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LETTER TO THE EDITOR

# Cluster shapes in lattice gases and percolation

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**Abstract.** An analysis is undertaken of the mean surface $\bar{s}$ of clusters of size $n$ from Monte Carlo data simulating a two-dimensional Ising model. At sufficiently high temperatures the data represent a percolation process and it is found that the clusters are completely ramified (tree- or sponge-like). At temperatures just below $T_c$ the data do not correspond to circular droplets as assumed in standard nucleation theory and typical samples show a great deal of ramification. It is concluded that the parameters in Fisher's droplet model are empirical and should not be given a direct physical interpretation.

The droplet model of condensation was first introduced in the 1930s and has subsequently played a central role in nucleation theory. It showed how supercooled droplets can exist in a metastable condition and provided an apparatus for the calculation of the lifetime of these metastable states (for a review of early ideas see Frenkel 1945). The droplets were always considered to be spherical; it was realized that other shapes of droplet could arise but a detailed investigation was considered unnecessary since such a refinement could not seriously affect the general conclusions of the theory.

The first suggestion that the model might be useful in describing critical point behaviour was made by Essam and Fisher (1963) and the treatment was amplified and extended by Fisher (1967). Droplets of all shapes were taken into account and the shape was conveniently characterized by a single parameter $\sigma$, where for large $n$

$$s \sim An^{\sigma}. \tag{1}$$

Here $n$ is the number of molecules in the droplet and $s$ its surface area; $\sigma$ could take all values from a minimum $\sigma_0$ corresponding to the most compact shape ($\frac{1}{2}$ in two dimensions and $\frac{2}{3}$ in three dimensions) to 1 corresponding to tree-like or sponge-like shapes; and $A$ is a constant of order unity to differentiate for example between spherical and cubical shapes with the same value of $\sigma$.

For the number of droplets of given $\sigma$ Fisher proposed the form

$$(\lambda(\sigma))^s/s^{\tau(\sigma)} \tag{2}$$

where $\lambda(\sigma)$ is a constant related to the surface entropy and $\tau(\sigma)$ an exponent similar to that for self-avoiding polygons on a lattice (see eg Domb 1969). He constructed a partition function and, using standard ideas of statistical mechanics, assumed that one particular set of parameters $\bar{\sigma}$, $\bar{\tau}$ would dominate asymptotically. The thermodynamic behaviour of the condensing gas could then be described in terms of these two parameters. Critical exponents could be expressed in terms of $\bar{\sigma}$, $\bar{\tau}$ and, using exact results in

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two dimensions and estimates based on high-temperature series expansions in three dimensions, the following numerical values were assigned to $\bar{\sigma}$ and $\bar{\tau}$:

$$\begin{aligned} &\bar{\sigma} = \tfrac{8}{15} \simeq 0{\cdot}533, \qquad \bar{\tau} = \tfrac{31}{15} \simeq 2{\cdot}067 \qquad \text{in two dimensions} \\ &\bar{\sigma} \simeq 0{\cdot}64, \qquad\qquad \bar{\tau} \simeq 2{\cdot}2 \qquad\qquad \text{in three dimensions.} \end{aligned} \tag{3}$$

The above development provided a great stimulus to the theory of critical behaviour since it proposed exact relations between critical exponents and paved the way for the subsequent scaling hypothesis (see eg Vicentini-Missoni 1972). However on closer scrutiny a number of defects of the model became apparent. The excluded volume effect is ignored, ie that no two droplets can occupy the same region of space; this is of increasing importance as the density of droplets increases. Gaunt and Baker (1970) pointed out that the model gives invalid results when $T > T_c$. Fisher himself noted that the three-dimensional estimate of $\bar{\sigma}$ quoted above is geometrically impossible, and the exponent must therefore be regarded as an 'effective average'.

In view of these defects it is important to investigate whether the exponent values in (3) can be given the physical interpretation that the droplet shapes remain fairly spherical (or circular) as the temperature moves up to the critical region or whether they are to be treated as empirical parameters. It has been suggested recently by one of us (Domb 1975) that, whereas at sufficiently low temperatures compact spherical droplets should dominate, at higher temperatures *ramified* droplets of tree-like or sponge-like shape should become increasingly important. The dominant value of $\sigma$ should therefore change with temperature.

Droplet or cluster shapes are also important in the theory of percolation and dilute magnetism (see eg Essam 1972). Here the clusters considered are purely random and no excluded volume effects arise. It has been argued (Domb 1974) that only ramified clusters with $\sigma = 1$ play a significant role in the problem (see also Stauffer 1975). Attention has also been recently drawn to percolation effects in Ising systems at different temperatures (Müller-Krumbhaar 1974, Coniglio 1975); in three-dimensional systems infinite clusters can occur at temperatures well below $T_c$.

The aim of this letter is to present Monte Carlo data obtained with a two-dimensional one-spin-flip Ising model (Glauber model) in an endeavour to throw some light on to these points. For a detailed discussion of our Monte Carlo procedure and its limitations in the simulation of an infinite system we refer to Stoll *et al* (1973) and Schneider and Stoll (1974). Here we have considered a $110 \times 110$ Ising system on a simple quadratic lattice subject to periodic boundary conditions. On this basis we identified the clusters by labelling all down-spins connected by nearest-neighbour bonds in such a way that different clusters contain spins with different labels. To evaluate the surface of a particular cluster we counted the number of spin reversals at the cluster border. This takes account of inclusions which may be important for ramified clusters. In this manner we calculated the distribution function $P(n, s)$ where $s$ is the surface and $n$ the cluster size. Given this distribution it is then possible to calculate the size dependence of the mean surface

$$\bar{s}(n) = \frac{\Sigma s P(n, s)}{\Sigma P(n, s)}. \tag{4}$$

Our data revealed that the function $P(n, s)$ as a function of $s$ for fixed $n$ has a single peak.

In figure 1 the mean surface given by (4) is plotted against $n$ for a series of different temperatures above and below $T_c$. The scale is logarithmic so that the asymptotic value

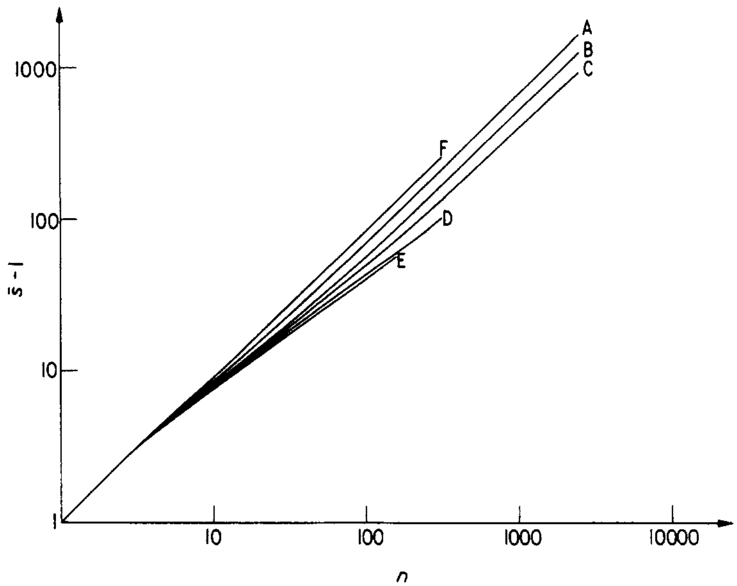


**Figure 1.** Mean surface $s$ as a function of cluster size $n$ plotted on a logarithmic scale for a number of temperatures. (Each point is the average of at least 100 clusters; sparser data are not reproduced.)
A, $T/T_c = 2{\cdot}0$; B, $T/T_c = 1{\cdot}3$; C, $T/T_c = 1{\cdot}1$; D, $T/T_c = 0{\cdot}958$; E, $T/T_c = 0{\cdot}9$; F, $T = \infty$.

of the slope should provide an estimate of $\sigma$ in (1). The convergence to this limit must necessarily be slow because of the logarithmic scale, but for values of $n$ of the order of a few hundred or more the data should provide reasonable estimates.

When $T \to \infty$ the problem is one of pure percolation and we find that $\sigma \to 1$ quite accurately (0·005) for $n > 50$. The data therefore confirm that ramified clusters are dominant in the percolation problem. When $T/T_c = 2$ we still find that $\sigma \to 1$ accurately when $n > 250$; the problem is still one of almost pure percolation although the value of $A$ in (1) must be slightly decreased. When $T/T_c = 1{\cdot}3$, although the limiting value of $\sigma = 1$ is still attained with the same accuracy, asymptotic behaviour does not set in until $n \sim 500$, and the value of $A$ required to fit the data asymptotically must be further decreased. When $T/T_c = 1{\cdot}1$ the limiting asymptotic behaviour of $\sigma = 1$ is not attained until $n \sim 1500$, and $A$ must be further decreased.

When $T < T_c$ the data are much more restricted since the probability of obtaining large clusters in the sample is much smaller. For $T/T_c = 0{\cdot}958$ we find an effective value of $\sigma = 0{\cdot}75 \pm 0{\cdot}05$ for $80 < n < 320$; for $T/T_c = 0{\cdot}9$ we find approximately the same value for $80 < n < 160$. We do not think the asymptotic limit has yet been attained but we consider that these values are sufficiently far from the estimate (3) to justify the conclusion that ramified droplets are important near $T_c$.

To confirm this view we have plotted a few typical cluster shapes for $T/T_c$ slightly less than 1 in figure 2. For a more extensive demonstration we refer to the motion picture of Stoll and Schneider (1972, 'Computer simulation of a two-dimensional Ising model', available on request) showing in detail the evolution of spin configurations. From figure 2 it will be seen that the clusters manifest tree-like characteristics and differ markedly from a circular shape. In an earlier communication (Stoll and Schneider 1972) attention was drawn to disagreement between Monte Carlo simulations and the

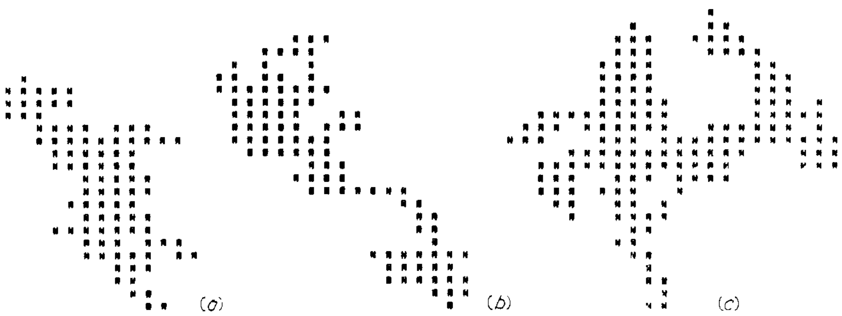


**Figure 2.** Some typical clusters for $T < T_c$: (*a*) $T/T_c = 0{\cdot}958$. (*b*) $T/T_c = 0{\cdot}979$. (*c*) $T/T_c = 0{\cdot}99$. The 'tree-like' character of the clusters should be noted, and the increase of ramification as $T$ approaches $T_c$.

simple Frenkel picture of a circular or spherical-shaped droplet; the observed deviation from circularity in droplet shapes was then termed 'border anisotropy'.

Our conclusions for $T/T_c = 0{\cdot}9$ differ from those of Binder and Stauffer (1972) who attempted a similar analysis and found a value of $\sigma$ close to $\frac{1}{2}$ for larger $n$ (in fact they suggested that the slope of the curve in figure 1 *decreases* with increasing $n$ which is the reverse of our general finding). We believe that the statistics at our disposal and those which were available to Binder and Stauffer need further investigation.

We should like to comment finally on the significance of clusters in the percolation and Ising problems. As we lower the temperature from $T = \infty$ to $T = 0$ in the Ising model we pass gradually from clusters which have purely geometric significance and play no part in the physics of the problem, to clusters which are true 'droplets' and are of central physical importance. In the former case neighbouring spins are uncorrelated and the linking together of neighbouring spins in a cluster is a geometrical property. As the temperature is lowered physical effects begin to manifest themselves as a result of correlation between neighbouring spins, but in the neighbourhood of $T_c$ it is doubtful whether these clusters themselves are relevant to critical behaviour. Instead of clusters we should perhaps use configurations consisting of correlated groups of spins which become identical with physical droplets at sufficiently low temperatures but which break down into individual spins as $T \to \infty$ (similar ideas have been put forward recently by Binder *et al* 1975). Such configurations cannot readily be studied by Monte Carlo methods, but they can help in a theoretical understanding of condensation and critical behaviour (Domb 1975).

We summarize our main conclusions as follows:

(i) In percolation phenomena ramified clusters play a dominant part.

(ii) Near $T_c$ clusters deviate markedly from a compact circular shape.

(iii) The parameters in Fisher's droplet model should be treated as empirical averages and should not be related directly to cluster shapes.

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