup> which have very unusual or "nonuniversal" exponents. Again, their fractal dimension is approximately the same as other critical systems.

The above result was first recognized by Suzuki,<sup>(21)</sup> who postulated that all critical systems have the same value of the magnetic scaling power (this is usually called the extended universality hypothesis). In the next section we provide a physical interpretation of this "grand" universality based on fractal concepts.

### 4. EQUILIBRIUM CONFORMATION OF RANDOM FRACTALS

What are the basic equilibrium shapes of random fractals? In order to answer this question, let us begin by listing the values of the fractal dimensions of models of random fractals in Table II. By looking at this table we see that the numbers fall essentially into three groups. In Sections 4.1–4.3 we use a Flory-type theory<sup>(22–27)</sup> to explain why random fractals have only three possible equilibrium shapes. We also determine the fractal dimension and the upper critical dimension  $d_c$ , for each case.

d	Model	D
2	Lattice animals (random clusters, branched polymers)	1.56
	Critical clusters (see Table I)	~1.9
	Compact clusters	2
3	Lattice animals, (random clusters, branched polymers)	2
	Critical clusters (see Table I)	~2.5
	Compact clusters	3

 Table II.
 Fractal Dimension of Equilibrium Models of Random

 Fractals in Two and Three Dimensions

## 4.1. Extended State

Let us begin by considering the following question: Under what physical condition(s) will a random fractal have the most ramified structure? Clearly when there are many clusters in the system they all compete for the available volume. Therefore, presence of other clusters limits the extent to which a cluster can become ramified; a cluster is more ramified in isolation than in a concentrated "solution."

The second factor which affects the shape of a cluster is the presence or absence of attractive interactions. For example, attractive interactions are less effective at high temperatures than at low temperatures. Therefore, an isolated random fractal at high temperatures will have a more ramified structure than at low temperatures. The best examples of this type of object are isolated polymers<sup>(24)</sup> in good solvents or at high temperatures. We call this state of a random fractal the *extended state*, because in this state a random fractal has its smallest value of D.

Let us use the Flory theory approach of Isaacson and Lubensky<sup>(26)</sup> and Daoud and Joanny<sup>(27)</sup> to determine the fractal dimension of an isolated,

random cluster. This method is based on finding the most probable conformation of a cluster from an approximate free energy. The free energy is written as a sum of the elastic free energy and the repulsive free energy. The elastic free energy (i.e., the entropic term) tends to make the cluster radius Requal to its radius  $R_0$  in the ideal state where there are no repulsive interactions present (see Section 3). The elastic free energy is written

$$F_{\rm el} = R^2 / R_0^2 \tag{4}$$

This term is assumed to be the same for any random structure. Since  $N \sim R^4$  for an ideal branched fractal, then

$$F_{\rm el} = R^2 / N^{1/2} \tag{5}$$

In an isolated cluster the excluded volume effect acts like a repulsive twobody interaction. Within the Flory theory  $^{(22-27)}$  the interactions between each of the N elements of the cluster and all the N-1 elements is approximated as an average interaction spread over a volume  $\mathbb{R}^d$ . This repulsive free energy is written  $^{(22-27)}$ 

$$F_{\rm rep} = N^2 / R^d \tag{6}$$

Minimization of the total free energy

$$F = F_{\rm el} + F_{\rm rep} = \frac{R^2}{N^{1/2}} + \frac{N^2}{R^d}$$
(7)

with respect to R gives

$$N \sim R^D$$
, with  $D = \frac{2(d+2)}{5}$  (extended state) (8)

This result was first obtained by Isaacson and Lubensky<sup>(26)</sup> and by Daoud and Joanny<sup>(27)</sup> for branched polymers in the dilute limit.

An inspection of the expression for D shows that it has two important characteristics: First, it depends strongly on the spatial dimension d, and second, there exists a critical dimension  $d_c = 8$  at which D takes on the value D = 4 corresponding to the ideal random model, i.e., the model with no correlations. Above the critical dimension correlations due to the excluded volume effect are unimportant and D does not change any longer. The value  $d_c = 8$ , is the well-known result for branched polymers and lattice

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animals.<sup>(17,25)</sup> Lattice animals<sup>(8,17-19)</sup> are random configurations obeying equilibrium statistics and can be considered as a model for isolated random fractals. Predictions of the Flory formula, (8), are in excellent agreement with the known results for lattice animals. Recently, Schaefer and Keefer<sup>(48)</sup> have used small-angle X-ray scattering technique to measure D for silica condensation polymers and find that their result is in agreement with (8).

#### 4.2. Compensated State

In the previous section we investigated the conformation of a single isolated cluster and found that it has a highly extended structure with a relatively low D. In this section we consider the effects which tend to increase D. Let us begin by studying the conformation of one large cluster of size N in the presence of other clusters having a distribution of cluster sizes. Clearly, the large cluster does not feel the presence of the other clusters as long as their average size is small compared to the size of the large cluster. On length scales much larger than the radius of an average cluster the cluster interacts only with itself and is swollen. However, as pointed out by Edwards<sup>(49)</sup> and de Gennes<sup>(25)</sup> in the context of polymers, as the average size of the other clusters increases they tend to screen the excluded volume effect experienced by the large cluster. If the average cluster size is M, then the ("screened") repulsive free energy for the large cluster is written<sup>(25)</sup>

$$F_{\rm rep} = \frac{1}{M} \frac{N^2}{R^d} \tag{9}$$

Note that as long as M is independent of N, i.e.,  $M \sim O(1)$ , (9) is identical to (6) and presence of other clusters does not change the conformation of the large cluster. However, when M does depend on N, then the screening effect changes the fractal dimension of the cluster.

The best examples of systems with a distribution of cluster sizes are percolation and thermal critical phenomena, e.g., Ising model, Potts model, etc. Under equilibrium conditions, in a critical system consisting of a distribution of cluster sizes, the mean cluster size diverges at the critical point. Simple scaling arguments give that  $M \sim N^{\nu/D^{\nu}}$ , where  $\gamma$  and  $\nu$  are the susceptibility and the correlation length exponents, respectively. In the spirit of the Flory theory,<sup>(22-27)</sup> we use the mean-field values  $\gamma = 1$ ,  $\nu = 1/2$ , and D = 4 to find  $M \sim N^{1/2}$  at the critical point. Substituting this result in (9) we find

$$F_{\rm rep} = N^{3/2} / R^d \qquad (\text{critical point}) \tag{10}$$

Adding (10) and (5) together we find the total free energy at the critical point

$$F = \frac{R^2}{N^{1/2}} + \frac{N^{3/2}}{R^d}$$
 (critical point) (11)

Minimization of (11) with respect to R gives

$$N \sim R^D$$
, with  $D = \frac{d+2}{2}$  (compensated state) (12)

This result is expected to hold for any random critical cluster in the presence of a distribution of clusters whose mean cluster size diverges as  $N^{1/2}$ . The best examples are critical clusters in thermal critical phenomena.

We can determine the upper critical dimension for critical clusters by letting D = 4 in (12). The result,  $d_c = 6$ , is well established for percolation,<sup>(25,26)</sup> but appears to disagree with the result  $d_c = 4$  for thermal critical phenomena.<sup>(50)</sup> In order to resolve this problem, let us first point out that the usual upper critical dimension,  $d_c = 4$ , refers to the dimension above which thermal correlations in a critical system become unimportant. Above  $d_c = 4$ , the thermal and the magnetic scaling powers take on their mean field values<sup>(50)</sup> ( $y_T = 2$ ,  $y_h = 3$ , respectively), indicating that thermal fluctuations above four dimensions are of the mean field type. In contrast to the usual critical exponents, deviation of D from its mean field value (i.e., D = 4) is a measure of the geometrical correlations (i.e., excluded volume) in the system. Therefore the result  $d_c = 6$  for the fractal dimension implies that although thermal fluctuations above d = 4 are unimportant, geometrical correlations are nontrivial.

In order to show how D can vary above d = 4, in analogy with percolation,<sup>(4,20)</sup> we introduce a ghost field, H, which couples to active bonds in a critical cluster (see Section 2). The essential difference between this field and the usual mgnetic field is that the magnetic field is conjugate to the order parameter,<sup>(50)</sup> whereas the ghost field is conjugate to the geometrical size of the cluster, i.e., N.<sup>(8)</sup> Therefore, for  $d \leq 4$  the magnetic scaling power  $y_H$  calculated from the ghost field is identical to the usual  $y_h$ , whereas for d > 4 this field determines the fractal dimension D (which does not coincide with  $y_h$  above d = 4). Renormalization group results<sup>(29)</sup> for the Ising model in six dimensions can be interpreted as giving  $D = y_H = 4$ , in agreement with Eq. (12).

The above results show that the presence of many clusters and the divergence of the mean cluster size at the critical point leads to the screening of the excluded volume effect and an increase in D. Therefore, we call this the *compensated state* of a random fractal, because in this state the excluded volume effect is compensated for by the screening effect.

Although the Flory theory properly takes the screening into account,  $^{(25,26)}$  it ignores the details that depend on the statistics of the system, e.g., *n* and *q* in the *n*-vector and the *q*-state Potts models, respectively. However, as can be seen from Table I, these details do not seem to be important, because systems belonging to many different universality classes appear to have approximately the same value of *D*. Therefore, it is important to ask the question: Why do all critical clusters have almost the same value of *D*?

In order to answer this question, let us introduce two types of length scales in the problem: First, the thermal correlation length  $\xi$  which diverges with the usual thermal correlation length exponent at the critical point, and second, the geometrical length scale of the cluster R, e.g., the cluster diameter, which also diverges at the critical point, but with the exponent 1/Dand as a function of the cluster size N [see Eq. (2)]. Away from  $T_c$ , there exist large clusters in the system whose geometrical length R is large compared to the thermal correlation length  $\xi$ . The reason is that in contrast to  $\xi$  which is strongly temperature dependent, R depends only on the geometrical correlations that are mainly function of the spatial dimension. Therefore, only a few bonds are needed to join these large clusters together to form an infinitely large critical cluster at the critical point. As the critical point is approached, these bonds are formed by the increase in thermal correlations between the elements belonging to neighboring clusters. Thus, the only effect of thermal correlations is to produce a few links between existing clusters and join them together. The overall geometrical shape of the critical cluster is independent of the details of the thermal interactions and depends primarily on the dimensionality d and the screening effect discussed above. Quantities which determine the symmetry of the order-parameter, e.g., n in the n-vector model or q in the q-state Potts model, act as symmetrybreaking perturbations which produce slight differences in the values of the fractal dimension of the different models. If these were not present, then all critical phenomena would have had exactly the same fractal dimensions. This result provides a clear physical interpretation for the Suzuki universality hypothesis.<sup>(21)</sup>

The above description of the compensation of the repulsive interactions was based on the concept of screening. Another way to reduce the strength of the repulsive interactions is by introducing attractive interactions between the elements of an isolated cluster.<sup>(27,51)</sup> In order to discuss this case we write the repulsive free energy (5) in its generalized form<sup>(27,51)</sup> as a power series in the density  $\rho = N/R^d$  as

$$F_{\rm rep} = N(w_2 \rho + w_3 \rho^2 + \dots)$$
(13)

where  $w_2, w_3, \dots$  are the virial coefficients which include the effect of

interactions between pairs, triplets, etc. The extended state discussed in Section 4.1 corresponds to the limit where the first term in (13) dominates. However, in the presence of attractive interactions  $w_2$  vanishes and it becomes necessary to keep the  $w_3$  term on the right-hand side of (13).<sup>(27,51)</sup> The point at which the second virial coefficient  $w_2$  vanishes is called the Flory theta point in polymer science.<sup>(22,24)</sup> Therefore, at the compensation point the repulsive free energy is<sup>(27,51)</sup>

$$F_{\rm rep} = N^3/R^{2d}$$
 (compensation point) (14)

Minimization of the total free energy

$$F = \frac{R^2}{N^{1/2}} + \frac{N^3}{R^{2d}} \tag{15}$$

with respect to R gives

$$N \sim R^D$$
, with  $D = \frac{4(d+1)}{7}$  (compensated state) (16)

This result was first obtained by Daoud and Joanny<sup>(27)</sup> for the conformation of an isolated branched polymer at the theta point. In analogy with the previous results for the compensated state, (12), this expression also predicts  $d_c = 6$ .

In summary, at the compensation point, which can be brought about by either geometrically screening the excluded volume effect, or, by compensating it by introducing attractive interactions, the fractal dimension D is increased. Large clusters at the critical point in thermal phase transitions and polymers at the theta point are examples of random fractals in the compensated state.

### 4.3. Collapsed State

Let us now study the conformation of a random fractal, first, among other clusters in a system below the critical point, and second, in isolation, but in the presence of attractive interactions such that the second virial coefficient  $w_2 < 0$ . We shall find that in both cases the fractal object has a highly compact, or globular, conformation; we call this the *collapsed state* of the random fractal in analogy with the collapsed state of polymers.

Consider a large cluster of size N among other clusters in a system below its critical point. Under this condition, the mean cluster size M is not a critical object and the relation  $M \sim N^{1/2}$  does not hold. The reason is that owing to the mutual excluded volume effect, in concentrated solutions clusters can grow only by joining to other clusters, i.e., by nucleation. This means that we can no longer distinguish between a characteristic cluster size N and a mean cluster size M; the whole system is now composed of clusters having roughly the same size, i.e.,  $M \simeq N$ .

In order to determine the conformation of the clusters under this condition, we follow the approach used by Daoud and Joanny<sup>(27)</sup> to study the conformation of a branched polymer in a monodispersed melt, i.e., a solution of polymers of the same size, and write the total free energy of the system as

$$F = \frac{R^2}{N^{1/2}} + R^d \left[ \frac{1-\phi}{N} \log(1-\phi) \right]$$
(17)

where  $\phi$  is the volume fraction occupied by the elements of the fractal of size N. Assuming that  $M \simeq N$ , and minimizing (17) with respect to R one finds

$$N \sim R^D$$
, with  $D = d$  (collapsed state) (18)

Comparing this result with the purely random fractal, D = 4, we find  $d_c = 4$ . This implies that this type of random clusters are compact up to d = 4. For this reason, we call this state of the random fractals the *collapsed state*.

Now let us consider the case of an isolated fractal below the compensation point, i.e., when the attractive interactions are dominant. In this case the second virial coefficient,  $w_2$ , is negative and cannot be ignored in (13). Conformation of the fractal is determined by the balance between the first two terms in (13). The result is identical to (17) with  $d_c = 4$ , as before. Therefore, the conformation of a single random fractal below the compensation point is the same as that of a large cluster below the critical point.

#### 5. SUMMARY AND CONCLUSIONS

Conformation of a random fractal formed in an equilibrium process depends on the strength of the excluded volume effect. When the excluded volume effect is the only dominant force in the system, e.g., for an isolated cluster without attractive interactions, the fractal has an extended or highly ramified structure. We call this the *extended state* of a random fractal. The excluded volume effect can be reduced either by introducing other clusters in the system, or, by introducing attractive interactions between the elements of an isolated cluster. Within the Flory theory, both effects lead to more compact structures having an upper critical dimension of six. We call this the *compensated state* of a random fractal. In particular, critical clusters in thermal phase transitions have this type of structure, i.e., they are in the compensated state. Therefore, from the point of view of the geometry of their clusters, all critical phenomena can be regarded as belonging to a single "grand" universality class. One consequence of this result is that the upper

critical dimension for the fractal dimension of critical clusters is 6 instead of 4. Finally, when the attractive interactions fully compensate the repulsive interactions the random cluster has a *collapsed* structure, i.e., D = d for low dimensions, but for higher dimensions (d > 4) it has the structure of an ideal random fractal without excluded volume, with D = 4.

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