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Ising Cluster Kinetics at the Critical Point

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The scaling laws of Edwards *et al.* for cluster fragmentation at the two-dimensional percolation threshold, recently confirmed also in high dimensions, explain the Alexandrowicz observation that the Becker–Döring equation of classical nucleation theory and its generalization by Katz, Saltsburg, and Reiss fail right at the critical point.

KEY WORDS: Glauber kinetics; percolation; fragmentation.

Nucleation theory explains reasonably well the formation of liquid droplets out of a supersaturated vapour, as well as the differences between stretched exponential and simple exponential relaxation of the *d*-dimensional Ising model above and below its critical temperature. These phenomena away from equilibrium are approximated by the Becker–Döring equation:(1)

$$\partial N_s/\partial t = \frac{\partial}{\partial s}\left[R_s N_s \frac{\partial}{\partial s}(N_s/n_s) \right] \tag{1}$$

Here R_s is a cluster growth rate, $N_s(t)$ is the non-equilibrium number of clusters containing s molecules each, and n_s the stationary (equilibrium) number. Earlier literature is cited in ref. 2.

Alexandrowicz,(3) however, found that right at the critical point Eq. (1) fails, and this failure was confirmed by Monte Carlo simulations of the two- and three-dimensional Ising models.(4) Eq. (1) was derived(1) by assuming clusters to grow and shrink only by single molecules, but it has

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been generalized long ago$^{(5)}$ to the incorporation and splitting-off of small j-clusters; in this case the growth rate becomes

$$R_s = \sum_j j^2 K(s, j)\, N_j \tag{2}$$

with the coagulation rate K at which two clusters collide and merge. Here one takes $j \ll s$; more generally with clusters of all possible size merging and splitting again, the coagulation-fragmentation equation is:$^{(6)}$

$$\begin{aligned} \partial N_s/\partial t = {} & \tfrac{1}{2} \sum [K(i, j)\, N_i N_j - F(i, j)\, N_{i+j}] \\ & - \sum [K(s, j)\, N_s N_j - F(s, j)\, N_{s+j}] \end{aligned} \tag{3}$$

Here $F(i, j)$ is the rate at which a cluster with $s = i + j$ molecules splits into two with i and j particles; the first sum goes over all i and j with $s = i + j$, and the second over all j. In equilibrium,

$$K(i, j)\, n_i n_j = F(i, j)\, n_{i+j} \tag{4}$$

must hold. A Taylor expansion for small j then gives Eq. (3). However, right at the critical temperature the growth rate R_s was found to go to zero for $t \to \infty$ at fixed s, indicating something fundamentally wrong, as pointed out by Alexandrowicz.$^{(3)}$

Now we estimate the coagulation coefficients from binary fragmentation studies of random bond percolation,$^{(7)}$ assuming that site percolation is in the same universality class as bond percolation and that Ising clusters have the same structure of the scaling equation and only slightly different critical exponents. For example, at the critical point, both percolation and Ising clusters (properly defined) are fractal and have nearly the same fractal dimensions, like 2.53 and 2.48 in three dimensions.

Edwards *et al.*$^{(7)}$ showed at the bond percolation threshold that a finite fraction of all cluster bonds are fragmenting in the sense that without them the cluster of size s splits into two parts of sizes i and j with $i + j = s$. Moreover, the probability b_{js} for a fragmenting s-cluster to split off a fragment of size j (j was called s' in ref. 7) followed a scaling law

$$b_{js} = s^{-\phi} f(j/s) \tag{5}$$

where ϕ varies only slightly between 1.6 and 3/2 in two to infinite dimensions. For small j this probability is independent of s according to Edwards *et al.*,$^{(7)}$ and thus $f(x \to 0) \propto x^{-\phi}$ or $b_{js} \propto j^{-\phi}$ for $1 \ll j \ll s$. Numerically in two dimensions they found $f \propto [x(1-x)]^{-\phi}$ as in the Bethe lattice, for all x.

For clusters in a Glauber kinetic Ising model, not all spins will flip or all bonds will break with the same probability. But these probabilities are purely local and thus their mutual ratios do not depend on the cluster size. Thus as far as the exponents for the fragmentation probability are concerned, we approximate the Ising fragmentation probability $F(i, j)$ by the percolation fragmentation probability b_{js} with $s = i + j$; only the exponent ϕ is expected to be slightly different. Thus

$$F(i, j) = F(j, i) \propto s^{-\phi} f[i/s] \simeq (ij/s)^{-\phi} \qquad (s = i + j) \tag{6}$$

at the Ising critical point.

The equilibrium cluster numbers at the Curie point are known to decay asymptotically as $n_s \propto s^{-\tau}$ with τ varying from about 2 to 5/2 for $d = 2$ to infinity; then the coagulation rates are

$$K(i, j) = n_s F(i, j)/(n_i n_j) \propto (ij/s)^{\tau-\phi} \qquad (s = i + j) \tag{7a}$$

which for $1 \ll j \ll i$ and thus $i \simeq s$ reduces to

$$K(s, j) \propto j^{\tau-\phi} \tag{7b}$$

as an approximation for $K(s, j)$ in Eq. (2).

Now the equilibrium growth rate R_s of Eq. (2), which converges away from the critical point because of the exponential decay of the cluster numbers there, right at the critical point is proportional to $\sum_j j^2 j^{\tau-\phi} j^{-\tau} = \sum_j j^{2-\phi}$ which is clearly diverging. Thus the simple approximation (2), which is based on small clusters colliding with big ones, is no longer good right at the Ising critical point; there coalescence of clusters of about equal sizes, requiring the solution of the full Eq. (3) instead of (1), seems to be the dominating process and explains the failure of the Becker–Döring equation.$^{(3)}$ However, we do not exclude other sources of deviations from the Becker–Döring equation, like excluded volume effects$^{(8)}$ or other cluster-cluster correlations.

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