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Correlation-induced reentrant spin-glass behavior in an Ising model with random interactions

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An Ising spin-glass calculation is presented showing that stable "reentrant" spin-glass behavior, which is absent in mean-field theory, occurs when local correlations are taken into account. The method is a simple Kikuchi approximation starting from the exactly soluble classical spin-glass model proposed by van Hemmen. The resulting phase diagram is in good agreement with experiment.

Much recent theoretical work on disordered magnetic systems has concentrated on the role which spatial correlations play in magnetic spin-glass ordering[1] and the possibility of mixed phases and a "reentrant" spin-glass phase.[2,3] Model calculations which try to clarify these points of interest and aim at a spin-glass description beyond the mean-field level, are very relevant to the confrontation between theory and experiment.

Here, we investigate effects of local correlations of magnetic and spin-glass order parameters, starting from an Ising spin-glass model introduced and solved exactly, *in the mean-field limit,* by van Hemmen.[4] The replica-free mean-field solution of this "simplest nontrivial model" can already reproduce several genuine spin-glass features. Among these are the susceptibility cusp at the freezing temperature T_f and the field-induced transition away from the spin-glass phase, at finite magnetic field. However, with particular regard to dynamical behavior below T_f, the model appears too simple to describe a spin-glass in the "full sense."[5] Even if so, it appears that all *Ising* spin-glass models studied so far, including the celebrated Sherrington-Kirkpatrick (SK) model, cannot describe *stable* "reentrant" spin-glass behavior at the mean-field level.[2] In the SK model, this can be remedied, at the expense of using *Heisenberg* spins.[3]

We find that van Hemmen's mean-field phase diagram changes drastically when effects of short-range interactions are taken into account: a "reentrant" spin-glass phase appears and the new phase diagram agrees much better with experiment.

van Hemmen considered[4] the N-spin Hamiltonian

$$\mathcal{H}_N = -\frac{J_0}{N}\sum_{i,j} s_i s_j - \sum_{i,j} J_{ij} s_i s_j - h\sum_i s_i \ ,$$

with $s_i = \pm 1$, ferromagnetic exchange J_0, magnetic field h, and weakly correlated random spin-glass exchange $J_{ij} = J \times (\xi_i \eta_j + \xi_j \eta_i)/N$, assuming identically distributed, independent random variables ξ and η on every site. Randomness and frustration are abundantly present in this model and, in the mean-field limit, van Hemmen derived the equations of state ($\beta = 1/kT$):

$$\begin{aligned} m &= \langle \tanh\{\beta[J_0 m + h + Jq(\xi+\eta)]\}\rangle \ , \\ q &= \langle (\xi+\eta)\tanh\{\beta[J_0 m + h + Jq(\xi+\eta)]\}\rangle/2 \ , \end{aligned} \tag{1}$$

for the magnetization $m = \lim_{N\to\infty}(1/N)\sum_i s_i$ and spin-glass order $q = \lim_{N\to\infty}(1/N)\sum_i \xi_i s_i$ (the symmetry between ξ and η is never broken). The brackets denote the average over ξ and η.

Examining all the solutions of the equations of state and selecting those which minimize the free energy leads to very appealing phase diagrams, typically consisting of a paramagnetic ($q = m = 0$), ferromagnetic ($q = 0$, $m \neq 0$), and spin-glass ($q \neq 0$, $m = 0$) phase, meeting at a multicritical point M_c, and a mixed phase ($q \neq 0$, $m \neq 0$) at low temperatures T. However, neither in the case of a discrete distribution ($\xi = \pm 1 = \eta$), for which the spin-glass phase is even unstable at low T, nor for a Gaussian distribution of ξ and η, does van Hemmen's solution allow spin-glass behavior at "magnetic atom concentration" J_0/J greater than 1 (i.e., the multicritical point value). Experiments, though, reveal[6] that spin-glass phases extend far above $J_0/J = 1$.

In a pure ferromagnet, it is well known how to improve over a mean-field description by including effects of local correlations, for example, in a first-order Kikuchi (or Bethe-Peierls) approximation.[7] What one gains hereby is a "dimensionality dependence" of the phase diagram (relating space dimension to lattice coordination number) and an improvement of the critical-point location. In the Ising model, for instance, the Bethe-Peierls approximation correctly predicts the lower critical dimension d_{LC} to be 1 and shifts K_c in $d = 2$ from 0.25 (mean field) to 0.347 (cf. Onsager's exact value 0.441), on the square lattice.

Our first step is to replace van Hemmen's Hamiltonian by an analogous *short-range* Hamiltonian

$$\mathcal{H}_N = -\sum_{\langle ij\rangle}[J_0 s_i s_j + J(\xi_i \eta_j + \xi_j \eta_i) s_i s_j] \ ,$$

where $\langle ij\rangle$ indicates a sum over all nearest-neighbor pairs on a lattice. An external field term may be added, but we

will not consider it here. The first-order Kikuchi approximations may be described as follows. Consider a set of two nearest-neighbor sites with spin variables s_1 and s_2. The states on such a two-site system may be described by the probability distributions $p_2(s_1,s_2;\xi_1,\eta_1,\xi_2,\eta_2)$, giving the probabilities for finding spin values s_1 and s_2, when the ξ and η are known. Some obvious conditions should be satisfied, such as

$$\sum_i p_{2i} \equiv \sum_{s_i} p_2(s_1,s_2;\xi_1,\eta_1,\xi_2,\eta_2) = 1 \quad (\text{for } i=1,2) \ ,$$

$$p_2(s_1,s_2;\xi_1,\eta_1,\xi_2,\eta_2) = p_2(s_2,s_1;\xi_2,\eta_2,\xi_1,\eta_1)$$
$$= p_2(s_1,s_2;\eta_1,\xi_1,\xi_2,\eta_2) = \cdots \ .$$

These distributions can also be characterized by order parameters such as

$$Q_1 = m = \left\langle \sum_i p_{2i} s_j \right\rangle = \langle \{s_j\} \rangle \ ,$$

$$Q_2 = q = \left\langle \sum_i p_{2i} s_j \xi_j \right\rangle = \left\langle \sum_i p_{2i} s_j \eta_j \right\rangle = \langle \{s_j \xi_j\} \rangle \ ,$$

$$Q_3 = r = \left\langle \sum_i p_{2i} s_j \xi_j \eta_j \right\rangle = \langle \{s_j \xi_j \eta_j\} \rangle \ ,$$

$$\cdots \ ,$$

where $j=1$ or 2. $\langle \cdots \rangle$ indicates an average over the random variables ξ and η, while $\{\cdots\}$ indicates an average over the s variables, weighted with the probabilities p_2. The Q_k with $k>3$ contain variables on both sites simultaneously. We can also define the single-site distributions $p_1(s;\xi,\eta)$. These p_1 are functions $p_1(m,q,r)$ of m,q, and r alone. With these distributions, one defines in an obvious way the single- and two-site entropy functions:

$$S_1(m,q,r) = -\langle \{\ln p_1\} \rangle \ ,$$

$$S_2(\vec{Q}) = -\langle \{\ln p_2\} \rangle \ .$$

For a lattice with coordination number z, one then defines the following approximation to the free energy per lattice site:

$$F(\vec{Q}) = -\frac{z}{2} \langle [J_0 s_1 s_2 + J(\xi_1\eta_2 + \xi_2\eta_1) s_1 s_2] \rangle$$
$$- T \left[\frac{z}{2} S_2(\vec{Q}) - (z-1) S_1(m,q,r) \right] \ ,$$

where m,q, and r are equal to Q_1, Q_2, and Q_3, respectively. The Kikuchi approximation is then determined by minimizing F with respect to the Q_i.

The Kikuchi approximation may be approached in an alternative way, which is completely equivalent to what we have just described, but much more practical to use. Let the single-site system be determined by a Hamiltonian $\mathcal{H}_1$, which couples the spin to some molecular-field-like order parameters m_0, q_0, and r_0 on its z neighbors:

$$\mathcal{H}_1 = -z [J_0 m_0 s + J q_0 (\xi + \eta) s + r_0 \xi \eta s] \ .$$

Minimizing the free energy of this system determines m_1, q_1, and r_1 as a function of m_0, q_0, and r_0:

$$m_1 = \langle \{s\} \rangle, \text{ etc.} \ ,$$

where $\{\cdots\}$ indicates here a thermal average with respect to the Hamiltonian $\mathcal{H}_1$. The value of the minimal free energy itself is given by

$$-\beta F_1(m_0,q_0,r_0) = \left\langle \ln \sum_s \exp(-\beta \mathcal{H}_1) \right\rangle \ .$$

At this point, it is interesting to note that, up to a rescaling of the interaction strengths, van Hemmen's mean-field equations (1) are recovered simply by imposing

$$m_0 = m_1; \quad q_0 = q_1; \quad r_0 = 0 \ .$$

For the two-site system we use an analogous Hamiltonian:

$$\mathcal{H}_2 = -[J_0 s_1 s_2 + J(\xi_1\eta_2 + \xi_2\eta_1) s_1 s_2] - (z-1)\{J_0 m_0 (s_1+s_2) + J q_0 [(\xi_1+\eta_1)s_1 + (\xi_2+\eta_2)s_2] + r_0(\xi_1\eta_1 s_1 + \xi_2\eta_2 s_2)\} \ .$$

Remark that there is no term corresponding to r_0 in the direct interaction between sites 1 and 2. One may again determine the order parameters

$$m_2 = \langle \{(s_1+s_2)/2\} \rangle, \text{ etc.} \ ,$$

where $\{\cdots\}$ is now a thermal average with respect to $\mathcal{H}_2$, and a free energy F_2. The Kikuchi free energy is then defined as

$$F(m_0,q_0,r_0) = \frac{z}{2} F_2(m_0,q_0,r_0) - (z-1) F_1(m_0,q_0 r_0) \ .$$

Extremizing this free energy with respect to m_0, q_0, and r_0 yields the equations

$$m_1 = m_2; \quad q_1 = q_2; \quad r_1 = r_2 \ .$$

By exploring the free energy in the neighborhood of the solutions of this set of equations, we may furthermore determine whether or not we have found an equilibrium state (minimum of F), a metastable (relative minimum), or an unstable (saddle point or maximum) state.

When the resulting phase diagram is examined for different values of the coordination number z, we find that below $z=2$ (meaning $d \leq 1$) there are no ferro (F) or spin-glass (SG) phases at $T>0$. For $2<z\leq 3$, there is F but *no* SG at $T>0$ (in terms of Bravais lattices, $z=3$ corresponds to a honeycomb lattice in $d=2$). For $z>3$, both F and SG can exist at finite temperatures. What we have found is that the "lower critical coordination number" z_{LC} equals three for the spin-glass transition, whereas it is known to be two for ferromagnetism. A standard attitude is to associate a dimensionality d to z through $z=2d$; a relation which holds on hypercubic lattices. Our approximation would then predict $d_{LC}=\frac{3}{2}$, apparently a crude underestimation of the true lower critical dimension in spin-glasses.[1] Noteworthy, however, is that, since $\frac{3}{2}>1$, our result is qualitatively correct in showing that the SG phase disappears before F, as d is lowered.

Figure 1 shows all the equilibrium (stable) phases for $z>3$ (e.g., $z=4$) in the "concentration" (J_0/J)-"temperature" (T/J) plane. The important physical improvement over van Hemmen's phase diagrams[4] lies in the dominance of SG over F at low T, over a wide J_0/J range,

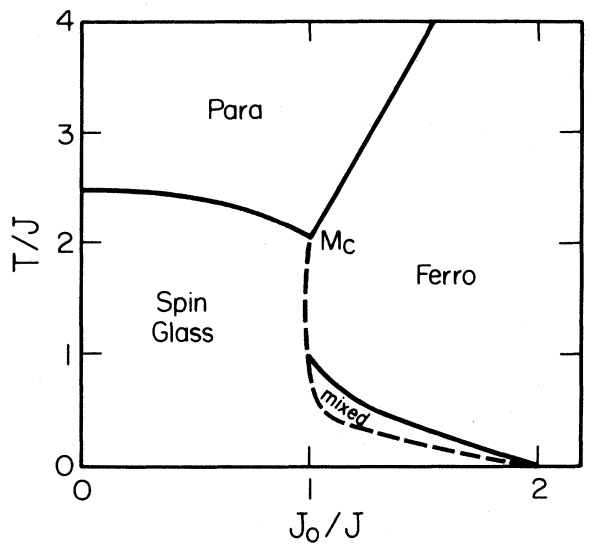

FIG. 1. Equilibrium phase diagram for $z = 4$. Phase transitions of second-order (full lines) and first-order (dashed line) separate para, ferro, and spin-glass phases and meet at a multicritical point M_c. In addition, a mixed phase occurs, separated from ferro by second-order transitions (full line).

and, hence, the occurrence of *a "reentrant" spin-glass transition.* Suppose we cool the system at fixed concentration $1 < J_0/J < 2$, starting from the paramagnetic phase. First, the system undergoes a second-order transition into a ferromagnetic phase (a small $m \neq 0$ is acquired). Computations show that m increases as T is further lowered, reaches a maximum, and then *decreases.* Next, the system enters a mixed phase via a second-order transition (apart from having $m \neq 0$, also a small $q \neq 0$ is acquired). A little further down in T, say at T_f, the system makes a first-order "freezing" transition to the pure SG phase ($m = 0$, $q \neq 0$). This transition is only weakly first-order: the jumps in m and q from the mediating mixed phase toward the SG phase, are small.

When comparing with experiment, the thermal equilibrium scenario just described may be modified to include a discussion of the metastable mixed phase which we find below the line of first-order transitions from mixed to SG and which can give rise to hysteresis effects.

Recent measurement[8] of m vs T at reentrance in amorphous spin-glasses is in remarkable agreement with our description. Experiments in metallic[9] and insulating[10] glasses also revealed a phase diagram similar to ours. The observed reentrant transition appeared to be a second-order transition directly from ferro to SG, at T_f. However, the observed irreversible or frozen moment behavior at low T was found to persist to temperatures somewhat higher than T_f, suggesting[11] a coexisting of ferromagnetic and spin-glass order somewhat above T_f. Also, from experiments on degenerate semiconductors, a transition has been reported[12] from a mixed (ferro + SG) phase to the SG phase at low T.

In closing, we would like to draw attention to renormalization group calculations on hierarchical Ising models where a reentrant disordered (paramagnetic) phase was found.[13] This is seen to be due to short-distance correlations destroying long-distance ones.

In a more extensive publication, we will present full details, discuss correlation-induced corrections to the behavior of specific heat and magnetic susceptibility, and also describe the phase diagram in the presence of an external magnetic field.

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