Ordering and Phase Transitions in Random-Field Ising Systems

Amos Maritan,[a] Michael R. Swift, Marek Cieplak,[b] Moses H. W. Chan, Milton W. Cole, and Jayanth R. Banavar[c]

Department of Physics, 104 Davey Laboratory, The Pennsylvania State University, University Park, Pennsylvania 16802

(Received 23 May 1991)

An exact analysis of the Ising model with infinite-range interactions in a random field and a local mean-field theory in three dimensions is carried out leading to a phase diagram with several coexistence surfaces and lines of critical points. Our results show that the phase diagram depends crucially on whether the distribution of random fields is symmetric or not. Thus, Ising-like phase transitions in a porous medium (the asymmetric case) are in a different universality class from the conventional random-field model (symmetric case).

PACS numbers: 05.50.+q, 64.70.Fx, 75.10.Hk

There has been considerable progress in the understanding of the physics of the random-field Ising model [1]. It is now believed that the ground state in three dimensions is ferromagnetic [2], that the continuous transition between the ordered and paramagnetic phases becomes first order when the random-field strength exceeds its tricritical value [3], and that the ferromagnet-paramagnet phase transition is characterized by irreversibility and strong history dependence [4]. The random-field Ising model is experimentally realized in dilute antiferromagnets in a magnetic field [5]. Recently, it has been proposed [6] that the phase separation of binary fluids in a porous medium [7] and the liquid-vapor critical point in a porous medium [8] may provide other experimental realizations of random-field Ising systems. The "random" porous medium provides the random field because of its preference for one of the binary fluids over the other, or for its preference of the liquid phase over the vapor phase. In order to get a critical point, not only does the temperature have to be adjusted to T_c, but also the chemical potential tuned to obtain exactly the right "negative" field on the other binary fluid or the vapor phase so that more than one pure phase can coexist. While almost all the insights regarding the random-field Ising model have been obtained for symmetric distributions of the random field, the porous-medium case corresponds to asymmetric distributions.

Recently, Wong and Chan [8] have carried out detailed thermodynamic studies of the liquid-vapor transition of ^{4}He confined in silica aerogel with a porosity ϕ of 95%. Sharp heat-capacity signatures were observed and used to map out the liquid-vapor coexistence boundary. These experimental results are in striking contrast in several ways to those expected for a conventional random-field Ising system. Specifically, the unusual features are (i) a continuous transition with an exponent $\beta=0.28\pm0.05$ (the bulk β is approximately $\frac{1}{3}$, whereas the random-field Ising model [9] is expected to have a much smaller $\beta\simeq0.05$); (ii) no evidence of hysteresis; (iii) a coexistence region that is an order of magnitude narrower than that in the bulk system; and (iv) in addition to the sharp heat-capacity signature at T_c, broad anomalies centered at a temperature slightly below T_c. The intriguing results of Wong and Chan open up several questions. Is the random-field Ising-model interpretation appropriate? Is the fractal characteristic of the aerogel [10] responsible for correlated randomness and thus a new universality class? Or is the critical behavior still bulklike in spite of the confinement in a random host?

In this Letter, we take a first crack at addressing some of the above questions. Our analysis is based on an exact solution of the *asymmetric* random-field Ising model with infinite-range exchange interactions. A rich phase diagram with several coexistence surfaces and lines of critical points is found. Our results are readily understood using simple physical reasoning and provide an explanation for several of the puzzling features of the Wong-Chan experiments. Our analysis also predicts hitherto unstudied phase transitions in a possible new universality class. Strikingly, the asymmetry is highly relevant, leading to qualitatively different behavior compared to the conventional random-field model.

Consider an Ising ferromagnet on a three-dimensional lattice with coordination number z described by the Hamiltonian

$$H=-J\sum_{\langle ij\rangle}\sigma_i\sigma_j-\sum_i\hat{h}_i\sigma_i\,, \qquad (1)$$

where $\langle ij\rangle$ refers to nearest-neighbor sites, $J>0$, $\sigma_i=\pm1$, and the probability distribution of the field $\hat{h}_i$ is given by

$$\hat{P}(\hat{h}_i)=p\delta(\hat{h}_i-h_0)+(1-p)\delta(\hat{h}_i+h_1)\,. \qquad (2)$$

A fraction p of the sites have a magnetic field h_0, whereas the rest of the sites have a field $-h_1$. For simplicity, let us first consider the case of uncorrelated random fields, at $T=0$ and in the extreme dilution limit ($p\sim0$). We further assume that h_0 and $h_1\geq0$. The case h_0 and $h_1\leq0$ is equivalent by symmetry whereas the other two cases (all $\hat{h}_i$ with the same sign) are trivial. There are three possible ground states denoted by $+$, $-$, and $+-$. These correspond to all spins being $+1$, all spins being -1, and the sites with h_0 (h_1) being $+1$ (-1), respectively. The corresponding energies per site

$(J=1)$ are $E_+=-ph_0+(1-p)h_1-z/2$, $E_-=ph_0-(1-p)h_1-z/2$, and $E_{+-}=-ph_0-(1-p)h_1-z/2+2zp$, where the $2zp$ term comes from the "broken" bonds around the sites having the field h_0. The magnetizations (per site) of the three phases are $m_+=+1$, $m_-=-1$, and $m_{+-}=2p-1$. The $T=0$ phase diagram in the h_0-h_1 plane is shown in Fig. 1(a). The bold lines indicate first-order transitions between the ground states—the point $P(h_0=z, h_1=zp/(1-p))$ is a triple point.

In order to extend our analysis to nonzero temperatures and to arbitrary p, we study the mean-field model

$$H_{\rm MF}=-(J/N)\sum_{i\neq j}\sigma_i\sigma_j-\sum_i \hat{h}_i\sigma_i\,, \qquad (3)$$

where N is the number of sites and the first sum is over all pairs of sites. The field distribution is still given by Eq. (2). Following Schneider and Pytte [11], the quenched free energy per site

$$f=\lim_{N\to\infty}\frac{1}{N}\int\prod_i[d\hat{h}_i\hat{P}(\hat{h}_i)]\ln {\rm Tr}e^{-\beta H_{\rm MF}}$$

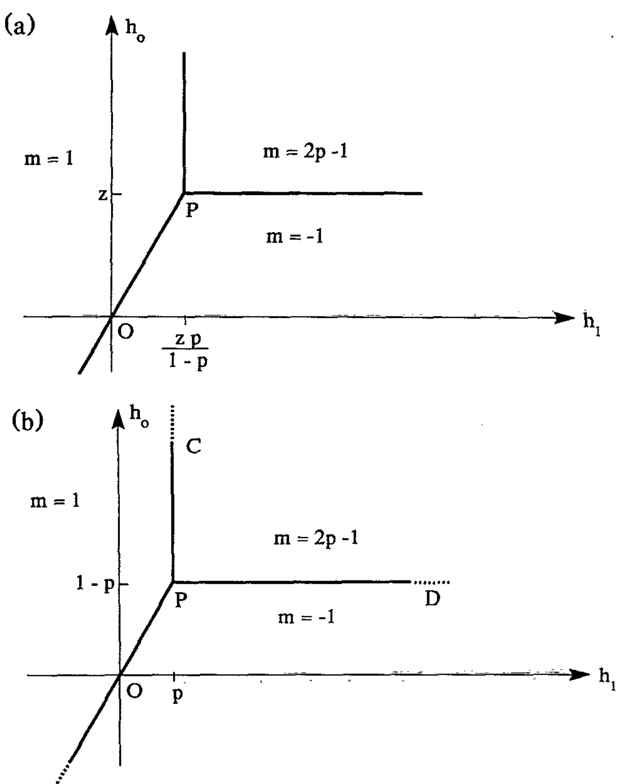

FIG. 1. (a) A two-dimensional schematic phase diagram at $T=0$ in the h_0-h_1 plane for a three-dimensional random-field Ising model in the $p\to 0$ limit. (b) A two-dimensional schematic phase diagram at $T=0$ in the h_0-h_1 plane for an infinite-range Ising ferromagnet in a random field for arbitrary p.

can be evaluated exactly to yield

$$f=Jm^2-(1/\beta)\{p\ln\cosh[\beta(2Jm+h_0)]+(1-p)\ln\cosh[\beta(2Jm-h_1)]\}\,. \qquad (4)$$

The equilibrium states are obtained from the minimum of f as a function of m at fixed J, h_0, h_1, and $\beta=1/T$ $(k_B=1)$. The phase diagram (for arbitrary p) at $T=0$ (assuming $J=\frac{1}{2}$) in the h_0-h_1 plane is shown in Fig. 1(b). The topology and the nature of the transitions are just as in the previous three-dimensional analysis.

OP, CP, and DP denote first-order transitions between + and −, + and +−, and − and +−, respectively. The triple point P has the coordinates $h_0=1-p$ and $h_1=p$. Each of the first-order transitions is manifested as a competition between the two corresponding minima in the f vs m curve, except in a neighborhood of P where all three states are at a local minimum and compete with each other. The point O refers to the pure Ising ferromagnet. As $p\to 0$, h_0 disappears from the problem and h_1 becomes an external field in the pure Ising model. The $p>\frac{1}{2}$ case corresponds to an interchange of the h_0 and h_1 axes.

The nonzero-T phase diagram can be determined using Eq. (4) and is shown schematically in a three-dimensional (h_0-h_1-T) plot in Fig. 2. Each of the three first-order lines in the $T=0$ plane (OP, CP, and DP) become coexistence surfaces terminating at the dashed lines denoting continuous transitions. The point P^* that marks the termination of the line of continuous transitions $D'P^*$ lies on the coexistence surface $CPOO'P'C'$. O' is the critical point of the pure Ising model. At $p=\frac{1}{2}$, $P^*\equiv P'$ and the lines $C'P'$ and P^*D' are interchangeable by a symmetry operation. In this limit, it is likely that O' and $P'=P^*$ are unstable fixed points whereas the lines $O'P'$ and $C'P'\cup P'D'$ are in different universality classes. The line $O'P'$ corresponds to the conventional random-

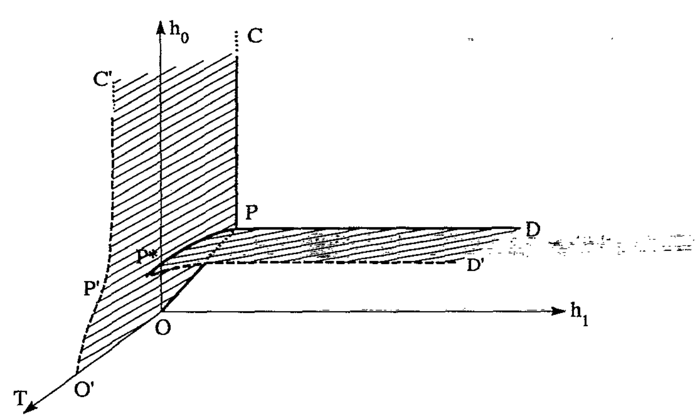

FIG. 2. A three-dimensional schematic phase diagram in the h_0-h_1-T space for an infinite-range Ising ferromagnet *in a random* field.

field Ising-model universality in the $p \equiv \frac{1}{2}$ limit.

For $p < \frac{1}{2}$, $P' \neq P^*$. We now consider the following cases: (a) Along a line with h_0=const on the $C'CPP'$ surface, the coexistence at low temperatures is between m_+ and m_{+-}, and the T_c in the high-field limit is $\sim T_{O'}(1-p)$, where $T_{O'}$ is the bulk critical temperature. (b) Along a line with h_0=const on the $O'OPP'$ surface, the coexistence is between m_+ and m_-, and as the line approaches OO', the critical temperature approaches the bulk value of $T_{O'}$. (c) Along a line with h_1=const on the $DD'P^*P$ surface, the coexistence is between m_- and m_{+-}, with the critical temperature in the infinite-field limit approaching $pT_{O'}$.

In all the above cases, the free energy as a function of m develops a third minimum when the line is close to the line PP^*. Indeed, along PP^* we find a coexistence curve shown schematically in Fig. 3. Thus at T^*, a continuous and a first-order transition occur simultaneously. There could be interesting dynamical effects depending on the state of the system: Preparing the system at $m = m_+$ leads to a clear first-order transition at $T = T^*$ with the continuous transition possibly masked due to dynamical effects associated with the first-order transition. On the other hand, preparing it with m_- or m_{+-} leads to a continuous transition followed by a first-order one.

Based on general considerations, we expect qualitatively the same phase diagram even for a broadened distribution of the $\hat{h}_i$ fields around h_0 and $-h_1$—however, too large a broadening causes some of the transitions to disappear. We will show below that we expect the Wong-Chan experiments [8] to probe the line $O'P'C'$. This line is expected to be robust since its existence relies on a sufficiently narrow distribution of h_1. On the other hand, the line P^*D' exists only as long as the h_0 field distribution is not too broad. The principal effect of the broadening is a depinning of m_+ and m_- from the $+1$ and -1 values, leading to a shrunk coexistence curve. A similar effect is caused by the presence of correlation in the random-field distribution, i.e., $\langle h_i h_j \rangle \neq \langle h_i \rangle \langle h_j \rangle$ for $i \neq j$.

In order to investigate the reliability of the infinite-range interaction approximation, we have carried out studies on a 3D simple cubic lattice using local mean-field

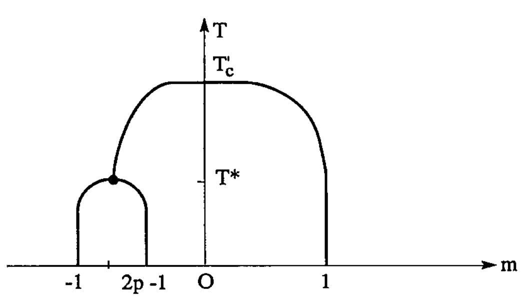

FIG. 3. The schematic coexistence curve along the line PP^*.

theory. Our calculations have been done with uncorrelated random fields and also with the h_0 field located on the sites of a fractal connected cluster (we have employed the percolation cluster for this purpose). The results obtained are in qualitative accord with the analytic calculations—the main difference being that the coexistence region is shrunk significantly when there are correlations in the random field.

We note that the phase diagram for $p \neq \frac{1}{2}$ is qualitatively different from the conventionally studied $p = \frac{1}{2}$ case. In particular, the line of continuous transitions ($O'P' \equiv O'P^*$) that describes conventional random-field behavior is now part of the line $O'P'C'$ (note that $P^* \neq P'$). O' is described by the bulk Ising exponents—since the entire line $O'P'C'$ is at nonzero temperature it seems plausible that the behavior along this entire line is governed by the bulk Ising-model fixed point. On the other hand, the line $D'P^*$ is probably in an entirely different universality class with the point P^* being a critical end point and thus having the same exponent as the rest of the line $D'P^*$. For a smooth interface, the $D'P^*$ line would be analogous to a prewetting critical line [12] and thus be characterized by $D-1$ bulk Ising exponents ($\beta = \frac{1}{8}$ for $D=3$). The effect of a rough substrate may change this critical behavior.

We turn now to an analysis of the experiment. Our model ignores the 5% of the space taken up by the aerogel. The +, −, and + − states correspond to all liquid, all vapor, and vapor except for a liquid layer coating the glass, respectively. The helium-glass interaction is modeled by h_0—indeed, the h_0 value is large and is constant during the experiment. Thus the observed phase transition is between m_+ and m_{+-} and is described by the sheet $C'CPP'$ in the phase diagram (Fig. 2). In accord with the experiment, the coexistence region is shrunk and the transition temperature is depressed. The severe shrinking of the coexistence region in the experiment is probably due to the correlated nature of the aerogel. The exponent measured by Wong and Chan [8] is consistent with the bulk value and supports the notion that the $O'P'C'$ line is governed by a bulk three-dimensional Ising fixed point. The sheet $PDD'P^*$ corresponds to a transition between the vapor phase and the vapor with a liquid layer coating the glass and is reminiscent of a prewetting transition [12] on a planar substrate, where there is a transition from a thin absorbed liquid layer (thickness $\simeq 0$) to a thick adsorbed layer. The experimentally observed transition (on sheet $C'CPP'$) corresponds to a vapor plus liquid layer going to a liquid phase and is analogous to capillary condensation. Finally, the third transition vapor→liquid (on sheet $O'P'PO$) occurs for small h_0 and would correspond to the case of a high wetting temperature with capillary condensation occurring before any wetting phenomena are observed.

The heat-capacity measurements of Wong and Chan show a large bump at temperatures lower than the critical temperature. While the sharp signatures in the heat capacity are readily attributable to the above transition within the porous media and a bulk transition outside it, the bump is probably caused by isolated pockets of liquid that undergo a rounded and shifted liquid-vapor transition. Another possible complexity is the presence of the sheet $PDD'P^*$, in the vicinity of which specific-heat anomalies may be observed.

We alert the reader that our analysis has been carried out in a mean-field context. Indeed, for a three-dimensional model with arbitrary p, there is a possibility of a richer phase diagram. Further, qualitatively different behavior may be expected depending on whether p is less than or greater than p_c. Studies of the effects of fluctuations on the phase diagram and the nature of the transitions would be quite interesting, especially the elucidation of the universality class of the transition on line $D'P^*$. An experimental study of this phase transition would also be exciting—it will entail an interchange of the h_0-h_1 axes allowing a constant-h_0 experiment to probe the surface $PP^*D'D$.

Our results suggest that the phase separation of binary liquid mixtures in porous media [7] is in a qualitatively different class from the symmetric random-field model. While critical behavior analogous to that seen in the Wong-Chan experiments ought to be seen, at least in principle, in binary mixtures, several points are worth noting: (i) The composition of the fluid in the pores has to be adjusted just right in order to probe the critical point; (ii) unlike the liquid-vapor experiment the order parameter is conserved [13] in the binary-fluid case, leading to somewhat slower relaxation; and (iii) the porous medium used in the binary-fluid experiments has been primarily Vycor glass which has much smaller (~70 Å) pores. In this case, wetting in the confined geometry [14] may lead to interesting dynamical effects and indeed might make the observation of the critical behavior of the random-field model with an asymmetric field distribution difficult.

The dilute antiferromagnetic system does provide an example for the $p=\frac{1}{2}$ symmetric random-field Ising model. In Fig. 2, while the line $O'P'$ (note that $P^*\equiv P'$ in this case) is the conventional random-field critical line, the universality class of the lines $O'P'$ and $C'P'$ (which are the same by symmetry) are unknown and would be worth investigating experimentally and theoretically. The hysteresis and history-dependent behavior associated with the conventional random-field model is presumably due to the many metastable domain states with barriers between each other that prevent equilibration. In contrast to this $p=\frac{1}{2}$ situation, in the $p\rightarrow 0$ limit the number of metastable states is the same as that for a ferromagnet thus leading to no dynamical effects. It seems plausible that there is a threshold above which there is an onset of dynamical effects and history-dependent behavior.

We are indebted to Anil Khurana, Jon Machta, and Apollo Wong for valuable discussions. One of us (J.R.B.) acknowledges the warm hospitality of Dominique Salin and the University of Paris and Flavio Toigo and the University of Padova. This work was supported by grants from the NSF, NASA, and the Petroleum Research Fund administered by the American Chemical Society.

[a]On leave from Dipartimento di Fisica, Università di Bari and Sezione Instituto Nazionale di Fisica Nucleare di Bari, Bari, Italy.
[b]Also at Institute of Physics, Polish Academy of Sciences, 02-668 Warsaw, Poland.
[c]Also at the Materials Research Laboratory.

[1] Y. Imry and S. K. Ma, Phys. Rev. Lett. **35**, 1399 (1975); for recent reviews, see T. Nattermann and P. Rujan, Int. J. Mod. Phys. B **3**, 1597 (1989); D. S. Fisher, G. M. Grinstein, and A. Khurana, Phys. Today **41**, (12), 58 (1988).
[2] J. Z. Imbrie, Phys. Rev. Lett. **53**, 1747 (1984); J. Bricmont and A. Kupiainen, Phys. Rev. Lett. **59**, 1829 (1987).
[3] A. Houghton, A. Khurana, and F. J. Seco, Phys. Rev. Lett. **55**, 856 (1985).
[4] D. S. Fisher, Phys. Rev. Lett. **56**, 419 (1986).
[5] S. Fishman and A. Aharony, J. Phys. C **12**, L729 (1979); P. Z. Wong, S. Von Molnar, and P. Dimon, J. Appl. Phys. **53**, 7954 (1982); J. Cardy, Phys. Rev. B **29**, 505 (1984); R. J. Birgeneau, R. A. Cowley, G. Shirane, and H. Yoshizawa, Phys. Rev. Lett. **54**, 2147 (1985); D. P. Belanger, A. R. King, and V. Jaccarino, Phys. Rev. Lett. **48**, 1050 (1982).
[6] P. G. de Gennes, J. Phys. Chem. **88**, 6469 (1984).
[7] J. V. Maher, W. I. Goldburg, D. W. Pohl, and M. Lanz, Phys. Rev. Lett. **53**, 60 (1984); M. C. Goh, W. I. Goldburg, and C. M. Knobler, Phys. Rev. Lett. **58**, 1008 (1987); P. Wiltzius, S. B. Dierker, and B. S. Dennis, Phys. Rev. Lett. **62**, 804 (1989).
[8] A. Wong and M. H. W. Chan, Phys. Rev. Lett. **65**, 2567 (1990).
[9] A. T. Ogielski, Phys. Rev. Lett. **57**, 1251 (1986).
[10] R. Vacher, T. Woignier, J. Pelous, and E. Courtens, Phys. Rev. B **37**, 6500 (1988).
[11] T. Schneider and E. Pytte, Phys. Rev. B **15**, 1519 (1977).
[12] S. Dietrich, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and J. L. Lebowitz (Academic, New York, 1988), Vol. 12; E. Bruno, U. M. B. Marconi, and E. Evans, Physica (Amsterdam) **141A**, 187 (1987).
[13] D. A. Huse, Phys. Rev. B **36**, 5383 (1987).
[14] A. J. Liu, D. J. Durian, E. Herbolzheimer, and S. A. Safran, Phys. Rev. Lett. **65**, 1897 (1990).