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Abstract. We extend a functional recursion method for random Ising chains to a system in which random interchain interactions are treated in a self-consistent field approximation. This extension is applied to a random-bond binary Ising system. The phase diagram is obtained as a function of bond strengths and bond concentrations.

During the last 15 years, considerable effort has been spent on the understanding of random magnetic systems. Although much progress has been made, results in some of these areas, notably spin-glasses, are still controversial. This is because of the inherent difficulty of performing statistical mechanics calculations on general disordered systems.

An important early attempt at this problem was the work of Hone and co-workers [1], who studied an Ising system containing small concentrations of non-magnetic or weakly coupled impurities. In that paper the authors used a transfer-matrix approach to evaluate the correlation functions for a chain of spins, and then used linear response theory to find the effect of a mean-field perturbation. The principal weakness of this approach lies in the fact that the method used to evaluate the correlation functions becomes invalid in the presence of an applied field, and becomes equivalent to consideration of an ordered chain. That is to say, the transfer matrices all commute in the absence of an applied field, and can be redistributed in blocks, but do not commute when an applied field destroys the symmetry between up and down spins.

More recent progress in the study of the random-field Ising system includes a replica-trick approach [2] or a functional recursion method to average over the random field. In particular, in an important paper Bruinsma and Aeppli [3] applied to the random-field Ising chain a functional recursion method of the type first used by Fan and McCoy [4] to study random-bond Ising chains. Bruinsma and Aeppli [3] showed that the spin probability distribution has a multi-fractal structure, and found a crossover line in the phase diagram corresponding to the onset of frustration. More recently, Maritan and co-workers [5] investigated a three-dimensional random-field Ising system with spin–spin interactions treated in the mean-field approximation. They argued that the phase diagram of such a system depends crucially on whether the distribution of random fields is symmetric or not. The one-dimensional random-*bond* Ising chain has also been investigated. Györgyi and Ruján [6] applied a functional recursion method and found that this system too has a fractal structure in the probability density of the magnetization, while Tanaka and co-workers [7] studied free-energy fluctuations. In the present paper, we extend the functional recursion method to include the random coupling between disordered Ising chains within a self-consistent mean-field approximation. We have applied this extension to a three-dimensional

random-bond binary Ising system and obtained the phase diagram as a function of bond strengths and bond concentrations.

The system we have studied is defined by the Hamiltonian

$$H = -\sum_{i,j} J_{i,j}\sigma_{i,j}\sigma_{i,j+1} - \frac{\gamma}{2}\sum_{\substack{(i,i') \\ j}} J_{ii',j}\sigma_{i,j}\sigma_{i',j}. \qquad (1)$$

Here, $\sigma_{i,j}$ is an Ising spin on site j on chain i, $\sigma_{i,j} = \pm 1$; $\langle i, i'\rangle$ means that the summation extends over near-neighbour chains i and i', and the coupling constants $J_{i,j}$ and $J_{ii',j}$ are distributed according to the *same* distribution $\rho(J)$. In what follows, we will take this distribution to be $\rho(J) = C_A\delta(J - J_A) + (1 - C_A)\delta(J - J_B)$. The motivation for this choice comes from certain random copolymers, such as poly (*p*-chloroxylelene) [8], in which there can be two types of monomers, A and B, distributed at random but with fixed average concentrations C_A and $C_B = 1 - C_A$. Near-neighbour monomers can assume two inequivalent relative orientations, and in a first approximation one considers only two bond types, defined by the two near-neighbour monomers, and characterized by coupling constants J_A and J_B. Equivalently, (1) with this choice of $\rho(J)$ is a model of a random-bond binary Ising magnet.

We consider systems in which the interchain interactions are weaker than the intrachain interactions by a factor $\gamma < 1$. In this case, it is reasonable to treat the interchain interactions in a mean-field approximation, and we replace the Hamiltonian (1) by the approximation

$$H_{mf} = -\sum_{i,j} J_{i,j}\sigma_{i,j}\sigma_{i,j+1} - \frac{z\gamma}{2}\sum_{i,j} \widetilde{J}_{i,j}\sigma_{i,j}\overline{\langle\sigma\rangle}. \qquad (2)$$

Here the double average $\overline{\langle\sigma\rangle}$ is the thermal average $\langle\sigma_{i,j}\rangle$ of $\sigma_{i,j}$ for a particular chemical structure, averaged over all disordered structures, z is the in-plane coordination number, and $\widetilde{J}_{i,j}$ is distributed according to the distribution $\rho(J)$. This mean-field approximation leads to an effective single-chain Hamiltonian

$$H_{eff} = -\sum_j J_j\sigma_j\sigma_{j+1} - \sum_j h_j\sigma_j. \qquad (3)$$

The effective field h_j at site j is given by $h_j = z\gamma\widetilde{J_j}\overline{\langle\sigma\rangle}$ (we have dropped the chain index i for ease of notation). The effective field h_j is then a random variable with the distribution $\rho(h)$, which in our case is $\rho(h) = C_A\delta(h - J_A z\gamma\overline{\langle\sigma\rangle}) + (1 - C_A)\delta(h - J_B z\gamma\overline{\langle\sigma\rangle})$. We will generalize the functional recursion method [3] to allow us to obtain the magnetization $\overline{\langle\sigma\rangle}$ and free energy F averaged over disorder of the single-chain Hamiltonian (3) as a function of temperature T, concentration C_A and the ratio J_B/J_A of the coupling constants. From the free energy, we can then obtain the entropy S and specific heat C.

The transfer matrix relating the partition function for a chain with $N + 1$ spins, $Z_{N+1}(\sigma_{N+1})$, to that of a chain with N spins is

$$T_{N+1,N} = \begin{pmatrix} e^{\beta J_N - \beta h_{N+1}} & e^{-\beta J_N + \beta h_{N+1}} \\ e^{-\beta J_N - \beta h_{N+1}} & e^{\beta J_N + \beta h_{N+1}} \end{pmatrix}. \qquad (4)$$

By introducing the polar notation $Z_{N+1}(\sigma_{N+1} = 1) = r_{N+1}\cos\theta_{N+1}$ and $Z_{N+1}(\sigma_{N+1} = -1) = r_{N+1}\sin\theta_{N+1}$ we can write

$$\lambda_{N+1}\begin{pmatrix} \cos\theta_{N+1} \\ \sin\theta_{N+1} \end{pmatrix} = T_{N+1,N}\begin{pmatrix} \cos\theta_N \\ \sin\theta_N \end{pmatrix} \qquad (5)$$

with $\lambda_{N+1} = r_{N+1}/r_N$, so that

$$\lambda_{N+1}^2 = 2e^{-2\beta h_{N+1}}\cosh 2\beta J_N\cos^2\theta_N + 2e^{2\beta h_{N+1}}\cosh 2\beta h_{N+1}\sin^2\theta_N + 2\sin 2\theta_N.$$

$$(6)$$

From (5) we can derive the recursion relation between $z_N \equiv \tan \theta_N$ and $z_{N+1} \equiv \tan \theta_{N+1}$:

$$z_N = e^{-2\beta h_{N+1}} \frac{z_{N+1} e^{\beta J_N} - e^{-\beta J_N}}{e^{\beta J_N} - z_{N+1} e^{-\beta J_N}} \equiv f(z_{N+1}, h_{N+1}, J_N). \tag{7}$$

This recursion relation allows us to find the probability distribution $P(\theta)$ of the eigenvalues $\lambda(\theta)$. From the recursion relation (7), we obtain the result that the integrated probability distribution $W(\theta)$, defined by

$$W(\theta) = \int_0^\theta P(\theta') \, d\theta' \tag{8}$$

satisfies the relation

$$W(\theta) = \int \rho(h) \rho(J) W[f(\theta, h, J)] \, dh \, dJ. \tag{9}$$

The magnetization distribution is obtained from the probability distribution $P(\theta)$ and the distributions $\rho(h)$ and $\rho(J)$ as

$$\sigma(\theta) P(\theta) = \int \frac{-\cos^2 \theta + \sin^2 \theta T_{12}(h, J)/T_{21}(h, J)}{\cos^2 \theta + \sin^2 \theta T_{12}(h, J)/T_{21}(h, J)} \rho(h) \rho(J) \, dh \, dJ \, P(\theta). \tag{10}$$

Here, $T_{12}(h, J)$ and $T_{21}(h, J)$ are the off-diagonal elements of the transfer matrices:

$$T_{12}(h, J) = e^{-\beta J + \beta h} \qquad T_{21}(h, J) = e^{-\beta J - \beta h}. \tag{11}$$

The average magnetization is then given by $\overline{\langle \sigma \rangle}$:

$$\overline{\langle \sigma \rangle} = \int \sigma(\theta) P(\theta) \, d\theta. \tag{12}$$

Other quantities of interest are the Edwards–Anderson order parameter q_{EA}

$$q_{EA} = \int [\sigma(\theta)]^2 P(\theta) \, d\theta \tag{13}$$

and the mean-square disorder fluctuation $q_{EA} - (\overline{\langle \sigma \rangle})^2$. Finally, the free energy averaged over disorder is

$$F = -\frac{1}{\beta} \int \rho(J) \rho(h) P(\theta) \ln \lambda(\theta) \, d\theta \, dJ \, dh + \tfrac{1}{2} z\gamma \int dJ \rho(J) \overline{\langle \sigma \rangle}^2$$

$$= -\frac{1}{\beta} \int \left(W(\theta = \tfrac{1}{2}\pi) \ln \lambda(\theta = \tfrac{1}{2}\pi) - \int_0^{\pi/2} W(\theta) \frac{1}{\lambda(\theta)} \frac{\partial \lambda}{\partial \theta} \, d\theta \right) dh \, dJ$$

$$+ \tfrac{1}{2} z\gamma \int dJ \rho(J) \overline{\langle \sigma \rangle}^2. \tag{14}$$

The free energy can then be obtained by self-consistently solving (12) and (9), and thermodynamic functions, such as entropy and specific heat, can then be obtained from the free energy.

We have obtained the free energy, $\overline{\langle \sigma \rangle}$, entropy and specific heat for concentrations C_A ranging from 1.0 to 0.2 and J_B/J_A ranging from 0.9 to 0.25†. For all these values of C_A and J_B, there is an onset of ordering, defined by a temperature T_c below which $\overline{\langle \sigma \rangle}$

† Note that our approach breaks down for $J_B/J_A = 0$ and large concentrations of C_B, $C_B \gtrsim 0.25$. In this case, it predicts a finite critical temperature T_c below which there is ordering. (However, the magnetization does not saturate to unity as the temperature is reduced.) This appears to be inconsistent with the percolation threshold of 0.25 for a simple cubic lattice. The likely source of this inconsistency is the neglect of interchain fluctuations, which become important near the percolation threshold.
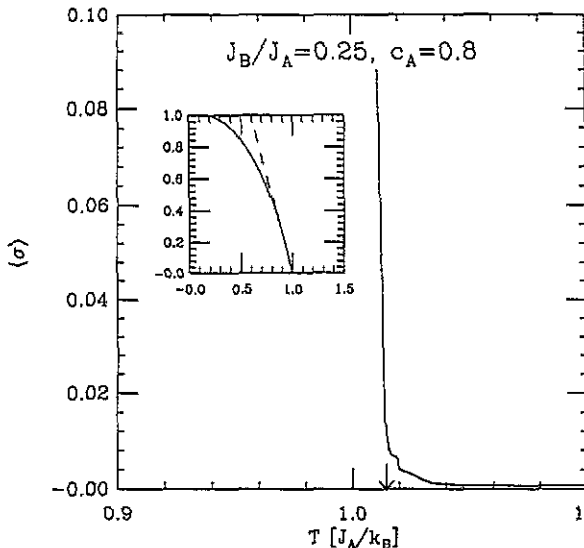
Figure 1. Detail of how $\overline{\langle\sigma\rangle}$ decreases with increasing temperature. The critical temperature T_c, indicated by the arrow, is defined as the temperature below which $\overline{\langle\sigma\rangle}$ behaves as $|T - T_c|^\beta$. Above T_c, $\overline{\langle\sigma\rangle}$ is small but non-zero for a small range of temperature. (The offset in $\overline{\langle\sigma\rangle}$ from zero at higher temperatures is due to numerical errors.) The insert depicts $\overline{\langle\sigma\rangle}^2$ as a function of temperature. For $T \gtrsim 0.9$, but below T_c, the curve is a straight line (indicated by the broken line), which demonstrates that $\beta = 1/2$.
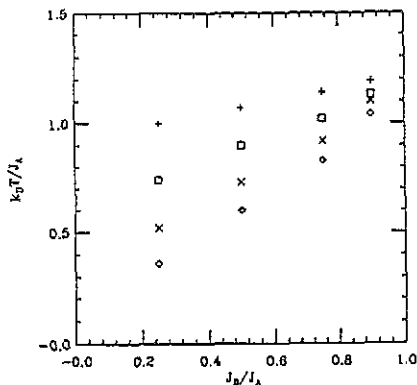

Figure 2. The critical temperature T_c is plotted against coupling constant ratio for $z\gamma = 0.25$ and for four different values of the concentration C_A, $C_A = 0.8$ (+), $C_A = 0.6$ (□), $C_A = 0.4$ (×), and $C_A = 0.2$ (◇).

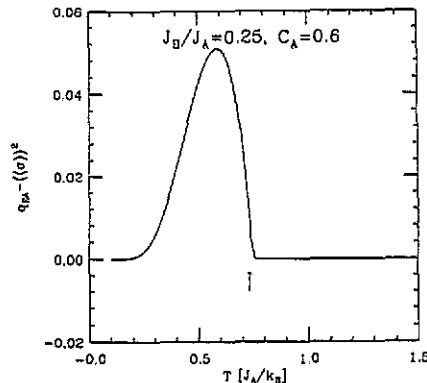**Figure 3.** The mean-square disorder fluctuation $q_{EA} - (\overline{\langle\sigma\rangle})^2$ is shown for $J_B/J_A = 0.25$ and $C_A = 0.6$. The critical temperature for this choice of parameters is 0.74 J_A/k_B and is indicated by the arrow.

behaves as $|T - T_c|^\beta$ with $\beta = \frac{1}{2}$ for temperatures near, but smaller than, T_c (see figure 1). At the temperature defined this way, there is also a discontinuity in the derivative of the entropy. The phase diagram defined by this T_c is depicted in figure 2. As the disorder increases, however, $\overline{\langle\sigma\rangle}$ remains non-zero but small for a range of temperature above this critical temperature but below the critical temperature $T_c(0)$ of the pure ($C_B = 0$) system. The reason is that, for $T < T_c(0)$, there are small regions of pure material. In each of these regions, the correlation grows to the size of the region as $T \to T_c(0)$. This gives rise to fluctuations in the magnetization density above the critical temperature T_c (see figure 3(c)). (Note that T_c is defined by the way the magnetization density *approaches* zero as the temperature increases, even though the magnetization density does not vanish at T_c. The critical temperature defined this way is nevertheless well defined.)

A measure of the disorder in the magnetization is given by the mean-square fluctuation $q_{EA} - (\overline{\langle\sigma\rangle})^2$. For structurally disordered systems, $q_{EA} > (\overline{\langle\sigma\rangle})^2$ for temperatures below T_c. This difference vanishes rapidly as T approaches zero and $\overline{\langle\sigma\rangle}$ tends to unity. This
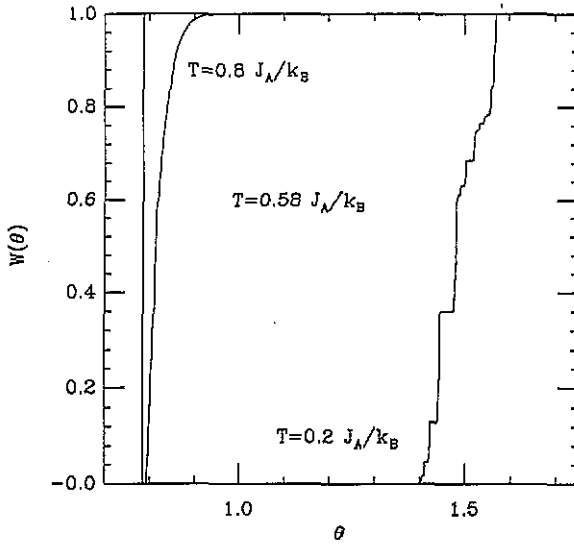
Figure 4. The integrated probability density $W(\theta)$ for the temperatures $T = 0.2\ J_A/k_B$, $T = 0.58\ J_A/k_B$, and $T = 0.8\ J_A/k_B$. Here, $J_B/J_A = 0.25$ and $C_A = 0.4$. The critical temperature for this choice of parameters is 0.52 J_A/k_B.

is depicted in figure 3. Hence, in this range of temperature, there is a higher degree of magnetic order locally than is indicated by the average magnetization for the whole system. This may help to explain why the ferroelectric phase of some random copolymers, such as the copolymers of vinylidene fluoride and trifluoroethylene (VF_2/F_3E), are more stable than those in the pure material. By mixing in a species with a stronger coupling, for example trifluoroethylene in VF_2/F_3E, one obtains a material that locally is ordered over a range of temperatures above the one at which the ferroelectric phase of the pure material becomes disordered.

It is also of interest to study the general behaviour of the integrated probability distribution $W(\theta)$ and the magnetization distribution $\sigma(\theta)$. We note the following general features. For ordered systems, $W(\theta)$ is a step function located at the fixed point of the map (7). For disordered systems, $W(\theta)$ approaches a step function at $\theta = \pi/2$ as $T \to 0$ and $\overline{\langle\sigma\rangle}$ approaches unity. At temperatures such that $0 < \overline{\langle\sigma\rangle} < 1$, $W(\theta)$ is a non-trivial function. As the temperature increases from $T = 0$, $W(\theta)$ is a function that is flat nearly everywhere, with a large number of discontinuities. At the widest plateau, $W(\theta) = c_B^2$. As the temperature continues to increase, the widths of the plateaus shrink to zero, $W(\theta)$ becomes continuous and finally approaches a single step-function at $\theta = 0$. This behaviour is shown in figure 4. The corresponding magnetization distributions are shown in figure 5. At low temperatures, the probability density is a sequence of delta functions, and so the magnetization distribution also consists of a sequence of delta functions. As the temperature increases, the distribution broadens and becomes smooth. Finally, at high temperatures the distribution approaches a single delta function.

In conclusion, we have extended the functional recursion method to systems of disordered Ising chains in an approximation in which the random interchain couplings are treated in a self-consistent mean-field approximation. We have applied this extension to a random-bond binary Ising system and obtained the phase diagram of this system. As the structural disorder increases, there is still an onset of magnetic ordering in the system, manifested by a non-zero value of $\overline{\langle\sigma\rangle}$, at a well defined temperature. The integrated probability distribution $W(\theta)$ at low temperatures becomes highly non-analytic as the disorder increases. As the temperature is increased, $W(\theta)$ becomes smooth, and finally approaches a step function at $\theta = 0$. The mean-square fluctuation $q_{EA} - (\overline{\langle\sigma\rangle})^2$ provides a
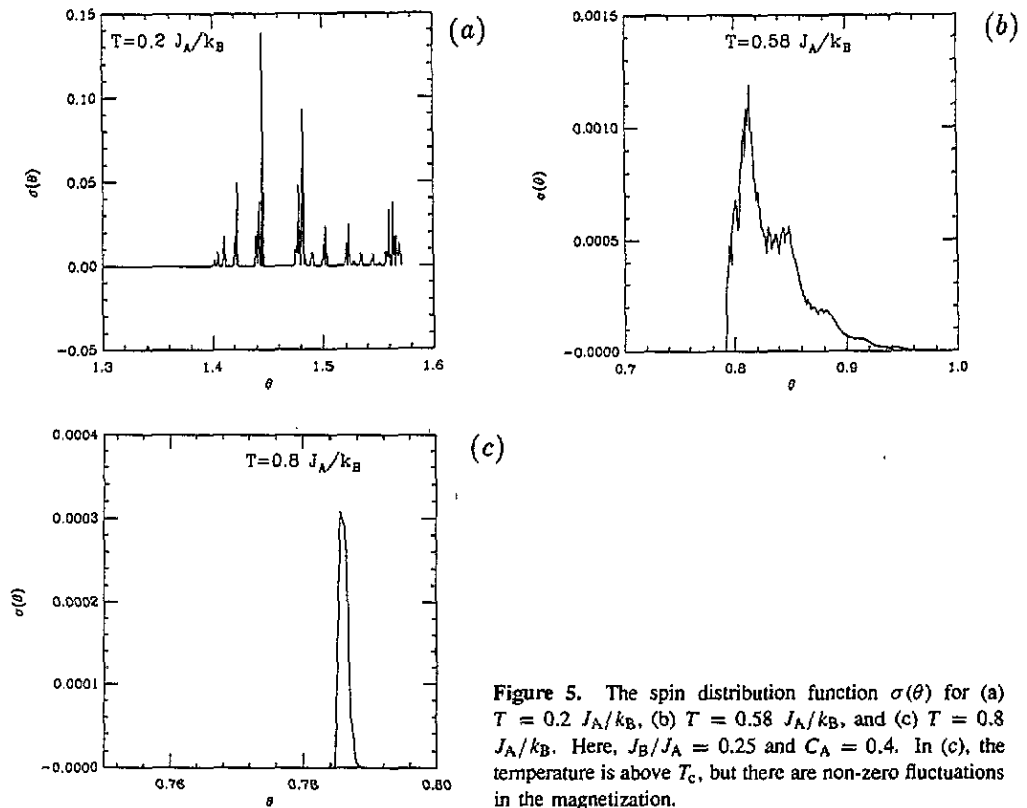
Figure 5. The spin distribution function $\sigma(\theta)$ for (a) $T = 0.2\ J_A/k_B$, (b) $T = 0.58\ J_A/k_B$, and (c) $T = 0.8$ J_A/k_B. Here, $J_B/J_A = 0.25$ and $C_A = 0.4$. In (c), the temperature is above T_c, but there are non-zero fluctuations in the magnetization.

measure of the magnetic disorder. For structurally disordered systems, $q_{EA} - (\overline{\langle\sigma\rangle})^2 > 0$ over a range of temperature below T_c.

We conclude by noting that the inherent difficulty in treating disordered Ising systems lies in the unavoidable complexity of the fractal nature of the distribution functions illustrated in figure 5. In an approach based on functional recursion, this complexity is treated exactly. Approaches based on the assumption that the transfer matrices commute do not predict this type of structure, and thus lead to results in qualitative and quantitative disagreement with the results presented here.

Acknowledgments

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