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LETTER TO THE EDITOR

Glassy behaviour in pyrochlores: a spin glass approach

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Abstract. Using a model for antiferromagnetism in disordered systems with many sublattices, it is suggested that the spin glass behaviour in pyrochlore systems is basically due to fluctuations in the spin interactions quenched on the time scale of measurements. A mean field calculation is carried out for antiferromagnetism in structures with many sublattices. By allowing quenched disorder (fluctuations) in the exchange interactions, our results clearly exhibit the interplay between the effects of lattice frustration and disorder on the system's properties. Even small fluctuations in the exchange parameters do induce sizeable glassy behaviour in structures with many sublattices. Spin glass behaviour in apparently non-disordered systems, such as certain pyrochlores, may thus be accounted for within the present context.

1. Introduction

Magnetic order in systems with a complicated lattice structure, such as pyrochlores, has been intensely studied recently [1]. In systems, such as for example $Tb_2Mo_2O_7$, strong geometrical frustration may be present in the interactions among the spins. Even for purely ferromagnetic interactions, lattice constraints may produce frustration effects in interacting Heisenberg spins [2]. The ground state and low-lying excitations of such systems are the subject of current interest and are largely unknown; notably the glass-like (meaning spin-glass-like) behaviour at low temperatures in apparently non-disordered systems [1–3]. Previous mean field theory studies [3, 4] of pyrochlores cannot account for its glassiness without disorder behaviour at low temperatures and here we shall argue that the missing ingredient is allowing for fluctuations in the interactions among the spins. These fluctuations are to be regarded as frozen on the time scale of measurements.

In the present work we study an Ising model which is good for multi-sublattice antiferromagnetic systems [5], allowing quenched disorder in the interactions which may originate in several ways: thermal, quantum, impurities etc. As Anderson pointed out long ago [5], the simple Néel theory for antiferromagnetism with two-sublattices is not applicable to most lattice structures encountered in actual antiferromagnets; a better agreement with experimental results is achieved by taking into account lattice structure. The model studied here is basically a generalization of Néel's molecular field theory and that of the Sherrington–Kirkpatrick (SK) model [6] (both valid for large d). It incorporates strong frustration effects among the sublattice spins through a mean exchange antiferromagnetic interaction J_0.

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2. The model

Here we want to study a mean field model (MFA) when the system of interest may be subdivided into many sublattices such as in pyrochlore lattices but with every spin seeing only spins on the other sublattices. In this case, to obtain sensible results, the long-range model must accordingly allow the possibility of the various orderings among the sublattices as in Anderson [5]. Thus, we consider the Hamiltonian

$$H = -\sum_{(\mu\nu)}\sum_{i,j=1}^{N} J_{ij}^{\mu\nu}\sigma_{i\mu}\sigma_{j\nu} - h\sum_{\mu=1}^{p}\sum_{i=1}^{N}\sigma_{i\mu} \tag{1}$$

where $i, j = 1, 2, 3 \ldots N$ number the sites on each sublattice, $\mu = 1, 2 \ldots p$, $(\mu\nu)$ means the distinct pairs of sublattices and h is an external magnetic field. The exchange interactions $J_{ij}^{\mu\nu}$ are assumed to be distributed according to a Gaussian

$$P(J_{ij}^{\mu\nu}) = (2\pi(J^{\mu\nu})^2/N)^{-1/2}\exp\left[-N(J_{ij}^{\mu\nu} + J_0^{\mu\nu}/N)^2/2J^2\right] \tag{2}$$

and for simplicity we assume a uniform mean and variance, $J_0^{\mu\nu} = J_0/(p-1)$ and $(J^{\mu\nu})^2 = J^2/(p-1)$, respectively. All energies are measured in units of J. For studying pure antiferromagnetism ($J = 0$) in complex lattice structures, this model is more appropriate than the simple Néel two-sublattice approach [5]. Weiss's molecular field theory for ferromagnetism is recovered for $J_0 < 0$, $p = 2$, $J = 0$, while Néel's theory is obtained for $J_0 > 0$, $p = 2$, $J = 0$. For $J_0 > 0$, $J = 0$ and very large p the model falls in the category of fully frustrated models [7]. It is a generalization of the SK model [6]. Following standard procedure the free energy per spin within the replica approach is given by

$$f = -\frac{\beta J^2}{4} + \frac{1}{p\beta}\lim_{n\to 0}\frac{1}{n}\Phi\left\{m_p^\alpha; q_p^{\alpha\beta}\right\} \tag{3a}$$

where

$$\begin{aligned}\Phi\left\{m_p^\alpha; q_p^{\alpha\beta}\right\} = &-\frac{\beta J_0}{(p-1)}\sum_{(\mu\nu)}\sum_{\alpha=1}^{n} m_\mu^\alpha m_\nu^\alpha + \frac{\beta^2 J^2}{(p-1)}\sum_{(\mu\nu)}\sum_{(\alpha\beta)}(1-q_\mu^{\alpha\beta})(1-q_\nu^{\alpha\beta}) \\ &-\ln \mathrm{Tr}\exp\Big[-\big(\beta J_0/(p-1)\big)\sum_{(\mu\nu)}\sum_{\alpha} m_\mu^\alpha\sigma_\nu^\alpha \\ &+\big(\beta^2 J^2/(p-1)\big)\sum_{(\mu\nu)}\sum_{(\alpha\beta)} q_\mu^{\alpha\beta}\sigma_\nu^\alpha\sigma_\nu^\beta + \beta h\sum_{\mu}\sum_{\alpha}\sigma_\mu^\alpha\Big]\end{aligned} \tag{3b}$$

where $\alpha, \beta = 1, 2 \ldots n$ are replica indices and m_μ^α, $q_\mu^{\alpha\beta}$ are variational parameters associated to the sublattice magnetization and spin glass order parameters. As usual, to explore the thermodynamics and possible ordering in the case of multi-sublattice antiferromagnetic systems, the first ansatz to solve (3) is to suppose a replica symmetric solution $m_\mu^\alpha = m_\mu$, $q_\mu^{\alpha\beta} = q_\mu$ together with the study of fluctuations around this solution [8–10]. The replica symmetric solution for the free energy per spin, from (3), is

$$\begin{aligned}f = &-\frac{J_0}{p(p-1)}\sum_{(\mu\nu)} m_\mu m_\nu - \frac{\beta J^2}{2p(p-1)}\sum_{(\mu\nu)}(1-q_\mu)(1-q_\nu) \\ &-\frac{1}{p\beta}\int\cdots\int\left(\prod_{\mu=1}^{p} Dz_\mu\right)\ln\left[2^P\prod_{\mu=1}^{p}\cosh(E_\mu)\right]\end{aligned} \tag{4a}$$

where

$$E_\mu = \beta\left(h - (J_0/(p-1))\sum_{\nu(\neq\mu)} m_\nu + z\beta J\left[\sum_{\nu(\neq\mu)} q_\nu/(p-1)\right]^{1/2}\right) \tag{4b}$$

$$Dz = (2\pi)^{-1/2}\exp(-z^2/2)\mathrm{d}z \tag{4c}$$

$$m_\mu = \int Dz\tanh(E_\mu) \qquad q_\mu = \int Dz\tanh^2(E_\mu). \tag{4d}$$

Study of the fluctuations of (3) around the replica symmetric ansatz (4) leads to eigenvalues whose zeros may yield the replica symmetric phase diagrams and to de Almeida–Thouless (AT) lines [8–10]. These are given by $\min[\lambda_{\mu\nu}] = 0$ where (replicon sector)

$$\lambda^{(1)}_\mu = 1 \qquad \mu = 1, 2 \ldots p \tag{5a}$$

$$\lambda^{(2)}_{\mu\nu} = 1 + (A_\mu A_\nu)^{1/2} \qquad \mu, \nu = 1, 2 \ldots p \qquad (\mu \neq \nu) \tag{5b}$$

$$\lambda^{(3)}_{\mu\nu} = 1 - (A_\mu A_\nu)^{1/2} \qquad \mu, \nu = 1, 2 \ldots p \qquad (\mu \neq \nu) \tag{5c}$$

where

$$A_\mu = \beta^2(J^2/(p-1))(1 - 2q_\mu + q^{(4)}_\mu) \qquad \mu = 1, 2, \ldots, p \tag{5d}$$

$$q^{(k)}_\mu = \int Dz\tanh^k(E_\mu) \qquad k = 2, 3, \ldots. \tag{5e}$$

As might have been anticipated, the replica symmetric solution is unstable over parts of the phase diagram calling into action replica symmetry breaking (RSB). However, even the replica symmetric solution is already sufficiently involved to be obtained for general p. Notice that all replica parameters are interdependent, such that RSB will act simultaneously on all of them. In addition, one is dealing with a parameter space much larger than the SK one for general p and it is not clear that Parisi's RSB prescription should be applied uniformly to all sublattices. For instance, one may or may not associate to each sublattice a double continuum of order parameters $q_\mu(x)$ and $\Delta_\mu(x)$, such as in the RSB approach of de Dominicis *et al* [11]. The SK model solution as found by Parisi corresponds to the gauge $\mathrm{d}[\Delta(x)]/\mathrm{d}x = -x\mathrm{d}[q(x)]/\mathrm{d}x$. It seems that one cannot simply invoke gauge invariance of the physical properties and use this gauge, since in the present model there are ergodic metastable solutions—in general, there will be p ergodic phases, p non-ergodic phases and as many metastable states—which also require breaking of replica symmetry, thus yielding a different physical state. Here we shall only consider the solutions of equations (4) and (5). Nonetheless, following previous works which consider the two-sublattice antiferromagnetic random energy model (REM) [12], extended for the present context, we find that the RSB scheme associated with ergodic metastable states is not the same as that associated with the global minimum of the free energy [13]

The sets of equations (4) and (5) have many possible solutions which depend on the values of T/J, h/J, J_0/J and the number p of sublattices considered. Here we have solved them numerically for the cases $p = 2, 4$ and 5. Our method was to iterate equations (4*d*) starting from low and high temperatures or fields. Figure 1 shows the zero field phase diagram for these cases. In zero field ($h = 0$) equations (4) and (5), for one-sublattice (with $J_0 < 0$) or $p = 2$ (with $J_0 > 0$), give the well-known SK phase diagram [8–10] where one finds the phases paramagnetic (PM), antiferromagnetic (AFM), spin glass (SG) and mixed antiferromagnetic-spin glass (MX). This is symmetrical with respect to the cases $J_0 \to -J_0$ (antiferromagnetism/ferromagnetism) for $p = 2$, and thus figure 1 would be symmetrical. For $p \geqslant 3$ there will always be present a lattice frustration among the sublattices. Notice that metastable states which appear in the AFM phase may now become unstable against breaking of replica symmetry, and this yields the line segment MO while the line ME is the instability line associated with the global minimum of the free energy, as shown for the case $p = 5$ (the multicritical point M in figure 1 is, in general, at $T/J = 1.0$ and $J_0/J = p - 1$). Not shown in figure 1, the phase boundary SG/MX for this requires full RSB calculation and shall be undertaken elsewhere. One interesting point to notice is the great increase in the spin glass

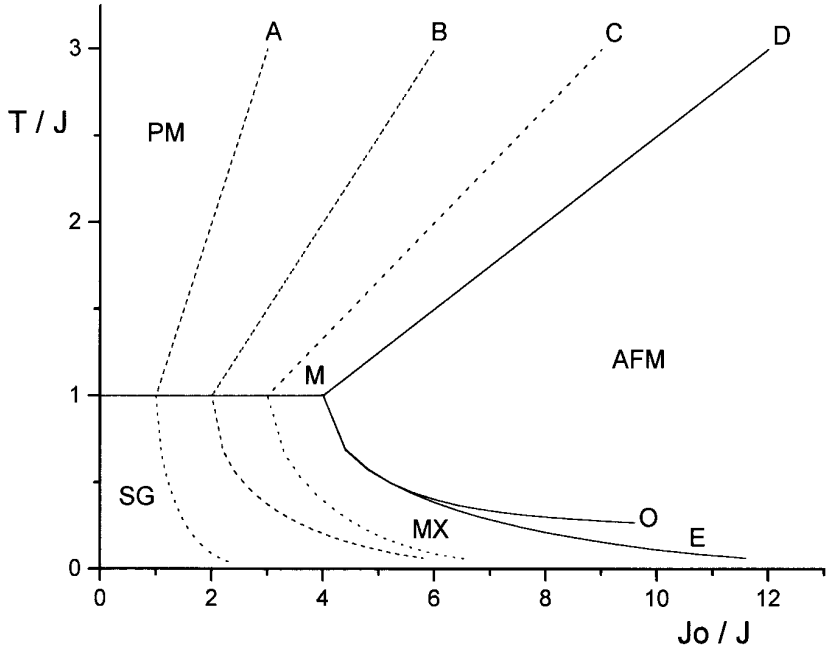

Figure 1. Zero field phase diagrams for the multi-sublattice antiferromagnetic SK model, shown for $p = 2$ (curve A), 3 (B), 4 (C) and 5 (D). The line segment MO is the instability line for the metastable solution while the line ME is for the global minimum of f. All phases are as indicated (see text). M is the AFM multicritical point.

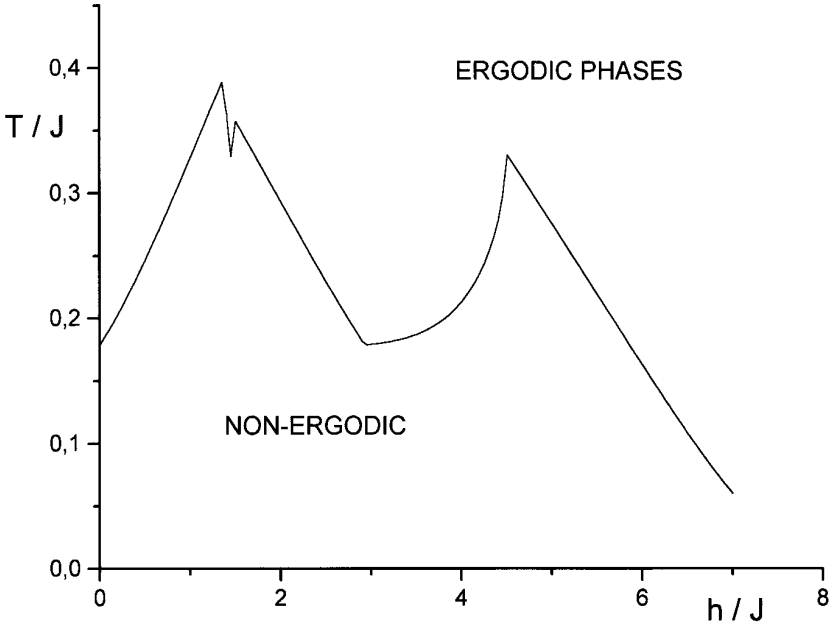

Figure 2. Field versus temperature phase boundary between the ergodic and non-ergodic phases for four-sublattices and $J_0/J = 5.1$.

phase as p is increased. For very large p there will be practically only the phases PM and SG separated by a flat phase boundary given by $T/J = 1$. This corresponds to a fully frustrated antiferromagnet where the fluctuations in the exchange parameters (J) can be arbitrarily small. The glass temperature is thus a measure of the fluctuations in the interaction parameters among the spins. A glassy behaviour has been systematically observed in pyrochlore systems [1, 2] finding a natural explanation in the present context. As long as fluctuations in the interactions are present (of a thermal, quantum or other nature) and assuming they may be considered as quenched on the time scale of the measurements, there will be a spin glass phase at low temperatures and this phase will persist in non-zero external fields. Figure 2 shows the AT line, separating the ergodic phases from the non-ergodic low temperature phases for $p = 4$ and $J_0/J = 5.1$. Notice that this phase boundary has a peaked structure for the considered values of the parameters which reflects the underlying four-sublattice system. It would be nice to have experimental data for comparison, but as far as we are aware this phase boundary has not been fully determined experimentally.

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