Thermal Phase Transition of the Dilute s-State Potts and n-Vector Models at the Percolation Threshold

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(Received 22 August 1980)

A general theory is given for the quenched dilute s-state Potts and n-vector models in any dimension d. It is shown that for $T \to 0$ at the percolation threshold the Potts thermal exponent ν_T equals the percolation exponent ν_p, implying a crossover exponent $\phi = 1$, for any s and d. For the n-vector model ($n > 1$), $\nu_T = \nu_p/\zeta_R$, where ζ_R is a resistivity critical exponent. Agreement with recent experiments for two-dimensional dilute Ising and Heisenberg systems is excellent.

PACS numbers: 64.60.Kw, 75.10.Hk

Dilute ferromagnets have received much attention recently, both experimentally[1,2] and theoretically,[3-7] because their comprehension is important for the general understanding of many other disordered systems. To illustrate the phenomena involved, consider an Ising model in which ferromagnetic bonds are randomly distributed with concentration p. If all the bonds are present ($p = 1$), the pure Ising model is recovered. As p decreases, the average ferromagnetic interaction also decreases. As a consequence the critical temperature decreases and approaches zero at the percolation threshold p_c. Below this value, only finite clusters of ferromagnetic bonds are present, and therefore no ferromagnetic order is possible. The special point $Q(p = p_c, T = 0)$ is very intriguing as both connectivity and thermal fluctuations become critical.[3-5] As this point is approached along the path $p \to p_c$ at $T = 0$, no thermal fluctuations are present and the critical behavior is characterized by percolation exponents. Much less is known when Q is approached along the path $T \to 0$ with $p = p_c$.

Let us briefly review the present theories[3-5] describing the thermal phase transition along this path. At p_c the system can be viewed as an Ising model defined on a typical incipient infinite cluster (IIC). This is the infinite cluster at p_c which occupies a zero fraction of the lattice. Stanley *et al.*[4] and Lubensky[5] argue that magnetic correlations spread through the IIC along an effective one-dimensional (1D) path of $\mathcal{L}$ steps. If ξ_p is the end-to-end distance of the IIC,[8] a new exponent ν_T can be defined through $\mathcal{L}^{\nu_T} \sim \xi_p$. If, at some temperature T, the Ising correlation length $\xi_2(T)$ satisfies $\xi_2(T) \sim \xi_p$, then $\mathcal{L} \sim e^{2J/kT}$, since this is the behavior of the correlation length of a 1D Ising system. Thus

$$\xi_2(T) \sim (e^{-2J/kT})^{-\nu_T}. \qquad (1)$$

Consequently ν_T defines the divergence of the Ising correlation length along the path $T \to 0$ with $p = p_c$. Since $\xi_p \sim |p - p_c|^{-\nu_p}$, one also has $\mathcal{L} \sim |p - p_c|^{-\nu_p/\nu_T}$. Since the critical behavior along the path $p \to p_c$ at $T = 0$ is given by the percolation exponent ν_p, one finds the crossover exponent $\phi = \nu_p/\nu_T$ with application of scaling.[3,4] Lubensky[5] argues that $\nu_s \leq \nu_T \leq 1$, where ν_s is the self-avoiding-walk (SAW) exponent, while Stanley *et al.*[4] make the *Ansatz* $\nu_T = \nu_s$. In two dimensions, with use of $\nu_T < 1$ and[9] $\nu_p = 1.33$, it follows that $\phi > 1$, while the SAW *Ansatz* predicts more precisely that $\phi = 1.8$. Neither of these results agree with the ϵ expansion,[6] or with the experimental data[2] on dilute Ising systems, which both give $\phi = 1$. The result $\phi = 1$ is quite puzzling, because in two dimensions where $\nu_p > 1$ it implies that the length $\mathcal{L}$ is smaller than the direct end-to-end distance, contrary to the intuitive idea from theory.[4,5] The above theories were assumed to be valid also for the dilute n-vector model. The most recent experimental data of Birgeneau *et al.*[1] for a 2D Heisenberg system give $\phi = \nu_T/\nu_p = 1.48 \pm 0.15$ (with use of $\nu_p = 1.33$), which does not agree well with the value predicted by the SAW *Ansatz*.

Although the theories described above are largely heuristic, they provide a concrete picture which can be used as starting point for further developments. In this paper, I develop a general theory which describes the properties of the s-state Potts and n-vector models on the IIC. From this theory, without any *ad hoc* conjectures, I predict the critical exponents for both models and find the predictions to agree with the available exact results and experimental data. I also make new predictions that can be tested experimentally and/or numerically.

Let us first consider the dilute s-state Potts model on a d-dimensional regular lattice. The partition function for a given bond configuration can be written as

$$Z\{J_{ij}\}=\sum_{\{\sigma\}}\exp[\beta\sum_{\langle ij\rangle}J_{ij}(s\delta_{\sigma_i\sigma_j}-1)], \qquad (2)$$

where J_{ij} are random variables which assume a value J or 0 with probability p or $q=1-p$, and $\sigma_i=1, \ldots, s$ are the Potts variables. The sum is over all nearest neighbors, and $\beta\equiv 1/kT$. The free energy F_s is obtained by averaging $\ln Z\{J_{ij}\}$ over all the bond configurations. It is easy to show (just as for the dilute Ising case) that the free energy describes bond percolation for any s in the limit $T\to 0$ and therefore we expect a general phase diagram of the form of Fig. 1.

Consider now the case $s=1$ for any value of T. It is possible to show,[10] if one starts from (2), that $\lim_{s\to 1}F_s/(s-1)$ describes bond percolation on an already diluted lattice with bond probability $p_1=1-e^{-J/kT}$. This is equivalent to a bond percolation on a full lattice with an effective bond probability $\bar{p}$ given by $\bar{p}=pp_1$. Consequently, the critical curve is given by $pp_1=p_c$, with percolation critical exponents all along this curve. In particular, as the Q point is approached along the path $T\to 0$ with $p=p_c$, the $s=1$ correlation length ξ_1 diverges as

$$\xi_1\sim(p_1-1)^{-\nu_p}\equiv(e^{-J/kT})^{-\nu_p}. \qquad (3)$$

This implies that for the $s=1$ Potts model $\nu_T=\nu_p$.

I now show that this result is true for any s. Consider the s-state Potts model defined on a typical realization of the IIC. To solve this model we apply an exact renormalization procedure, which maps this cluster into a single bond with renormalized coupling constant J' and rescaling parameter ξ_p. To show how to construct the recursion relation, let us consider first the case $s=1$. In this case the renormalized bond probability $p_1'=1-e^{-J'/kT}$ is given by the probability of getting across[11] from one end to the other of the cluster (Fig. 2). A simple calculation gives

$$p_1'=p_1^{L}[p_1^{N}+Np_1^{N-1}q_1+O(q_1^2)], \qquad (4)$$

where L is the number of *singly connected* bonds (shown as bold lines in Fig. 2), and N is the number of multiply connected bonds (shown as light lines). $O(q_1^2)$ are terms of higher order in $q_1=1-p_1$. The term p_1^{L+N} corresponds to all the bonds occupied and $Np_1^{L+N-1}q_1$ corresponds to one multiply connected bond missing. No matter which of the N bonds is missing it will be always possible to get across (see Fig. 2), hence the factor N. The eigenvalue of (4) at $T=0$ ($p_1=1$) is given by $(dp_1'/dp_1)_{T=0}=L$.

For the general s-state Potts model we decimate[12] all the internal sites of the IIC except the two ends. Following the same line as before, we find a scaling field given by $e^{-sJ/kT}$ with the same eigenvalue L at $T=0$ [see the example in Fig. 2(b) where $L=2$] and therefore the same exponent ν_T

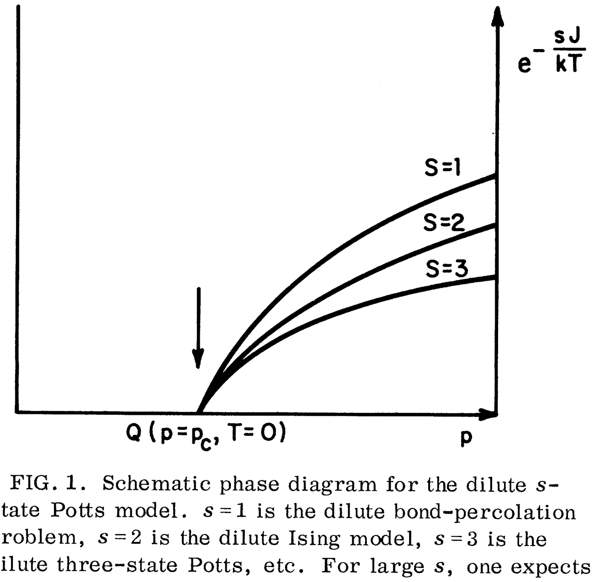

FIG. 1. Schematic phase diagram for the dilute s-state Potts model. $s=1$ is the dilute bond-percolation problem, $s=2$ is the dilute Ising model, $s=3$ is the dilute three-state Potts, etc. For large s, one expects a line of first-order phase transitions ending at a second-order phase transition that is characterized by percolation exponents.

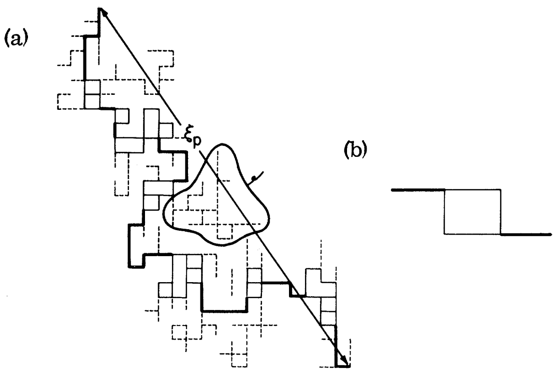

FIG. 2. (a) Computer-generated incipient infinite cluster for the bond percolation problem (from Ref. 8). The backbone bonds are shown as full lines and the "dangling ends" are shown as broken lines. For the s-state Potts model only the backbone bonds that are *singly* connected (shown as bold lines) contribute to the effective 1D length L through which the thermal correlation spreads. As an example, in (b) $L=2$. For the n-vector model ($n>1$) the effective 1D length L_R is given by the equivalent 1D resistance of the backbone. In (b) $L_R=3$.

as for $s=1$. Since for $s=1$, $\nu_T=\nu_p$, it follows that $\nu_T=\nu_p$ for any s and d. Consequently the crossover exponent $\phi=1$ for any s and d.

As a by-product we also find $\nu_T=\ln\xi_p/\ln L$, since L is the eigenvalue and ξ_p the rescaling parameter. Because $\nu_T=\nu_p$ we obtain $L^{\nu_p}=\xi_p$, which relates the percolation and the thermal exponent to the geometrical properties of the IIC. Finally, since $\xi_p \sim |p-p_c|^{-\nu_p}$, it follows that

$$L \sim |p-p_c|^{-1}. \qquad (5)$$

This relation is new in the context of percolation theory and is amenable to direct computation.

The quantity L plays the role of an effective 1D length along which thermal information is transmitted. In fact the same renormalization group, applied to a 1D length of L steps, gives the same eigenvalue at $T=0$ and therefore the same critical behavior near $T=0$. Physically only the singly connected bonds contribute to L, because the spins in the blobs of multiply connected bonds are very strongly correlated at low temperatures, and therefore do not offer any "resistance" to the spread of thermal correlations. Note that L can be *smaller* than ξ_p, and therefore there is no inconsistency in two dimensions where $\nu_p>1$.

Since $\phi=1$, I expect percolation exponents also for the susceptibility,[7] i.e. $\gamma_T=\gamma_p$. The experimental data[2] for the dilute 2D Ising system ($s=2$) are in excellent agreement with my predictions (Table I). No other experimental data exist presently for $d>2$ and $s>2$ to compare with these predictions. I hope that this theory will stimulate further experiments.

For the n-vector model ($n>1$), application of the same exact renormalization procedure near $T=0$ yields a new effective 1D length L_R. Using the steepest-descent method, one can prove (details will be given elsewhere) that this length is obtained by constructing the equivalent 1D electrical resistance along the backbone of the IIC. To be more precise, if a unit electric resistance r is associated to each bond of backbone and R is the total resistance of the backbone,[13] then $L_R=R/r$. As an example, in Fig. 2(b) $L_R=3$, as easily follows from Kirchhoff's laws.

Now the blobs do contribute to the effective length. The physical reason for this is that the spins in the blobs are not as strongly correlated, as in the s-state Potts model, because of the low energy excitations of the spin waves. Therefore a different value of the exponent ν_T can be obtained through $L_R^{\nu_T}\sim\xi_p$. Consequently $L_R \sim |p-p|^{-\zeta_R}$ with $\zeta_R=\nu_p/\nu_T$. Following the same procedure adopted before, I find that the correlation length ξ_n diverges as $\xi_n \sim (kT/J)^{-\nu_T}$ as $T\to 0$ and the crossover exponent $\phi=\zeta_R=\nu_p/\nu_T$, while[7] $\gamma_T=\gamma_p/\phi$. Using the available numerical data[14] for ζ_R and the best estimates for the percolation exponents,[10,15,16] I find the values of ϕ, ν_T, and γ_T, for $d\leq 6$ (see Table I). The agreement with the recently available experimental data of Birgeneau *et al.*[1] in two dimensions is remarkably good. I hope that experimental data in three dimensions will eventually be available for comparison also.

In conclusion, I have given, I believe for the first time, a complete unified theory for both the quenched dilute s-state Potts and the n-vector model. This theory is based on a renormalization procedure that I have been able to perform exactly near $T=0$. The available exact results and experimental data agree with the theory. I have made new predictions which, I hope, will be tested by experiments and numerical calculations.

TABLE I. Predicted exponents for the dilute s-state Potts and n-vector models for $T\to 0$ with $p=p_c$. The error bars are no more than two units of the last decimal. The experimental data are in parentheses.

	s-state Potts model			n-vector model ($n>1$)		
d	ν_T	γ_T	ϕ	ν_T	γ_T	φ
2	1.33	2.4	1	0.92	1.6_5	1.43
	(1.32 ± 0.04^a)	(2.4 ± 0.1^a)	(1^a)	(0.91 ± 0.1^b)	(1.5 ± 0.15^b)	(1.48 ± 0.15^b)
3	0.8_3	1.7	1	0.7_4	1.5	1.12
4	0.7	1.5	1	0.6_6	1.4	1.05
5	0.6	1.2	1	0.5_9	1.2	1.02
6	0.5	1	1	0.5	1	1

[a] Ref. 2.
[b] Ref. 1.

I would like to acknowledge many useful comments and conversations with F. Family, W. Klein, H. Nakanishi, S. Redner, P. J. Reynolds, H. E. Stanley, and D. Stauffer. This work was supported by grants from the U. S. Army Research Office and Air Force Office of Scientific Research.

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Discovery of a Neutral-to-Ionic Phase Transition in Organic Materials

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(Received 25 August 1980)

A systematic study of mixed stacked organic charge-transfer compounds shows that these materials are either neutral or ionic and that they range from being near the neutral-ionic boundary to being far away. Under pressure, several neutral compounds near this boundary are found to undergo a reversible phase transition to an ionic ground state. This is the first observation of a neutral-to-ionic transition in any kind of material.

PACS numbers: 61.50.Lt, 72.80.Le

Organic charge-transfer solids are composed of planar, aromatic donor (D) and acceptor (A) molecules that tend to stack either in separate donor and acceptor stacks or in mixed stacks where donors and acceptors alternate along the stack.[1] Interest in these materials has been focused on those exhibiting high conductivity, but recently has expanded to cover other properties and aspects of this large class of materials. In this paper, we report the observation of new effects in insulating charge-transfer compounds that have mixed stacks. These materials are either neutral or ionic, but we find that under pressure certain neutral compounds become ionic. This is the first observation of a neutral-to-ionic transition in any kind of material. It is thus appropriate to begin with a discussion on what factors makes one compound neutral and another ionic.

In contrast to inorganic compounds, the overlap (transfer integral) between neighboring molecules in an organic solid is small compared with other energies involved and can be neglected. In this case, the degree of charge transfer is determined by the competition between two energies[2]: $(I - A)$, the cost of ionizing a donor-acceptor (D-A) pair (I is the ionization potential of the donor and A is the acceptor electron affinity),