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Essential singularities in dilute magnets

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Harris argued in favor of an essential singularity at zero magnetic field in the equation of state for randomly dilute low-temperature ferromagnets. His assumptions and conclusions are reexamined and criticized with the help of earlier Monte Carlo data on the number W_n of clusters with n spins each. These data suggest for large clusters $\log W_n \propto -n$ on the paramagnetic side, whereas $\log W_n \propto -n^{0.4}$ on the ferromagnetic side. More Monte Carlo work is suggested, if $n \sim 100$, for square site percolation near $p = 0.40$ or square bond percolation near $p = 0.35$.

Griffiths[1] showed that the magnetic equation of state $M(p,H,T)$ cannot be analytic in H for a randomly dilute quenched ferromagnet where only a fraction p of lattice sites are occupied by magnetic atoms. Instead, a nonanalyticity has to occur at magnetic field $H=0$ for $T_C(p) < T < T_C(p=1)$, where $T_C(p)$ is the Curie temperature of the dilute magnet and $T_C(p=1)$, the "pure" Curie temperature. For very low temperatures, the $M(p,H,T)$ problem reduces to the percolation problem,[2] and we restrict ourselves to this limit. Harris[3] argued that (in this limit) this nonanalyticity is an "essential" singularity where all derivatives of M with respect to H are finite but where the radius of convergence R of the Taylor series

$$M = \sum_{i=0}^{\infty} a_i H^i \qquad (1)$$

vanishes. But, Harris[3] proved this result for the Bethe lattice (Cayley tree) only,[2,4,5] which is some sort of mean-field approximation.[4] For real lattices[2,6,7] he made an assumption about the average number W_n of clusters of size n which was too simple, as we will discuss below. In the present paper we arrive, on the basis of earlier Monte Carlo simulations,[6] at a different behavior of these cluster numbers and look at the resulting essential-singularity answers.

Such essential singularities (more precisely, Taylor expansions with finite derivatives but zero radii of convergence) have been discussed before[8,9] for pure Ising magnets (or fluids) on the coexistence curve. Our Fig. 1 shows the three cases discussed so far: the pure Ising model ("Fisher"), the dilute low-temperature limit ("this work and Harris"), and the general dilute paramagnet ("Griffiths"). It is evident from Binder's discussion[9] that the arguments for the Fisher singularity do not apply to the paramagnetic region (Griffiths). Similarly, the Griffiths results for finite temperatures are not necessarily valid in the $T \to 0$ low-temperature limit (this work and Harris). Thus, since a rigorous relation seems lacking at present between these three singularities, one has to deal with each case separately. But, the mathematical methods and physical scaling assumptions of this work are very similar to the earlier discussions of the pure Ising model.

In a random quenched ferromagnet with nearest-neighbor interactions (for simplicity we take the spin-$\frac{1}{2}$ Ising model), the spins form clusters connected by nearest-neighbor interactions. We denote by W_n the average number of clusters (per spin) containing n spins each. For very low temperatures, as assumed here, all n spins within one cluster are parallel, whereas the orientations of different clusters are uncorrelated. In a magnetic field H (in suitable units), the dimensionless magnetization density M due to these clusters is

$$M = \frac{1}{2} \sum_{n=1}^{\infty} n W_n \tanh\left(\frac{nH}{2kT}\right). \qquad (2)$$

For p larger that the percolation threshold p_c, one infinite percolating network[2] appears and adds the spontaneous magnetization, independent of H, to

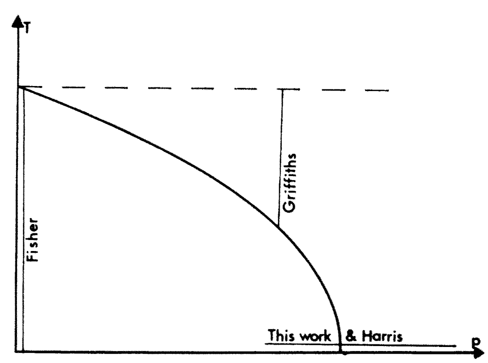

FIG. 1. Schematic phase diagram, with the position of various (essential) singularities, as discussed in Refs. 8 and 9 (Fisher), Ref. 1 (Griffiths), and Ref. 3 (this work and Harris).

the M of Eq. (2). Then $a_{i=0} \neq 0$ without other changes, and thus Eq. (2) alone determines the convergence of the expansion (1). Another quantity of interest is the generating function[7]

$$M' = \text{const} - \sum_{n=1}^{\infty} n W_n e^{-nH/kT} \tag{3a}$$

$$= \sum_{i=0}^{\infty} a'_i H^i , \tag{3b}$$

which is similar to the Mayer cluster expansion for liquid-gas systems.

Taylor expansions of Eqs. (2) and (3a) give

$$a_i = \tfrac{1}{2}(2kT)^{-i}(i!)^{-1} V_i \left(\frac{d}{dx}\right)^i \tanh(x)_{x=0} , \tag{4a}$$

$$a'_i = (-1)^{i+1}(kT)^{-i}(i!)^{-1} V_i , \tag{4b}$$

where

$$V_i = \sum_{n=1}^{\infty} n^{1+i} W_n . \tag{4c}$$

Asymptotically,[3]

$$\left(\frac{d}{dx}\right)^i \tanh(x)_{x=0} \sim \left(\frac{2}{\pi}\right)^i i!$$

for odd i whereas the derivatives vanish for even i. The radii of convergence R and R' for the expansions (1) and (3b), respectively, are given by

$$\frac{R'}{kT} = \lim_{i\to\infty} \left| \frac{a'_{i-1}}{a'_i} \right| = i \frac{V_{i-1}}{V_i} \quad (i \to \infty) , \tag{5a}$$

$$\frac{R}{kT} = \pi \frac{V_{i-1}}{V_i} \quad (i \to \infty) . \tag{5b}$$

For the higher moments $V_{i\to\infty}$ in Eq. (4c) only the large n are relevant. If the large cluster sizes behave asymptotically as

$$W_n \sim \exp(-\text{const} \times n^{\zeta}), \quad 0 < \zeta < \infty \tag{6}$$

apart from preexponential factors [or more generally, if $\ln(W_n)/n^{\zeta} \to \text{const} > 0$ for large n], then for large i the V_i vary as gamma functions $\Gamma(\text{const} + i/\zeta)$ and

$$V_{i-1}/V_i \propto (i/\zeta)^{-1/\zeta} \quad (i \to \infty) . \tag{7}$$

Thus, for large i,

$$\frac{R}{kT} \propto \lim_{i\to\infty} i^{-1/\zeta} , \tag{8a}$$

$$\frac{R'}{kT} \propto \lim_{i\to\infty} i^{1-1/\zeta} . \tag{8b}$$

Therefore the Taylor expansion (1) for M always has zero radius of convergence [if Eq. (6) is correct], whereas in expansion (3b) for M' the radius of convergence vanishes only for $\zeta < 1$.

For the Bethe lattice[5] one has $\zeta = 1$ for both $p < p_c$ (paramagnet) and $p > p_c$ (ferromagnet), giving essential singularities everywhere for both M and M'. In real lattices, such essential singularities occur in M independent of the actual cluster-size distribution W_n, whereas M' remains analytic in H, if W_n decays asymptotically at least as quick as $\exp(-\text{const} \times n)$, i.e., if $\zeta \geq 1$. In the droplet model of three-dimensional "pure" liquid-gas or Ising-magnet phase transitions,[8,9] Binder argued that $\zeta = \frac{2}{3}$ on the "ferromagnetic side" [now corresponding to $T < T_C$ since $p = 1$ is no longer a variable then; Eq. (2) is inadequate[9]]; and the same arguments apply to dilute ferromagnets at finite temperatures: essential singularity in $M'(T < T_C)$. Kretschmer *et al.*[9] argue for $\zeta = 1$ on the pure paramagnetic side: no essential singularity in M' above $T_C(1)$. Reatto[10] suggested the same value $\zeta = 1/(\beta + \gamma)$ for ferromagnetic and paramagnetic pure behavior, leading to essential singularities in M' everywhere.

For dilute low-temperature ferromagnets on real lattices (i.e., not on trees), Harris *assumes* $\zeta = 1$, apparently for both $p < p_c$ and $p > p_c$, since a factor $p^n(1-p)^s$ enters W_n, where s is the cluster perimeter.[2] Erroneously Harris takes $s \propto n^{2/3}$ as for the *outer* surface of a droplet, thus giving $\zeta = 1$ from the now dominating p^n factor. But, according to Domb,[11] one needs $s/n \to \text{const}$ for large n in order to find the desired $p_c < 1$. Even then the factor $p^n(1-p)^s$ does not necessarily lead to $W_n \sim \exp(-\text{const} \times n)$ as discussed in Ref. 12. In fact, a simple analysis[13] of older Monte Carlo data[6] gave $\zeta \simeq 0.36$ in two dimensions for the ferromagnetic side. The present paper, after this simple review, analyzes the asymptotic decay of the Monte Carlo[6] W_n for the paramagnetic side in order perhaps to clarify the essential-singularity question there.

The analysis for $p < p_c$ is more difficult than the one for $p > p_c$ since the "scaled" cluster numbers $W_n(p)/W_n(p_c)$ first increase, then decrease with n. This maximum[13] leaves only the largest clusters as candidates for the asymptotic behavior where data are few and inaccurate. For $n > 10$, Dean and Bird[6] only give sums like $\sum_{n=101}^{1000} W_n$. Reference 13 simply identified this sum with $(1000 - 100) W_n$ evaluated at the geometric mean $n = (1000 \times 101)^{1/2}$, etc. This simple but crude approximation works well for p very close to p_c but is inaccurate farther away from the phase transition. Thus, instead, we work here with the partial sums

$$S_n(p) = \sum_{m=n}^{\infty} W_m(p) \Big/ \sum_{m=n}^{\infty} W_m(p_c) , \tag{9}$$

thus avoiding such "geometric-mean" plots. If W_n asymptotically decays exponentially in n, as assumed in Eq. (6), then

$$S_n(p) \sim W_n(p) \quad (n \to \infty) \tag{10}$$

apart from preexponential factors.

Figure 2 shows a selection of data for the paramagnetic region. The two-dimensional triangular site and the (very similar) square bond percolation W_n are suited best since there $p_c = \frac{1}{2}$ exactly.[2] We see that for sufficiently large clusters the S_n seem to decay as $\exp(-\text{const} \times n)$, i.e.,

$$\ln(S_n) \propto -n \quad (n \to \infty,\ p < p_c)\ , \tag{11a}$$

whereas in the ferromagnetic region Stauffer[13] gave

$$\ln(S_n) \propto -n^{0.36} \quad (n \to \infty,\ p > p_c) \tag{11b}$$

for the critical region, p near p_c. [With the scaling assumption[13] for the critical region, $W_n \propto n^{-2-1/\delta} f((p-p_c)n^{1/\beta\delta})$ and $\beta\delta = \beta + \gamma$, Eqs. (11) mean $\ln f(x) \propto -x$ for the ferromagnetic side, $x > 0$, and $\ln f(x) \propto -(-x)^{\beta\delta}$ for the paramagnetic side, $x < 0$, for $n \to \infty$ in both cases. However, the ferromagnetic result (11b) is valid approximately[13] for all values of the scaling variable $x = (p - p_c)n^{1/\beta\delta}$, whereas we get the paramagnetic result (11a) for large arguments x only.] This asymmetry between Eqs. (11a) and (11b) is the main result of our paper and agrees with the above-mentioned phenomenological arguments[9] for usual phase transitions.

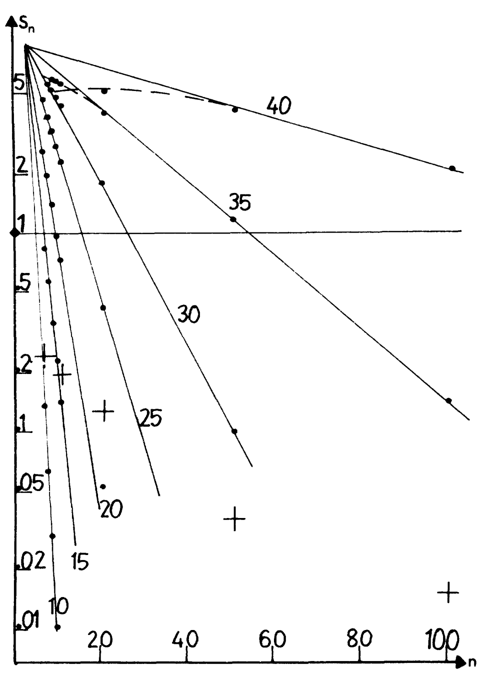

FIG. 2. Logarithmic plot of S_n, Eq. (9), versus cluster size n for two-dimensional square bond percolation. The numbers on the curves give the concentration p of spins in percent. Asymptotic decay along straight lines, as suggested by this figure, gives $\zeta = 1$ in Eq. (6). The Monte Carlo error is a few percent for large S_n and about a factor 2 for the smallest S_n shown. The crosses denote a ferromagnetic example, $p = 0.55$.

The asymptotic straight lines in our logarithmic plot all go through the same fixed point

$$S_n(n = n_F) = S_F\ , \tag{12}$$

where n_F and S_F are independent of p. This property facilitates a determination of the asymptotic behavior for inaccurate data. The intercepts n_{ic}, where $S_n(n = n_{ic}) = 1$, vary with the concentration as

$$n_{ic} = n_0[(p_c - p)/p_c]^{1/\sigma}\ , \tag{13}$$

where $\sigma = 0.40$ fits these intercepts for $0.1 < p < p_c$. Similar behavior with different fixed points is found (not shown) for other two- and three-dimensional lattices; the resulting fit parameters are given in Table I.

If our data were close to the critical point $p \to p_c$, $n \to \infty$ (which is *not* the case) then scaling arguments[8,9] would require the exponent $1/\sigma$ in Eq. (12) to equal $\beta + \gamma$ [usual notation: susceptibility $\propto (p - p_c)^{-\gamma}$, spontaneous magnetization $\propto (p - p_c)^{\beta}$]. For two dimensions, $\beta + \gamma = 1/0.39$ according to Ref. 7, whereas $\beta + \gamma = 1/0.46$ in three dimensions according to Kirkpatrick.[6] Our "effective" exponents σ in Table I are reasonably consistent with these predictions, since they were taken from p rather far away from p_c. Note the systematic increase of σ with p_c in two dimensions.

{Again we may try the scaling assumption[13] mentioned already after Eq. (11). Plotting S_n as a function of the "scaled cluster size" $n_s \propto (p_c - p)^{\beta+\gamma} n = |x|^{\beta\delta}$, we find roughly one curve for all p in the square bond percolation case. And if, following universality ideas, the proportionality factor in Eq. (13) is fit appropriately, then all two-dimen-

TABLE I. Parameters fitted on the Monte Carlo data, Eqs. (11) and (12). NN denotes site percolation in the square lattice with nearest- and next-nearest-neighbor interactions; SB means bond percolation in the same lattice with nearest neighbors only. The other cases refer to nearest-neighbor site percolation in two and three dimensions: triangular, square, honeycomb; simple cubic and face-centered cubic.

	p_c	$\ln S_F$	n_F	n_0	σ
NN	0.41	2.4	2.0	2.9	0.35
SB	0.50	2.2	3.0	3.8	0.40
tr	0.50	2.4	2.2	2.7	0.42
sq	0.59	2.2	2.6	3.0	0.46
hc	0.70	2.4	2.4	3.0	0.49
sc	0.32	1.2	2.8	1.9	0.42
fcc	0.21	2.4	3.9	3.5	0.42

sional cases of Table I are described roughly by the same curve for $\frac{1}{3}p_c \lesssim p < p_c$ (not shown). Consistent with our more direct plot in Fig. 1, these "scaling-universality" points agree somewhat better with a straight line [$\ln(S_n)$ vs n] than with a parabola: The asymptotic decay is described better by an $\exp(-\text{const}\times n)$ law than by an $\exp(-\text{const}\times n^\sigma)$ law. Thus for the paramagnetic region the somewhat incomplete data available to us seem to favor a simple $\exp(-\text{const}\times n)$ decay for $W_{n\to\infty}$, i.e., $\zeta = 1$.}

For the ferromagnetic region, data differ clearly from the paramagnetic behavior. Our analysis is consistent with the results of Ref. 13 and suggests $\ln(S_n) \propto -n^{0.35}$ for two dimensions. Also, right at p_c we find the same result, $\sum_{m=n}^{\infty} W_n \propto n^{1-\tau}$ or $W_n \propto n^{-\tau}$, with $\tau = 2 + \beta/(\beta+\gamma) \simeq 2.0$ for $n \gtrsim 10$ in two dimensions. (Note that in a scaling plot S_n vs n_s the fixed point n_F is negligible if $p \to p_c$ and $n \to \infty$; thus n_F gives a correction to scaling.)

Thus present data seem to favor a strong asymmetry[14] about p_c: Perhaps $\zeta = 1$ for the paramagnetic side and $\zeta = 1/(\beta+\gamma) \simeq 0.4$ for the ferromagnetic side. If this result is true we find, concerning the essential-singularity question, the answers given in Table II.

Of course, even more accurate Monte Carlo data never can *prove* some asymptotic behaviors, in particular, if complicated crossover effects are possible.[9] Nevertheless they would help in establishing more reliably our present tentative numerical conclusions. The square-lattice site percolation problem presumably is the easiest choice for computer simulations, and its size effects have been studied.[15] If $n \sim 10^2$, we suggest more Monte Carlo work here near $p = 0.4$, whereas $p = 0.5$ is less helpful.[16] For the square bond or triangular-site problem, where $p_c = \frac{1}{2}$ exactly, more data near $p = 0.35$ would be helpful in determining more accurately whether $\zeta = 1$. For sufficiently large samples, plots of the W_n directly instead of various sums only would be possible.

TABLE II. Does there exist an essential singularity (Taylor series with zero radius of convergence) in dilute magnets? Our answers for M' are based on the tentative Monte Carlo conclusion $\zeta(p > p_c) < 1$ and $\zeta(p < p_c) = 1$ for the ferromagnetic and paramagnetic side, respectively, with ζ defined in Eq. (6). Harris, Ref. 3, also gives yes for M but would get no for M' even in the ferromagnetic case where we have yes.

	$p < p_c$	$p > p_c$
M, Eq. (2)	yes	yes
M', Eq. (3a)	no	yes

In conclusion, to the extent made possible by the old Monte Carlo data of Ref. 6, our Table II clarified the question of essential singularities at zero magnetic field in randomly dilute low-temperature Ising ferromagnets. We criticized the assumptions of Harris[3] and found the region most profitable for future Monte Carlo studies of the same problem. Present results suggest a strong asymmetry in the asymptotic decay of the cluster-size distribution.

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