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## Are Random Fractal Clusters Isotropic?

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We have studied the shape of large clusters in the lattice-animal, percolation, and growing-percolation models. By calculating the radius of gyration tensor we find that in these models the clusters have an anisotropic shape. The results suggest that the critical droplets in related isotropic equilibrium models, such as the Ising model, may also be anisotropic. We have also determined the leading nonanalytic correction-to-scaling exponent by analyzing the anisotropy data and find that for percolation in two dimensions  $\theta \approx 0.47$ .

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In this Letter we address the following question: Is the shape of random scale-invariant clusters isotropic? The answer to this question is of considerable interest from both practical and fundamental points of view. The behavior of a wide variety of condensed systems, from polymer solutions and colloidal suspensions to smoke and dust, is determined by the conformation of the large clusters or aggregates that they are made of. Filtration, sedimentation, adsorption, transport, and rheological properties of such systems depend crucially on whether these clusters have a spherically symmetric shape or are anisotropic.<sup>1</sup> The existence of structural anisotropy in systems with isotropic coupling would represent a puzzling anomaly requiring theoretical explanation.

The basic interest in cluster shapes,<sup>2</sup> however, stems from the fact that the classical droplet models<sup>3</sup> in theories of phase transitions are based on the assumption of spherically symmetric clusters. In addition, models allowing only for spherical droplets contain an essential singularity at first-order phase transitions.<sup>3</sup> Theoretical studies<sup>2,4</sup> indicate that the details of this singularity are modified if deviations from sphericity are taken into account. Moreover, being the ratio of two diverging lengths, anisotropy is a quantity which is independent of the leading exponent and could allow

an unbiased determination of the correction-to-scaling exponent. According to the scaling theory of continuous phase transitions, the ratio of the amplitudes of two quantities diverging with the same leading exponent at the critical point is universal. Since anisotropy is the ratio of two such quantities, on the basis of the scaling theory, it is expected to be a universal parameter.

In this Letter we report the results from the investigations of the cluster shapes in three models: lattice animals,<sup>5</sup> percolation clusters,<sup>6</sup> and growing percolation clusters.<sup>7,8</sup> Lattice animals represent clusters in an equilibrium model away from the critical point ( $T = \infty$ ), percolation clusters at the percolation threshold are the *sin quo non* of the critical clusters in phase transitions, and growing percolation clusters represent a kinetic process which is closely related both to equilibrium models (e.g., percolation) as well as to a new class of kinetic aggregation phenomena<sup>9</sup> (e.g., diffusion-limited aggregation).

The anisotropy of a given  $N$ -site cluster can be determined from its radius of gyration tensor  $R_{ij}^2$  ( $i, j = x, y$  in  $d = 2$ ), by diagonalization of the tensor and calculation of the principal radii of gyration. The anisotropy  $A_N$  of an  $N$ -site cluster is then defined to be the ratio  $R_{N, \min}^2 / R_{N, \max}^2$ , where  $R_{N, \min}^2$  and  $R_{N, \max}^2$  are the smaller and the larger eigenvalues of

the radius of gyration tensor, respectively. The quantity  $A_N$  when properly averaged over all clusters of size  $N$  is denoted by  $\langle A_N \rangle$  and is an estimate of the anisotropy of  $N$ -site clusters in the ensemble. We allow for the leading nonanalytic and analytic correction to scaling and write the asymptotic form of the principal radii of gyration as

$$\langle R_{N,\sigma}^2 \rangle = r_\sigma N^{2\nu} (1 + a_\sigma N^{-\theta} + b_\sigma N^{-1} + \dots), \quad (1)$$

$\sigma = \min$  or  $\max$ . The coefficients  $r_\sigma$ ,  $a_\sigma$ , and  $b_\sigma$  are independent of  $N$ ,  $\nu$  is the leading scaling exponent and is equal to the inverse of the fractal dimension,  $N^{-\theta}$  is the leading *nonanalytic* correction-to-scaling term, and  $N^{-1}$  represents the leading *analytic* correction-to-scaling term. Using expression (1) we can write the anisotropy as

$$\langle A_N \rangle = \langle R_{N,\min}^2 / R_{N,\max}^2 \rangle = A_\infty [1 + (a_{\min} - a_{\max}) N^{-\theta} + (b_{\min} - b_{\max}) N^{-1} + \dots], \quad (2)$$

where  $\langle \dots \rangle$  denotes an average over all  $N$ -site clusters.

*Lattice animals.*—The statistics of connected clusters of sites or bonds on regular lattices—commonly referred to as lattice animals<sup>5</sup>—has been a subject of considerable recent interest because of its applications in a variety of diverse fields including the theory of chemical graphs, the cell-growth problem, homogeneous turbulence in fluids, spinodal decomposition, and dilute branched polymers.

An effective method for a systematic study of the asymptotic scaling behavior of lattice animals is the exact series enumerations. Recently, Privman and Fisher<sup>10</sup> (PF) have developed a technique for the determination of the correction-to-scaling exponents using exact-series-enumerations data based on cancellation of the leading nonanalytic terms. This method has been successfully applied to the analysis of the radii of lattice animals and percolation clusters.<sup>11</sup> The PF method is particularly well suited for the study of the anisotropy because the  $\langle A_N \rangle$  series *only* contains correction-to-scaling terms and allows an unbiased calculation of the exponents. We have enumerated all site animals of up to size 15 on a square lattice and have determined  $\langle R_{N,\min}^2 \rangle$ ,  $\langle R_{N,\max}^2 \rangle$ , and  $\langle A_N \rangle$ . Using the PF method we first analyzed the  $\langle R_{N,\min}^2 \rangle$  and the  $\langle R_{N,\max}^2 \rangle$  series assuming the scaling form (1) with  $\nu = 0.640 \pm 0.005$ .<sup>11</sup> The resulting biased estimate of  $\theta = 0.88 \pm 0.06$  for the leading nonanalytic correction-to-scaling exponent agrees with the previous estimate<sup>11</sup> for  $\theta$  based on the radius of lattice animals. As noted above, anisotropy does not involve the leading exponent and the analysis of the  $\langle A_N \rangle$  series in principle provides an unbiased estimate of  $\theta$ . In applying the PF method to the series for  $\langle A_N \rangle$  we found that the correction-to-scaling exponent is  $1.01 \pm 0.04$ . This result implies that either the amplitude of the  $N^{-\theta}$  term in (2) vanishes or is very small, and in this way only the leading *analytic* correction-to-scaling term remains. Since the  $N^{-\theta}$  term does not appear in (2), the asymptotic value of the anisotropy,  $A_\infty$ , can be simply estimated from the intercept of a plot of  $\langle A_N \rangle$  against  $1/N$ . Such a plot is shown in Fig. 1, from which we estimate that for lattice animals  $A_\infty \approx 0.29$ . This result indicates that large two-dimensional lattice animals have an anisotropic shape, even though the model is purely isotropic.

In addition to lattice animals, we have enumerated all animals without loops (i.e., trees) of up to size 16 and have applied exactly the same analyses to these series. We found that if we use the same  $\nu$  as for lattice animals the radius of gyration,  $\langle R_N^2 \rangle$ , and the principal radii of gyration,  $\langle R_{N,\min}^2 \rangle$  and  $\langle R_{N,\max}^2 \rangle$ , give the same correction-to-scaling exponent  $\theta$ . This is further support for the universality of animals with and without loops.<sup>5</sup> Similarly, the analysis of the series for the anisotropy of trees shows that again the  $N^{-\theta}$  term in (2) drops out. A plot of  $\langle A_N \rangle$  vs  $1/N$  for trees is also shown in Fig. 1. From this figure we can assume that animals with and without loops have the same degree of anisotropy. We have also calculated the fluctuations of the anisotropy, by determining  $\sigma(A_N)$ , which is the standard deviation of the quantity  $R_{N,\min}^2 / R_{N,\max}^2$ . The results for lattice animals are shown in Fig. 1, where it can be seen that the fluctuations of the anisotropy are essentially independent of  $N$ .

*Percolation.*—The study of percolation clusters pro-

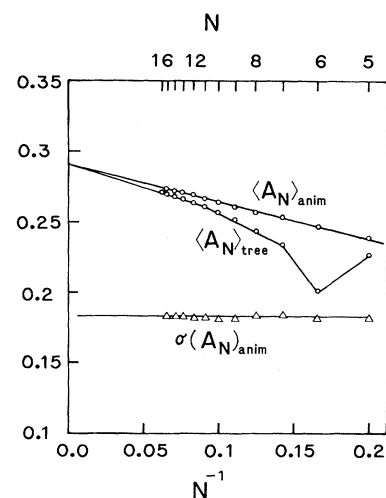


FIG. 1. Dependence of the average anisotropy of lattice animals,  $\langle A_N \rangle_{\text{anim}}$ , and lattice animals without loops (trees),  $\langle A_N \rangle_{\text{tree}}$ , in two dimensions, on  $1/N$ , where  $N$  is the number of sites in the cluster. Extrapolation to  $N \rightarrow \infty$  gives approximately the value 0.29 for the anisotropy of cluster shapes in both models. The fluctuation of the anisotropy of lattice animals,  $\sigma(A_N)_{\text{anim}}$ , is also plotted against  $1/N$ .

vides an additional possibility to investigate the shape of random fractal clusters. A simple way to generate percolation clusters is a process originally proposed by Alexandrowicz<sup>7</sup> and later in a modified form by Grassberger.<sup>8</sup> The process is started with a seed particle (occupied site) on a lattice. One of its nearest neighbors (perimeter sites) is chosen randomly, and if a random number  $r$  attributed to this site is less than a previously fixed parameter,  $p$ , this empty site is occupied and becomes part of the growing cluster. If  $r > p$ , the given perimeter site is discarded and is not considered in the future as an available perimeter site. In the next step one of the available perimeter sites of the new cluster is considered for occupation, and so forth. It is possible to generate very large clusters with use of this method and therefore extrapolation of the results to  $N \rightarrow \infty$  is expected to yield reliable estimates for the anisotropy.

The above process can go on indefinitely if our seed particle happens to belong to an infinite cluster, or terminate at a particular cluster size if the number of available perimeter sites becomes equal to zero. Accordingly, two kinds of percolation clusters can be generated by this method. The fractal dimensions of the *growing* percolation clusters (nonequilibrium clusters which can grow further because of the available perimeter sites) have been shown to be the same as the equilibrium percolation clusters.<sup>8,12</sup> Clusters which are obtained when the growth process terminates are ordinary *equilibrium* percolation clusters; therefore, the use of the Alexandrowicz method<sup>8</sup> allows us to study the shape of both equilibrium and nonequilibrium percolation clusters.

In order to get a good estimate of the anisotropy of an  $N$ -site *growing* percolation cluster,  $\langle A_N \rangle_{gr}$ , we have generated 30 000 clusters of up to 2500 sites each using the Alexandrowicz method for  $p = p_c = 0.5928$ , where  $p_c$  is the site percolation threshold on the square lattice. The anisotropy was determined for selected values of  $N$  from the ratio of the eigenvalues of the radius of gyration tensor. We also calculated the fluctuations of the anisotropy,  $\sigma(A_N)_{gr}$ . The results are presented in Fig. 2. If we plot  $\langle A_N \rangle_{gr}$  vs  $N^{-0.47}$  the data lie along a straight line, indicating that the exponent  $\theta$  in the correction-to-scaling term of the expression for the anisotropy, (2), has a value  $\theta \approx 0.47$ . We have also plotted  $\log(\langle A_N \rangle_{gr} - \langle A \rangle_\infty)$  against  $\log(N)$ . The slope of the straight line drawn through the data provided another estimate for the correction-to-scaling exponent, giving  $\theta \approx 0.47$ . While  $\langle A_N \rangle$  contains a nonanalytic correction-to-scaling term the fluctuations in  $\langle A_N \rangle_{gr}$  seem to be independent of  $N$  (see Fig. 2). The actual values  $A_{\infty,gr} \approx 0.485$  and  $\sigma(A_{\infty,gr}) \approx 0.18$  suggest that the shape of large growing percolation clusters is typically anisotropic and the fraction of the nearly circular clusters is relatively

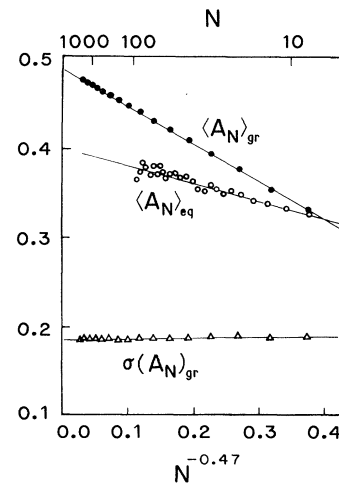


FIG. 2. The average anisotropy of equilibrium ( $\langle A_N \rangle_{eq}$ ) and growing ( $\langle A_N \rangle_{gr}$ ) percolation clusters in two dimensions as a function of  $N^{-\theta}$ , where  $\theta$  is the correction-to-scaling exponent. We used  $\theta = 0.47$  which gave the best straight-line fit to the data. The fluctuation in the anisotropy of growing percolation clusters,  $\sigma(A_N)_{gr}$ , is also plotted against  $N^{-0.47}$ .

small.

In the next series of simulations 80 000 *equilibrium* percolation clusters of sizes  $10 < s < 100$  were generated for  $p = p_c$  with use of the same method and calculation of the anisotropy of a cluster after the growth of the given cluster had been terminated. The statistics was not as good in this case as it was for the growing percolation clusters because the data obtained for a cluster contributed to the statistics only for the particular size at which the growth stopped. Correspondingly, our results, presented in Fig. 2, show a larger uncertainty. On the other hand, we can conclude from Fig. 2 that the equilibrium clusters are more anisotropic than the growing percolation clusters and the very large ordinary percolation clusters have an anisotropy approximately equal to 0.4.

A long-standing controversy exists in the studies of corrections to scaling in percolation in two dimensions, because existing estimates of  $\theta$  spread over two different ranges.<sup>13</sup> The lower values of  $\theta \approx 0.47$ – $0.49$  have been suggested by the generating-function and the PF-method analyses of series for various percolation properties.<sup>13</sup> The higher range of  $\theta \approx 0.6$ – $0.7$  has been obtained in other numerical studies.<sup>14</sup> According to the Aharony-Fisher theory,<sup>15</sup> the higher value is a next-to-leading correction term arising from the mixing of nonlinear scaling fields, and is given by  $\theta' = 1 - \sigma = \frac{55}{91} = 0.601$ . . . , where  $\sigma$  is the percolation gap exponent. The result  $\theta \approx 0.47$  obtained here suggests that the lower value of  $\theta$  is the leading nonanalytic correction-to-scaling exponent.

In addition, growing percolation and equilibrium percolation seems to have the same value of the correction-to-scaling exponent. This is perhaps not unexpected in view of the fact that the leading exponent (i.e., the fractal dimension) is known to be the same in the two models.<sup>8,12</sup>

Our results concerning the anisotropy of cluster shapes are both novel and interesting, because in contrast to certain models in which the clusters are trivially isotropic or anisotropic, there is no explanation available for the presence and magnitude of anisotropy in the lattice animal, percolation, and growing-percolation models. For example, diffusion-limited aggregates are expected to be isotropic because of the growth mechanism which is based on a spherically symmetric diffusing field. On the other hand, large aggregates in the cluster-cluster aggregation model are formed by coalescence of, say, two large clusters and are anisotropic.<sup>16</sup>

The existence of anisotropic cluster shape in percolation is significant because of the close connection between this model and a variety of other models exhibiting critical behavior. Since percolation is the  $q \rightarrow 1$  limit of the  $q$ -state Potts model,<sup>17</sup> we expect that the critical droplets in the Ising model (and the Potts model in general) are not spherically symmetric as well. Moreover, there are many other isotropic models in which the anisotropy of the cluster shapes could also be investigated.

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<sup>1</sup>C. J. S. Petrie, *Rheology* (Pitman, London, 1979).

<sup>2</sup>R. K. P. Zia and D. J. Wallace, *Phys. Rev. B* **31**, 1624 (1985).

<sup>3</sup>M. E. Fisher, *Physics* (Long Island City, N.Y.) **3**, 255 (1967).

<sup>4</sup>J. S. Langer, *Ann. Phys. (N.Y.)* **41**, 108 (1967).

<sup>5</sup>T. C. Lubensky and J. Issacson, *Phys. Rev. A* **20**, 2130 (1979).

<sup>6</sup>D. Stauffer, *Introduction to Percolation Theory* (Taylor and Francis, Bristol, 1985).

<sup>7</sup>Z. Alexandrowicz, *Phys. Lett.* **80A**, 284 (1980).

<sup>8</sup>P. Grassberger, *Math. Biosci.* **62**, 157 (1983), and to be published.

<sup>9</sup>*Kinetics of Aggregation and Gelation*, edited by F. Family and D. P. Landau (North-Holland, Amsterdam, 1984).

<sup>10</sup>V. Privman and M. E. Fisher, *J. Phys. A* **16**, L295 (1983).

<sup>11</sup>A. Margolina, F. Family, and V. Privman, *Z. Phys. B* **54**, 321 (1984).

<sup>12</sup>F. Family and T. Vicsek, *J. Phys. A* **18**, L75 (1985).

<sup>13</sup>J. Adler, M. Moshe, and V. Privman, in *Percolation Structures and Processes*, edited by G. Deutcher, R. Zallen, and J. Adler (Hilger, Bristol, 1983), p. 397.

<sup>14</sup>A. Margolina, D. Stauffer, H. E. Stanley, and H. Nakanishi, *J. Phys. A* **17**, 1683 (1984).

<sup>15</sup>A. Aharony and M. E. Fisher, *Phys. Rev. B* **27**, 4394 (1983).

<sup>16</sup>H. G. E. Hentschel, in Ref. 9, p. 117.

<sup>17</sup>F. Y. Wu, *Rev. Mod. Phys.* **54**, 235 (1982).