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Universality and cluster structures in continuum models of percolation with two different radius distributions

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Abstract. The percolation thresholds and the fractal cluster structures for continuum models *of percolation with uniform (CM1) and variable radius (CM2) distributions of discs and* spheres are investigated and compared with the results of ordinary lattice percolation. Configurations of up to 250 000 discs (2 dimensions) and 100 000 spheres (3 dimensions) are numerically simulated. In two dimensions we find distinctly different percolation concentrations for models CM1 and CM2. In the three-dimensional systems the percolation concentrations for both models cannot be distinguished within our limits of accuracy. The fractal dimensions of the cluster hull, surface and volume are the same as in the corresponding lattice models. The Harris criterion for the continuum percolation problem is confirmed by our simulation.

1. Introduction

Continuum percolation models have attracted increasing attention as the physical realization of percolating structures which are insensitive with respect to the microstructure (e.g. the lattice of a solid). Examples for such systems are the pore space in sedimentary rocks (Thompson *et al* 1987) and the cluster structures arising in the droplet model of first-order phase transitions (Binder and Stauffer 1976). In the latter example the phase transition is determined by the creation of critical nuclei of the stable phase and the growth of these nuclei until they impinge to one another forming larger and larger clusters. The largest cluster percolates in the critical stage of the transition. In the process of homogeneous nucleation and isotropic growth the clusters consist of overlapping discs (in two dimensions, 2D) or spheres (3D) with variable radii.

Lattice percolation models have been investigated frequently and the critical exponents at the percolation threshold and the fractal properties of clusters are suggested to be universal for various models and lattices in two and three dimensions (for a review see Stauffer 1985). Less work has been done for continuum models. Two typical radius distributions $W(r)$ for the cluster forming nuclei are of interest. In the first model (CM1), all discs or spheres are of one and the same size R_0, $W(r) = n_0\delta(r - R_0)$. n_0 denotes the density of nuclei. This model describes the cluster structure growing at first-order phase transitions where nucleation takes place instantaneously before the onset of growth. The second model (CM2) is characterized by a constant size distribution $W(r) = W_0$, $r \leq R_0$, which arises in the process of homogeneous

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nucleation and growth (Christian 1965). The model CM2 applies to various phase transformations in materials science and metallurgy.

Experimentally, percolating structures are verified by the observation of quenched silica aerogels (Devreux *et al* 1990), the aggregation of colloidal gold particles (Liu *et al* 1990), and high pressure phase separation in solutions (Lorenz *et al* 1991).

The percolation threshold, critical exponents and cluster structures for CM1 are determined by series expansion (Domb 1972, Haan and Zwanzig 1977), Monte Carlo simulation (Roberts 1967, Pike and Seager 1974, Gawlinski and Stanley 1981), renormalization group approach (Vicsek and Kertész 1981, Kertész and Vicsek 1982) and even more sophisticated methods (Rosso 1989). The results lead to the conjecture that the critical exponents at the percolation point and the fractal cluster structures are the same as in the lattice models and the continuum and lattice percolation models belong to one and the same universality class. The non-universal behaviour of specific quantities like the ratio of the susceptibilities above and below the percolation threshold, however, is in discussion (Balberg 1988, Lee 1990).

For a correct calculation of critical properties the percolation threshold density has to be determined with high accuracy. It is convenient to use the volume fraction Φ of the growing phase as the density parameter in order to compare the results for continuum models with different radius distributions. Φ is related to the model parameters by $\Phi = 1 - \exp\{-c_1 n_0 R_0^d\}$ and $\Phi = 1 - \exp\{-c_2 W_0 R_0^{d+1}\}$ for CM1 and CM2, respectively (Christian 1965). d denotes the spatial dimension and the c_i are constants which depend on the model and the shape of the nuclei. Several approximation techniques applied to the models CM1 and CM2 exhibit nearly similar threshold values Φ_c in two (Pike and Seager 1974, Kertész and Vicsek 1982) as well as in three dimensions (Pike and Seager 1974). It is not clear, however, whether or not the threshold values are independent of the radius distribution. Phani and Dhar (1984) have shown that at least for distributions of discs with two different radii R_1 and R_2 the threshold value for the onset of percolation differs distinctly from that of equally sized discs as far as $R_2/R_1 < 0.35$. Kertész and Vicsek (1982) suggested the validity of the Harris criterion for the continuum percolation models considering the variation of the radius distribution as an additional randomness. Then, the critical percolation exponents and the fractal dimensions should be equal for the models CM1 and CM2. It is of special interest to answer the question if the threshold value Φ_c represents a further universal quantity.

In the present paper we investigate the onset of percolation in the continuum models discussed above by a numerical simulation. We concentrate on the comparison of universal and non-universal quantities at Φ_c for the models CM1 and CM2 in two and three dimensions and the fractal properties of the cluster structures.

2. Numerical procedure

The simulation of the random system is performed using an extended and refined algorithm discussed previously (Orgzall and Lorenz 1988, Lorenz 1989, Orgzall and Lorenz 1992). A configuration is generated by randomly placing discs or spheres into a unit volume until the volume fraction Φ is covered. The radius distribution is chosen according to CM1 or CM2. The cluster structure is evaluated numerically using the technique of Hoshen and Kopelman (1976) extended to continuum problems by Gawlinski and Stanley (1981). Thus, systems of up to 250 000 discs and 100 000 spheres

were simulated on a RISC processor system. After labelling the various clusters in the system, the volume and the surface of each individual cluster are determined numerically within a relative error of less than 0.5%. In the two-dimensional models the outer surface (hull) of the clusters is calculated by an exact integration. The percolation threshold is determined by considering the extension of the largest cluster in the configuration using periodic as well as non-periodic boundary conditions.

In order to characterize the fractal properties, a measure of the linear 'size' of the cluster has to be defined. Although this definition is somewhat arbitrary, there exist arguments (Stauffer 1985) that in the scaling region (i.e. for large cluster sizes) the various definitions are equivalent. In our calculations three different measures were used and compared as follows.

(i) The average over the maximum extent of the cluster along the coordinate axes

$$R_m = \frac{1}{2d} \sum_{j=1}^{d} (x_{j,\max} - x_{j,\min}) \tag{1}$$

where d denotes the dimension of the system.

(ii) The radius of gyration of the cluster hull (2D)

$$R_g = \frac{1}{S} \int_{\text{hull}} \left[\sum_{j=1}^{d} (x_j - \bar{x}_j)^2 \right]^{1/2} \mathrm{d}s \tag{2}$$

where S and $\bar{x}_j$ denote the size and the centre of mass coordinates of the hull, respectively.

(iii) The mean square radius given by

$$R_s = \left[\frac{1}{V} \int_{\text{vol}} \left\{ \sum_{j=1}^{d} (x_j - \bar{x}_j)^2 \right\} \mathrm{d}v \right]^{1/2}. \tag{3}$$

$\bar{x}_j$ is now the centre of mass of the cluster volume V.

We have compared the values calculated according to the definitions (i) to (iii) and found a proportionality of R_m, R_g and R_s for large cluster sizes in agreement with the scaling prediction. In the following the suffix is suppressed and the cluster size is denoted by R.

3. Percolation threshold Φ_c

The best known values Φ_c for the onset of percolation have been determined for the model CM1 in two dimensions as $\Phi_c^{(1)} = 0.6766 \pm 0.0005$ (Rosso 1989) and in three dimensions as $\Phi_c^{(1)} = 0.294 \pm 0.003$ (Lee 1990). In the present calculation we estimate the percolation densities simulating systems up to 250 000 discs and 100 000 spheres. Φ_c is determined by extrapolating the percolation densities for finite systems of length L to the infinite system according to the scaling law (Gawlinski and Stanley 1981, Stauffer 1985)

$$|\Phi_c(L) - \Phi_c(\infty)| \sim L^{-1/\nu} \tag{4}$$

where ν is the critical exponent at Φ_c and L is the length of the system.

$\Phi_c(L)$ is derived from the percolation probability $P_L(\Phi)$ for fixed L. This probability is defined as the ratio of the number of percolating systems to the total number of

simulations for given Φ and exhibits a sigmoidal shape between 0 and 1. From a large number of computer experiments ($N \approx 2000$) for fixed L and Φ the data are derived and fitted to a smooth function $P_L(\Phi)$. Testing various functions within the limits $0.05 \leqslant P_L(\Phi) \leqslant 0.95$ the best fit is obtained using

$$P_L(\Phi) = \tfrac{1}{2}\{1 + \tanh^{-1}[b(\Phi - \Phi_c)]\}. \tag{5}$$

The $\tanh^{-1}$ denotes the inverse hyperbolic tangent which shows the sigmoidal dependence on Φ. Equation (5) is a convenient fit function since it contains only two fit parameters. Comparing the results with those from alternative functions containing up to four parameters we find the same values of $\Phi_c(L)$ within the statistical uncertainty. With (5) the percolation threshold $\Phi_c(L)$ is derived at $P_L(\Phi_c) = 0.5$. It is assumed that this definition does not influence the extrapolated value $\Phi_c(\infty)$. For a better extrapolation $\Phi_c(L)$ is determined taking into account periodic as well as non-periodic boundary conditions. Naturally, systems with periodic boundary conditions yield a larger $\Phi_c(L)$ and the difference vanishes for $L \to \infty$.

In order to test the accuracy of the procedure described above we investigated the model CM1 in two dimensions. The results for $\Phi_c(L)$ are given in figure 1 (lower curve set). The extrapolation according to (4), however, is problematic since the function (4) yields a nearly equally good fit to the numerical data for a broad set of pairs $\{\Phi_c, \nu\}$. The corresponding curves $\Phi_c(\nu)$ are shown in figure 3(a) for periodic and non-periodic boundary conditions. Obviously, the curves for both types of boundary conditions have a common intersection point which is chosen as the best fit of the parameters Φ_c and ν to equation (4). This procedure yields $\Phi_c(\infty) = 0.6764 \pm 0.0009$ and $\nu = 1.37 \pm 0.07$. The value of Φ_c is in excellent agreement with that of Gawlinski and Stanley (1981) (0.676 ± 0.003) and the most accurate determination by Rosso (1989) with a gradient percolation technique, $\Phi_c = 0.6766 \pm 0.0005$. Although the variance in

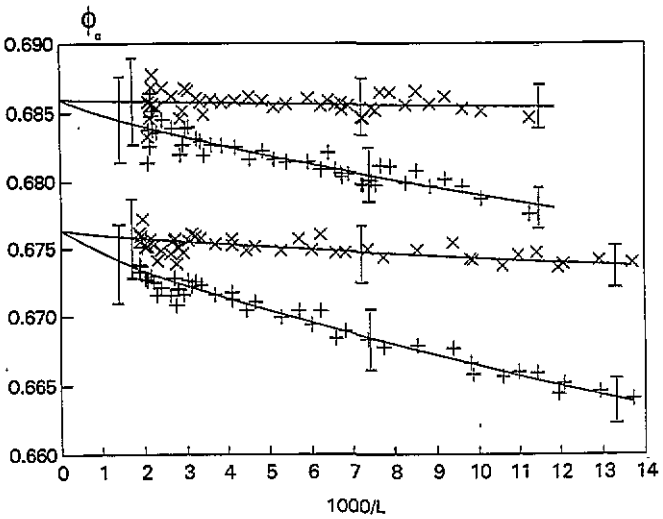

Figure 1. Finite size scaling plot of the percolation threshold value Φ_c for the continuum models CM1 (lower curve set) and CM2 in two dimensions. The symbols (×) and (+) denote the results obtained with periodic and non-periodic boundary conditions, respectively. The lines represent a fit to equation (4). The typical variance of the data for small, intermediate, and large systems are marked by the vertical bars.

our estimate is somewhat larger compared with the work of Rosso (1989) the accuracy of our procedure is further enhanced by the simulation of periodic and non-periodic boundary conditions and the coincidence of both extrapolated threshold values. The critical exponent ν obtained by our fit is comparable with the presumably exact value of $\frac{4}{3}$ and with other simulations (e.g. Kertész and Vicsek 1982, $\nu = 1.34 \pm 0.07$). Since the dependence of Φ_c on ν is very weak (figure 3(a)) the percolation threshold is determined with appreciably higher accuracy than the exponent ν.

From the above discussion we conclude that the simulation method can be applied with sufficient accuracy to the determination of the percolation thresholds in continuum models with a radius distribution. In the following we restrict our investigations to the model CM2. The results in two dimensions are shown in figure 1 (upper curve set). The upper line represents the data using periodic boundary conditions. Analogous to the extrapolation for CM1 the data for $\Phi_c(L)$ are fitted to the equation (4) and extrapolated to $L \to \infty$. The length scale of L is defined by $L = 1/R_0$, where R_0 represents the radius of largest disc. The statistical variation of the data for $\Phi_c(L)$ is dependent on the system size. It slightly increases from ± 0.0015 for the smallest systems (several hundred discs) to ± 0.003 (250 000 discs). The percolation density and the critical exponent are obtained as $\Phi_c(\infty) = 0.6860 \pm 0.0012$ and $\nu = 1.28 \pm 0.08$, respectively. Within the error limits the exponent ν is equal to that estimated for model CM1 and to the exact exponent $\nu = \frac{4}{3}$. However, the value of Φ_c is clearly different from the critical area fraction in the model with equally sized discs (CM1) given by the lower curve set in figure 1. Our estimate of Φ_c is more accurate than previous ones for model CM2. Pike and Seager (1974) obtained a value of 0.68 ± 0.02 for a system of 4000 discs using non-periodic boundary conditions which is comparable with our estimate for the same system size (figure 1). However, $\Phi_c(L)$ increases with L for non-periodic boundary conditions resulting in a larger limiting value. The variation of $\Phi_c(L)$ is considerably smaller for periodic boundary conditions so that $\Phi_c(\infty)$ is well approximated even by small systems. Kertész and Vicsek (1982) used a Monte Carlo renormalization group approach and obtained 0.70 ± 0.02. Due to the increased accuracy of our results we conclude that the percolation thresholds for models CM1 and CM2 are different and the suggestion that Φ_c might be independent of the radius distribution in continuum models of percolation is ruled out in two dimensions. This result is in qualitative agreement with the calculations of Phani and Dhar (1984) for their model including discs of two different radii. Because of the considerably larger systems and the finite size scaling procedure applied in the present study we reach an order of magnitude higher accuracy in determining Φ_c.

The results for the three-dimensional model CM2 are presented in figure 2. Contrary to the two-dimensional case $\Phi_c(L)$ decreases with L for both periodic and non-periodic boundary conditions. Obviously, the scaling regime is limited to large systems only whereas for smaller L the data systematically deviate from the scaling law (4). A crossover from non-scaling behaviour to the scaling regime is observed at $1/L \approx 0.003$.

The data in the scaling regime are fitted to (4) in the same way as in two dimensions. The corresponding curves $\Phi_c(\nu)$ along which the goodness of the fit is nearly constant are given in figure 3(b) for periodic and non-periodic boundary conditions. The two parameters at the intersection point of both curves are estimated as $\Phi_c = 0.297 \pm 0.006$ and $\nu = 0.87 \pm 0.07$. The value of ν is in good agreement with 3D lattice (Stauffer 1985, $\nu \approx 0.9$) and continuum models (Balberg and Binenbaum 1985, $\nu \approx 0.83 \pm 0.09$, model CM1). The value of Φ_c determined here has to be compared with that of Pike and Seager (1974), $\Phi_c = 0.303$. This larger value is probably due to the smaller system (8000

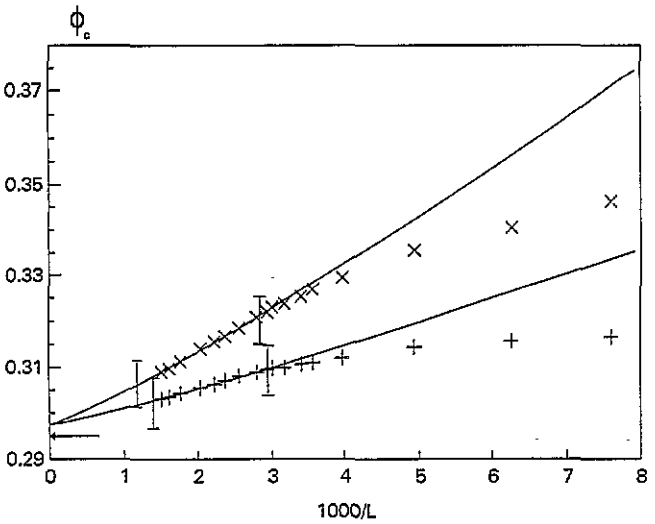

Figure 2. Percolation threshold Φ_c for the model CM2 in three dimensions. The symbols have the same meaning as in figure 1. The arrow denotes the percolation threshold for model CM1.

spheres in comparison with 100 000 spheres in the present simulation) simulated by them since $\Phi_c(L)$ decreases with L. The critical volume fraction as determined for CM2 is only slightly larger than that obtained for equally sized spheres by Pike and Seager (1974), $\Phi_c = 0.295 \pm 0.02$, Haan and Zwanzig (1977), $\Phi_c = 0.295 \pm 0.04$, and the very precise calculation of Lee (1990), $\Phi_c = 0.294 \pm 0.003$. The difference of the percolation thresholds for the models CM1 and CM2 is smaller than the error limits and both values cannot be distinguished in three dimensions.

4. Fractal cluster structures at Φ_c

Using the critical values Φ_c estimated in the previous section we determine numerically the geometric properties (volume, total surface, hull, anisotropy etc) of the clusters as a function of their size R (according to the definitions (1)-(3)). Thereby, we use dimensionless quantities dividing the volume by R_0^d, surface and hull by R_0^{d-1}, and the cluster size by R_0. For both continuum models (CM1 and CM2) several 10^5 clusters are evaluated and the results are averaged within equally divided intervals of the logarithm of cluster size, ln R. The data are represented in a double logarithmic plot. By definition, the fractal exponents are given by the slope of this plot in the scaling region (i.e. in the limit of large cluster sizes). In this range a measure of the statistical error of the averaged data is given by the deviation of the data points from the straight line. In order to exclude an error due to the influence of smaller clusters (outside the scaling regime) we apply the following fit procedure. At first all data are fitted to a straight line. Then we systematically exclude the smallest clusters until the exponents remain unchanged. We find the same fractal exponents as in the lattice models in two and three dimensions. The results are given in more detail for the model CM2 in the following sections.

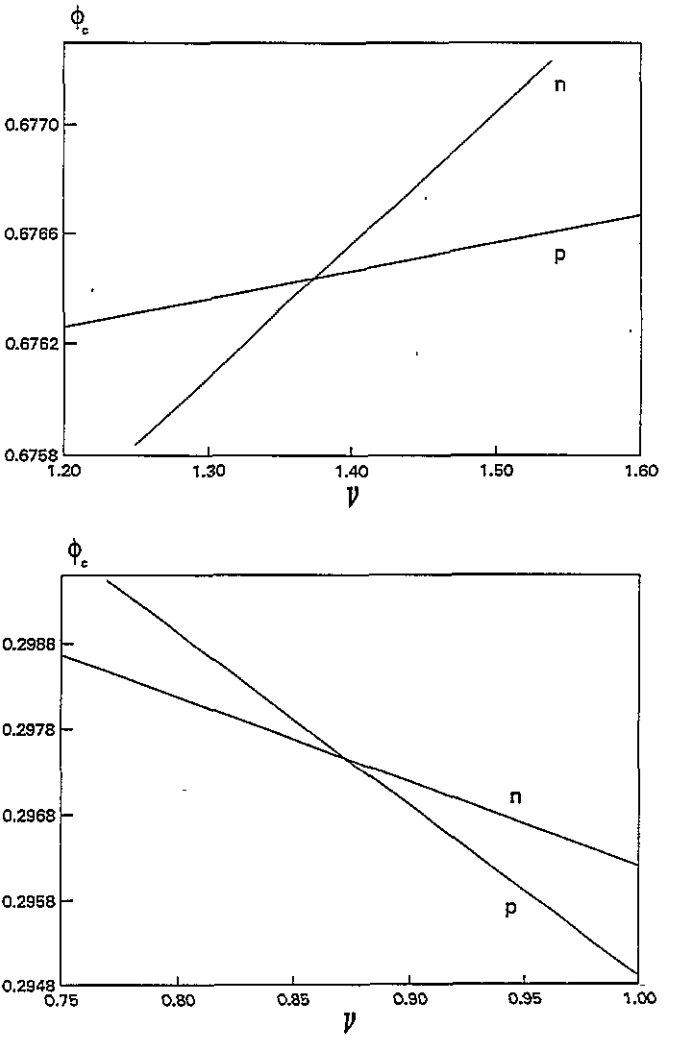

Figure 3. The functions $\Phi_c(\nu)$ along which the fit of the finite size data to the scaling relation (4) yields nearly the same quality for simulations with periodic (p) and non-periodic (n) boundary conditions. (a) Two-dimensional model CM1. (b) Three-dimensional model CM2.

4.1. Two-dimensional model CM2

In the scaling regime (i.e. for large clusters) the cluster volume as function of R is given by

$$V \sim R^{d_f} \tag{6}$$

where the fractal exponent d_f is estimated according to the procedure outlined above (figure 4) as $d_f = 1.894 \pm 0.003$ in excellent agreement with the best calculated values for lattice models including the presumably exact result $d_f = \frac{91}{48}$ (Stauffer 1985).

The same exponent (within the statistical error) is obtained for the cluster surface S which establishes the well known proportionality of the surface and the volume of large clusters, $S \sim V$ (Stauffer 1985). Similarly, we see that the number of discs N

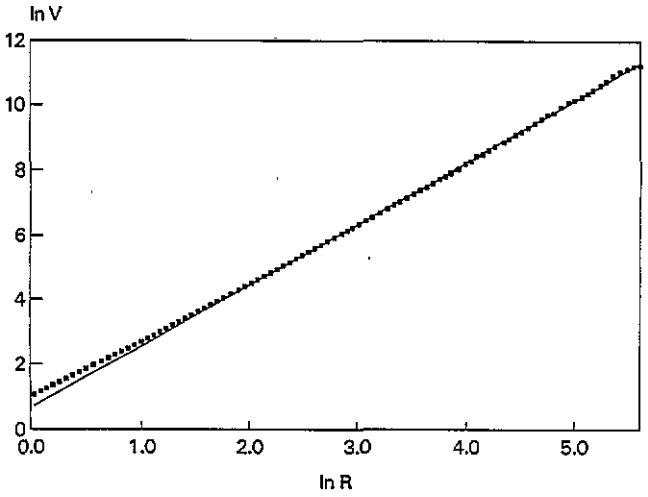

Figure 4. Double logarithmic plot of cluster volume versus cluster radius showing relation (5) with an exponent $d_f = 1.894 \pm 0.003$ (2D model).

within a cluster is proportional to its volume, $N \sim V$ even for very small cluster sizes. Both relations are shown in figure 5. The latter result gives the possibility of suppressing the time-consuming numerical calculation of the cluster volume, defining the cluster size R by equation (1) instead of (2) or (3). Then, with $N \sim R^{d_f}$ the exponent d_f can be determined simulating even larger systems.

The cluster hull H (the surface without internal holes) is shown in figure 6 as function of R. The estimated exponent $d_h = 1.751 \pm 0.002$ is very close to the exact value of $\frac{7}{4}$ for 2D lattice models (Sapoval *et al* 1985, Saleur and Duplantier 1987) and the value $d_h = 1.75 \pm 0.02$ determined by Rosso (1989) for the model CM1.

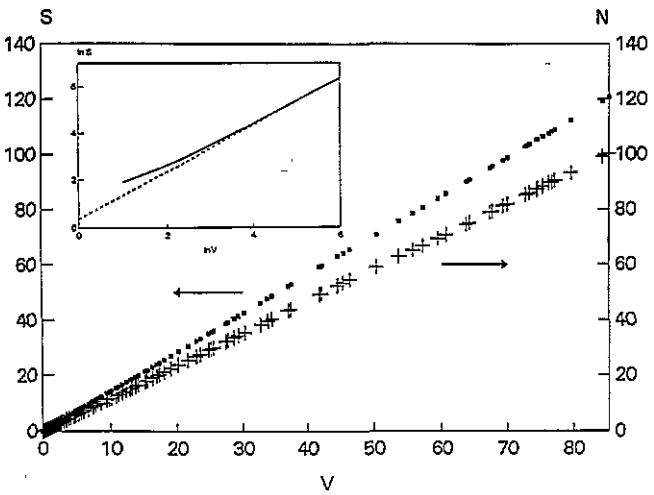

Figure 5. Linear dependence of total surface S (□) and the number of discs N (+) on the cluster volume V, 2D model CM2. For convenience, all values are reduced by a factor of 10^3. The inset shows the deviation from the scaling relation $S \sim V$ (dashed line) for intermediate cluster sizes.

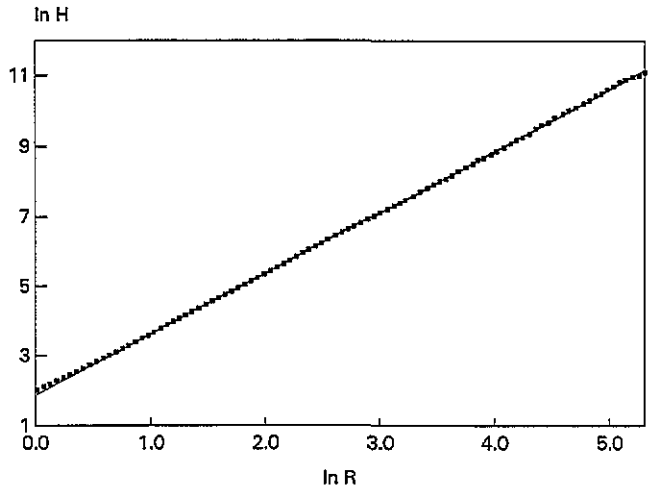

Figure 6. Double logarithmic plot of cluster hull versus cluster radius. The fractal exponent is given by $d_h = 1.751 \pm 0.002$ (2D model).

4.2. Three-dimensional model CM2

According to figure 2 the scaling relation (4) is fulfilled only for rather large systems with $1/L > 0.003$. An analogous situation is observed for the fractal cluster properties (figure 7). The double logarithmic plot ln V versus ln R shows a crossover to the scaling regime at ln $R \approx 2.2$. The fit in the scaling region yields $d_f = 2.512 \pm 0.012$ as the fractal exponent for the cluster volume which is in very good agreement with the corresponding lattice exponents given by Stauffer (1985), $d_f = 2.5$, and Adler *et al* (1990), $d_f = 2.536 \pm 0.047$.

As in 2D, the surface S and the number of spheres of a cluster are both proportional to the volume (figure 8). We have not evaluated the hull in 3D because of numerical

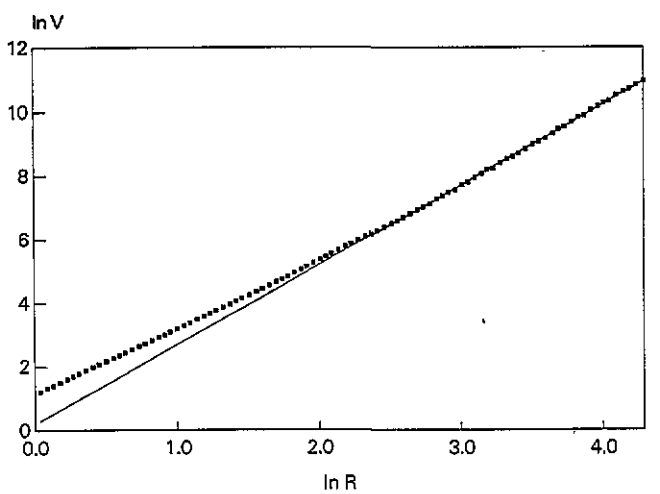

Figure 7. The same as figure 4 in three dimensions. The exponent d_f is given by $d_f = 2.512 \pm 0.012$.

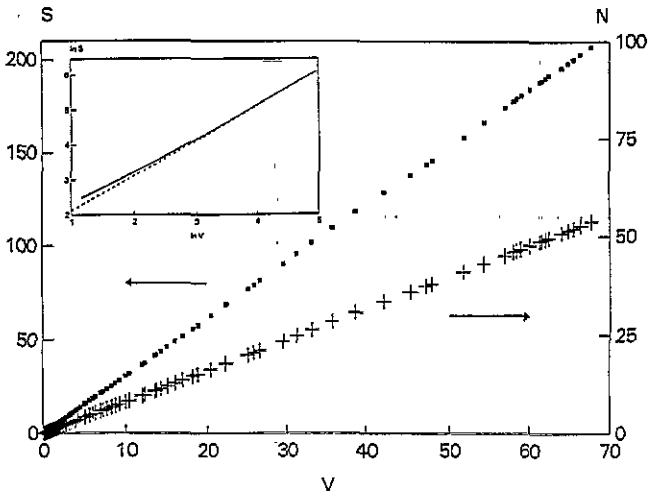

Figure 8. The same as figure 5 in three dimensions.

problems. In the two-dimensional model the hull can be measured by subsequently counting the sections between neighbouring knots (i.e. by calculating the intersections of the discs along the hull). In the three-dimensional continuum model the hull is topologically much more complicated and a suitable algorithm has not been found. However, from recent calculations on 3D lattice models it is strongly suggested that the cluster hull grows with the same exponent as the volume (Bradley *et al* 1991).

Finally, we have applied the numerical simulation technique also to the model CM1 (equally sized discs or spheres) and found within the error limits the same fractal exponents. Because of several recent papers on this model we omit a detailed discussion of our results.

4.3. Corrections to scaling and cluster shapes

The proportionality of cluster surface and volume is well accepted for large cluster sizes not only at Φ_c but also above and below the percolation threshold (Stauffer 1985). For smaller clusters, however, corrections to the scaling law are expressed by the equation

$$S \sim V - \text{const}\ V^{\zeta} \tag{7}$$

where the second contribution is assumed to behave like a common surface term at $\Phi > \Phi_c$, i.e. $\zeta = 1 - 1/d$. For $\Phi < \Phi_c$ the value of ζ is unity. At the percolation threshold it is expected from the results of lattice models that $\zeta = \sigma$, with $\sigma^{2d} = \frac{36}{91}$ and $\sigma^{3d} = 0.45$ (Stauffer 1985). In order to evaluate the exponent ζ at the percolation threshold Φ_c we fit the cluster data to (7) according to the procedure outlined at the beginning of section 4 (inset in figures 5 and 8). For the continuum model CM2 we get in two and three dimensions $\zeta^{2d} = 0.234$ and $\zeta^{3d} = 0.178$, respectively. Both exponents are far from the expected value σ as well as from the two limiting values below and above Φ_c. From a similar fit to (7) we get in the model CM1 $\zeta^{2d} = 0.256$ and $\zeta^{3d} = 0.366$. While ζ^{2d} is close to the corresponding value in the model CM2 there occur larger differences in three dimensions. The deviation of ζ from the lattice exponent σ is somewhat surprising in the view of universality of continuum and lattice models. However, in

the present simulation the size of the clusters contributing to the scaling correction may be too small to reproduce the correct exponent σ. Since (7) is valid only in the limit $V \to \infty$ even larger systems have to be simulated in order to evaluate this very sensitive quantity.

Another point of interest is the shape of the clusters. We measure the anisotropy of a cluster by calculating its principal moments of inertia I_j. An anisotropy factor f_a is defined by the quotient of the smallest and the largest moment. For an isotropic cluster this factor is 1 whereas for a long and thin structure it tends to zero. To obtain a reliable estimate for f_a we average over a large number of equally sized clusters. The resulting anisotropy factors are shown in figure 9 as functions of cluster size ln R. For large sizes f_a tends to a constant (0.4 for $d=2$ and 0.5 for $d=3$) well below the isotropic case ($f_a = 1$). From the data of figure 9 we conclude that large clusters up to the percolating cluster are anisotropic at Φ_c. In order to estimate the anisotropy factor of the percolating cluster we have performed averages over the f_as of percolating clusters only. The values obtained are $f_a = 0.416$ (2D) and $f_a = 0.503$ (3D). For a better understanding of the cluster anisotropy the cluster shapes should be investigated below and above Φ_c.

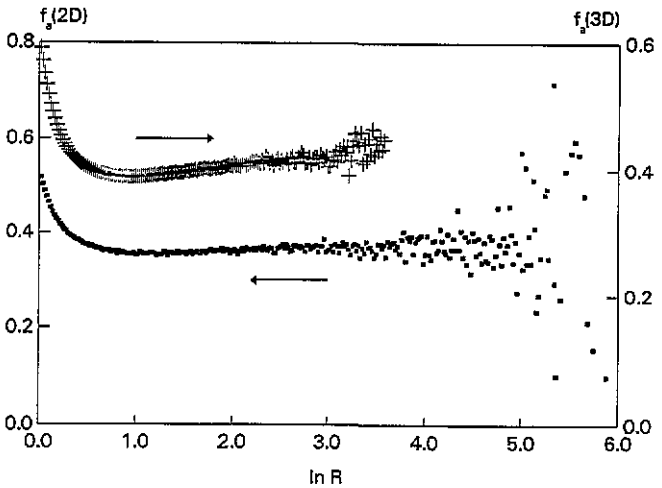

Figure 9. The anisotropy factor f_a as function of the logarithm of cluster radius R for the continuum model CM2 in two (left scale, lower data set) and three dimensions (right scale, upper data set). The scatter of the anisotropy at large radii results from the large configurational variance of f_a. A well defined mean value is obtained only for averaging over a large number of equally sized clusters as can be seen for small radii.

5. Conclusions

The comparison of the fractal dimensions at the percolation threshold confirms the conjecture that continuum models with different radius distributions of nuclei belong to the universality class of the corresponding lattice models. This is also supported by recent work on the growth of Eden clusters on 2D lattices which can be extrapolated to the continuum model of percolation (Evans 1990).

The disorder due to varying radius distributions should be an irrelevant quantity according to the Harris criterion (Harris 1974). Comparing the results of both continuum models CM1 and CM2, we have shown that the Harris criterion is valid in the continuum percolation problem.

The investigation of non-universal quantities shows small but distinct changes with the radius distribution in continuum percolation. The percolation thresholds Φ_c for the models CM1 and CM2 differ only by a small amount in 3D which is comparable with the accuracy of our analysis. However, in 2D we can rule out an equality of Φ_c for both models. At Φ_c, the exponent ζ describing the deviations from the proportionality of surface and volume of smaller clusters deviates from the expected lattice exponent σ. This open question should be investigated by more refined methods or simulations of even larger systems. The average shape of larger clusters at Φ_c is anisotropic. This feature should be investigated in more detail above and below Φ_c.

Acknowledgments

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