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A mean-field, effective medium theory of random magnetic alloys. II. The random Heisenberg model

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Abstract. A classical Heisenberg model is analysed. The interaction is of the RKKY type and only between sites randomly occupied by magnetic atoms. The possible phases are described at various temperatures and concentration of magnetic atoms. The procedure is realistic and not the 'exactly' solvable kind studied by earlier workers.

Keywords. Random magnetic alloys; spin glasses; random Heisenberg model; mean field; Heisenberg model.

1. Introduction

In an earlier paper (Mookerjee 1978, hereafter referred to as I) the author has discussed the stable phases of a random Ising model describing a disordered binary alloy of a magnetic and a non-magnetic constituent. The model incorporated a realistic long-ranged RKKY interaction between the atomic magnetic moments on the magnetic atoms and a realistic random-occupation type of statistical description. As discussed in detail in I, the approach avoided some of the serious objections raised against the previous works on the problem. Here, the formalism developed in I is applied to study the random Heisenberg model. Although the algebra closely follows that of the earlier work, the vector nature of the spins introduces features which are of interest in their own right. The short ranged order parameter is now tensorial in nature and essentially has two independent components $q_{\parallel}$ and $q_{\perp}$, which measure the single-site short ranged order parallel and perpendicular to the direction in which the symmetry is broken in the ordered phase (axis of quantisation). We shall find that there are now two types of phases with long ranged order: one in which $q_{\parallel} \neq 0$, $q_{\perp} = 0$ and one in which $q_{\parallel} \neq 0$, $q_{\perp} \neq 0$. This is a new feature absent in the Ising model.

The study of the Heisenberg model is of importance, because, unlike the Ising model, the Heisenberg Hamiltonian gives rise to the possibility of spin flip scattering of the local atomic 'spins' by the conduction electrons of the alloy. This is of special interest if we want to study the resistivity and magnetoresistance of spin-glass alloys. The freezing of the spins below the critical temperature drastically reduces the spin-flip scattering—and this may significantly reflect on the resistivity and magnetoresistance. The reduction of the spin-flip scattering certainly produces the characteristic cusp in the static susceptibility. This is the principal motivation for the work reported in this paper.

2. The classical Heisenberg model

Let us consider a model system in which the predominant energy is contributed by the indirect RKKY exchange interaction between local atomic spins situated at random and alloyed with a non-magnetic constituent. The Hamiltonian is a Heisenberg type:

$$\mathbf{H} = -\tfrac{1}{2} \sum_{k \neq j} \sum J(|\mathbf{r}_j - \mathbf{r}_k|)\, \mathbf{S}_j \cdot \mathbf{S}_k, \tag{1}$$

where $J(R) = A \cos(2k_F R)/(2k_F R)^3$ for $2k_F R \gg 1$.

The $\{\mathbf{r}_k\}$ refer to sites occupied by the magnetic atom labelled by k. Since the magnetic atoms are distributed randomly, the Hamiltonian is also random through its dependence on the $\{\mathbf{r}_k\}$. Note that this is quite different from assuming that the J_{jk}'s are themselves independent Gaussian variables. $\mathbf{S}_k$ is a normalised, classical Heisenberg spin variable characterised by two angles θ_k and ϕ_k, describing its orientation. Its magnitude is considered unity, while the normalising factor $S(S+1)$ may be absorbed into the expression for J_{jk}.

The partition function is then:

$$Z = \mathrm{Tr}\,(\theta_k, \phi_k) \exp\,[\tfrac{1}{2}\beta\, \Sigma\, \Sigma\, J(|\mathbf{r}_j - \mathbf{r}_k|)\, \mathbf{S}_j \cdot \mathbf{S}_k].$$

As before, the trace is intractable exactly, so we shall resort to the mean-field approximation. If we write $\boldsymbol{\sigma}_k = \langle \mathbf{S}_k \rangle$ (the thermal average), then, within this approximation

$$Z(\{\mathbf{r}_k\}) = \exp\,(-\tfrac{1}{2}\beta\, \Sigma\, \Sigma\, J_{jk}\, \boldsymbol{\sigma}_j \cdot \boldsymbol{\sigma}_k)\, \mathrm{Tr}\,(\theta_k, \phi_k) \exp\,(-\beta\, \Sigma\, \Sigma\, J_{jk}\, \mathbf{S}_j \cdot \mathbf{S}_k).$$

For discrete values of spin orientations the trace can be taken exactly giving rise to Langevin type functions $\Pi_j L_S(\beta \mathbf{h}_j)$, where $\{\mathbf{h}_j\}$ are local random Weiss fields given by $\mathbf{h}_j = \Sigma J_{jk} \mathbf{S}_k$ and h_j are their magnitudes. If we consider the spherical model, that is, when the spin orientations can take any directions on the unit sphere, then the trace can be replaced by integrals which give a result $\Pi_j (4\pi/\beta h_j) \sin(\beta h_j) = L_\infty$. The free energy is thus:

$$F(\{\mathbf{r}_k\}) = -\tfrac{1}{2}\, \Sigma\, \mathbf{S}_j\, \mathbf{h}_j + (1/\beta)\, \Sigma \ln\,[L_S(\beta h_j)]. \tag{2}$$

Note that the free energy is still a random function of the configuration variables $\{\mathbf{r}_k\}$. No configuration averaging is taken at this stage. To get the stable phases of the system we have to minimise the free energy with respect to the σ_j^μ ($\mu = 1, 2, 3$). As in I, the consistent solutions of the minimisation condition are

$$\sigma_k^\mu = h_k^\mu \left[L_S^{-1}(\beta h_k) \frac{\partial L_S(\beta h_k)}{\partial h_k} \right], \tag{3}$$

for all components μ and for all sites labelled by k.

So far no statistical information has been utilised. The free energy and the local magnetisations are all random functions of the occupation variables. Let us now introduce the configuration-averaged order parameters:

$$m^{\mu} = \int \sigma_k^{\mu} (\{h^{\mu}\}) \Pr (\{h^{\mu}\}) \Pi \, dh_k^{\mu},$$

$$q^{\mu\gamma} = \int \sigma_k^{\mu} (\{h^{\mu}\}) \, \sigma_k^{\gamma} (\{h^{\gamma}\}) \Pr (\{h^{\mu}\}) \Pi \, dh_k^{\mu}. \tag{4}$$

We have therefore to calculate the probability density of the local Weiss fields $\{\mathbf{h}_k\}$. We shall do this following the 'effective medium' approach as in Klein (1968) and I. If N is the number of available sites and n the number of magnetic atoms, then formally the probability density becomes:

$$\Pr (\{h^{\mu}\}) = \sum_{\mathbf{r}_1}^{N} .. \sum_{r_n}^{N} (1/N)^n \, (2\pi)^{-3} \iiint dk^{\mu} \exp [ik^{\mu} (\hat{h}^{\mu} - \Sigma \hat{J}_{oj} \sigma_j^{\mu})].$$

Note that because of the statistical homogeneity of the system the probability density is independent of the site label. Further $\hat{\mathbf{h}}$ is not the full Weiss field, but $\mathbf{h}/A$ where A is the factor appearing in the RKKY interaction and $J = A\hat{J}$. The reason why the effective medium approximation becomes necessary becomes quite obvious if we note that J_{oj} and $\boldsymbol{\sigma}_j$ are not independent variables and unlike the 'exactly solvable' models we cannot replace $\boldsymbol{\sigma}_j$ by a $\hat{\boldsymbol{\sigma}}_j$ independent of J_{oj} and neglect all other terms of the order $O(n^{-\frac{1}{2}})$. We may now combine this equation with (3) and proceed as in (I) replacing δ functions by their configuration averages. After some algebra we obtain:

$$\Pr(\{h^{\mu}\}) = (2\pi)^{-3} \iiint \Pi \, [dk^{\mu} \exp (ik^{\mu} h^{\mu})] \, (1 - G/N)^n,$$

where $$G = \sum_{\mathbf{r}} \iiint \Pi \, dx^{\mu} \{1 - [\exp \{J (|\mathbf{r} - \mathbf{r}_0|) f^{\mu} (\beta x^{\mu})\}]\},$$

$$f^{\mu} (x) = x^{\mu} L_S^{-1} (x) \, \partial L_S (x)/\partial x.$$

In the thermodynamic limit $N \to \infty$, $n \to \infty$ such that $n/N \to c$, we obtain

$$P(\{h^{\mu}\}) = (2\pi)^{-3} \iiint \Pi \, (dx^{\mu}) \exp (\Sigma ik^{\mu} x^{\mu} - cG). \tag{5}$$

We now proceed to evaluate the Fourier transform of the probability density, namely $\exp(-cG)$. We expand the exponential in the expression for G as an infinite series and neglect all moments of the RKKY interaction higher than the second. In I, we have shown that these moments fall off very fast, so that our approximation is not bad. This part of the approximation depends crucially on the nature of the RKKY interaction

$$G = iJ_0 \, \Sigma \, k^{\mu} m^{\mu} + \tfrac{1}{2} A_0^2 \, \Sigma\Sigma \, q^{\mu\gamma} k^{\mu} k^{\gamma} \quad J_0 = \sum_r J(r), \; A_0^2 = \sum_r J_0^2(r).$$

The inverse transform of the function exp $(-cG)$ can now be obtained easily to give a correlated three-dimensional Gaussian function:

$$P(h^x, h^y, h^z) = P_0 \exp\left[-\Sigma\Sigma\,(h^\mu - cJ_0\, m^\mu)\, A^{\mu\gamma}\,(h^\gamma - cJ_0\, m^\gamma)\right],$$

where $$A^{\mu\gamma} = (2cA_0^2)^{-1} \sum_\eta \mathbf{U}^T_{\mu\eta}\, q_\eta\, \mathbf{U}_{\eta\gamma}$$

$\mathbf{U}$ being the unitary matrix which diagonalises $q^{\mu\gamma}$

$$\text{i.e.}\ \sum_\eta \sum_\alpha \mathbf{U}^T_{\mu\eta}\, q^{\eta\alpha}\, \mathbf{U}_{\alpha\gamma} = q_\mu\, \delta_{\mu\gamma}, \tag{6}$$

P_0 is the normalising term.

3. A simplified classical Heisenberg model

Let us analyse the two equations (3) and (6) for a slightly simplified model. In the absence of external magnetic fields the Heisenberg model has complete spherical symmetry. This Hamiltonian by itself cannot provide a mechanism for symmetry breaking. We must begin with a finite external magnetic field and then go over to the field-free case as a limiting case of this. The system now possesses cylindrical symmetry about the axis of the external field (chosen as the z-axis). If there is no statistical anisotropy in our system we must have:

$$m^z = m,\ m^x = m^y = 0;\ q^{xx} = q^{yy} = q_2,\ q^{zz} = q_1,\ q^{xy} = q^{xz} = q^{yz} = 0.$$

Thus the matrix $\mathbf{U}$ in (6) $= \mathbf{I}$.

In the presence of an external magnetic field the probability density of the local Weiss fields get modified slightly:

$$P(\{\mathbf{h}\}) = (2\pi)^{-3} \iiint \langle \exp(-ik^z h^z)\rangle \exp\left\{\Sigma\left[ik^\mu\,(h^\mu - cJ_0\, m^\mu) - \tfrac{1}{2}\, k^\mu\, cA_0^2\, q^{\mu\mu}\right]\right\} \Pi\, dk^\mu.$$

If we now assume that the external field has a local effect h_j^z which has a Gaussian distribution with mean h_0 and variance h_1^2 we obtain

$$P(\{\mathbf{h}\}) = \frac{1}{(2\pi)^3\, cA_0^2\, q_2\, \sqrt{(h + cA_0^2\, q_1)}} \exp\left\{-(h_x^2 + h_y^2)/(2cA_0^2\, q_2) - (h_z - h_0 - cJ_0\, m)^2/[2(h^2 + cA_0^2\, q_1)]\right\}. \tag{7}$$

For the spherical model, combining (3) with (7) we obtain:

$$\begin{aligned} m &= \iiint dh_x\, dh_y\, dh_z\, (h_z/h)\, [\coth(\beta h) - 1/\beta h]\, P(\mathbf{h}),\\ q_1 &= \iiint dh_x\, dh_y\, dh_z\, (h_z/h)^2\, [\coth(\beta h) - 1/\beta h]^2\, P(\mathbf{h}) = q_{\parallel},\\ q_2 &= \iiint dh_x\, dh_y\, dh_z\, (h_x/h)^2\, [\coth(\beta h) - 1/\beta h]^2\, P(\mathbf{h}) = q_{\perp}. \end{aligned} \tag{8}$$

Unlike the Ising case, here the short-ranged order parameter is a tensor. In our simplified case, the statistical homogeneity in space reduces the six components of the symmetric $q^{\mu\gamma}$ to only two independent order parameters q_1 and q_2.

If we confine ourselves to that region of the c-T plane in the neighbourhood of the phase transitions from paramagnetic to the ordered phases, then all three m, q_1 and q_2 are small. We may expand the right side of equation (8) in terms of these variables and neglect all higher order terms. Knowing cot $hx \simeq 1/x + x - x^3/45\ldots$

$$m\,[(1 - cJ_0\,\beta/3) + c^2 A_0^2 J_0 (q_2 + 3q_1)\,\beta^3/45 + c^3 \beta^3 J_0^3\, m^2/45] = 0,$$

$$q_1\,(1 - c A_0^2 \beta^2/9) - c^2 \beta^2 J_0^2\, m^2/9 + 4c^3 \beta^4 J_0^2 A_0^2\, m^2\, q_2/135$$
$$+ 4c^3 \beta^4 J_0^2 A_0^2\, m^2\, q_1/45 + 2c^2 A_0^4 (2q_2 + 3q_1)\, q_1\, \beta^4/135 = 0,$$

$$q_2\,[(1 - c\beta^2 A_0^2/9) + 8c^2 \beta^4 A_0^4\, q_2/135 + 2c^2 \beta^4 A_0^4 q_1/135$$
$$+ 2c^3 \beta^4 J_0^2 A_0^2\, m^2/135] = 0. \qquad (9)$$

Note that we have gone to the limit of zero external field. In the presence of the external magnetic field the order parameters are not necessarily small and the equations (8) have to be numerically solved. The calculation of magnetoresistance does require such a calculation in the presence of the external magnetic field. This will be reported in a subsequent paper where we shall take up the problem of magnetoresistance.

Let us now analyse the various possible solutions of (11) which correspond to the different phases in the c-T plane.

(i) $m = 0$, $q_1 = q_2 = 0$. This is the paramagnetic phase in which there is neither long- nor short-ranged ordering. The lifetimes of the vector spins in any direction are very small and the spins flip from direction to direction driven by the random thermal field of the heat bath in which the system is immersed. The net magnetisation is zero. Such a picture is characteristic of the paramagnetic phase.

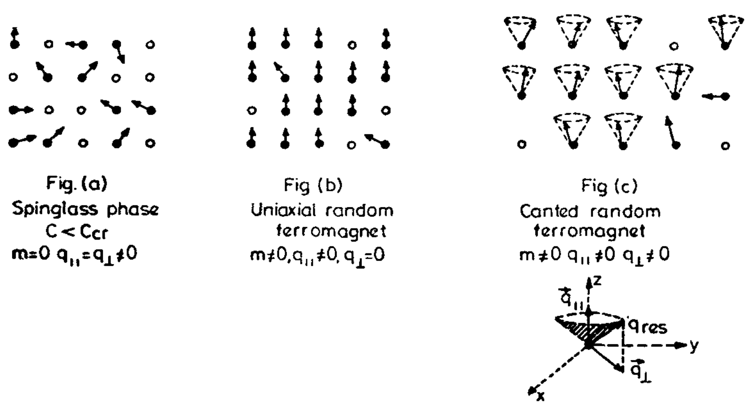

Figure 1. Schematic diagrams of frozen spins in (a) the spin glass phase (b) the 'uniaxial random' ferromagnetic phase and (c) the 'canted random' ferromagnetic phase.

(ii) $m = 0$, $q_1 = q_2 = 3\ (cA_0^2 - 9\beta^2)\ c^2A_0^4\ \beta^2$. Note that in the configuration-averaged formalism the phase corresponding to this solution reverts to complete spherical symmetry. This phase is stable if $k_B\ T \leqslant \sqrt{c}\ A_0/3$ and the paramagnetic phase is separated from this by the parabola $c = 9k_B^2\ T^2/A_0$ in the c-T plane.

The interpretation of the q-s is of interest. It can be shown that

$$q_{\parallel} = q_1 = \lim_{t\to\infty}\ [\langle S_z(t)\ S_z(0)\rangle],$$

$$q_{\perp} \equiv q_2 = \lim_{t\to\infty}\ [\langle S_x(t)\ S_x(0)\rangle] = \lim_{t\to\infty}\ [\langle S_y(0)\ S_y(t)\rangle], \text{ (Mookerjee 1977)}$$

$q_1 = q_2 \neq 0$, then, corresponds to a phase where the spins are frozen in completely random directions (which explains the spherical symmetry in the configuration-averaged description). This is the traditional spin-glass phase *a la* Edwards-Anderson ideas. Figure 1(a) shows a schematic diagram of spin-configurations in this phase.

(iii) If $T \geqslant \sqrt{c}\ A_0/3k_B$, then the only solution for q_2 is $q_2 = 0$. In this range if we further have $T \leqslant cJ_0/k_B$, then a possible set of solutions is

$$m = (cJ_0/3k_BT - 1)\ (1 - cA_0^2/9k_B^2T^2)\ (cJ_0^2/3A_0^2 + 8c^2J_0^2/27k_B^2T^2 -$$

$$4cJ_0/3k_BT)\ 15k_B^3T^3/c^2A_0^2J_0,$$

$$q_1 = (cJ_0/3k_BT - c^3J_0^3m^2/45k_B^3T^3)\ 15k_B^3T^3/c^2A_0^2J_0)\ q_2 = 0.$$

This is a phase with long-ranged order characteristic of a ferromagnetic phase. The spins in the direction of the broken symmetry (z-direction) have long lifetimes—it has both long-ranged and (consequently) short-ranged order. However, the spins in other directions are not frozen. Thus the frozen spins which constitute the long-ranged ordering has frozen components only in the z-direction. Figure 1(b) shows a schematic picture of these frozen spins. We may describe this phase as a 'uniaxial' random ferromagnetic phase. This phase is separated from the paramagnetic phase by the linear boundary $T = cJ_0/k_B$.

(iv) If $T \leqslant \sqrt{c}\ A_0/3k_B$ then we have the solution $m \neq 0$, $q_1 \neq 0$, $q_2 \neq 0$. In this region of the c-T plane, it is this and not the uniaxial random ferromagnetic phase that gives the lower free energy. Again, there is both long-ranged order and freezing of the z-component of the spins. However, the spin-components in the other two orthogonal directions are also frozen, but in random orientations. The resultant vector spins occupy random directions, but lie on the surface of cones whose axis coincides with the z-axis and whose vertex lies on the lattice sites. Because of the long-ranged order in the z-components, there is a resultant spontaneous magnetisation. Figure 1(c) shows a schematic picture of the frozen spins in this phase. We may call this the 'canted' random ferromagnetic phase.

Figure 2 describes the phase diagram in the c-T plane. Note however, that the spin-glass-ferromagnetic phase boundary must not be taken seriously. Near this boundary neither q_1 nor q_2 is small, so the expansion method fails. Even if we were to locate this boundary by numerically solving (8), we should note that the actual

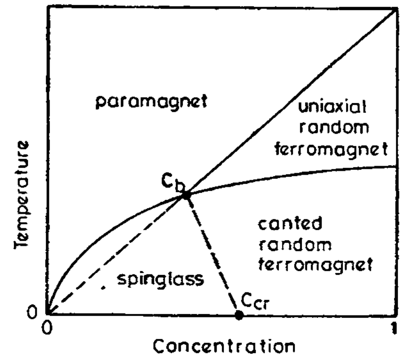

Figure 2. The phase diagram in the c-T plane for the random Heisenberg model.

spin glass ferromagnetic transition is not as simple as that described by this model. Near this boundary clusters gain in importance. We have completely ignored such mictomagnetism in our model. However, we may assert that the spin glass phase occurs in the low concentration and the ferromagnetic phases in the high concentration regimes. In the $T\to 0$ limit the critical concentration separating the spin glass from the ferromagnetic phases is $c_0=(3/8)^{\frac{1}{2}}\pi\, c_b$, where c_b is the concentration at the tricritical point where all the phases meet.

The phase analysis of the Heisenberg model is not as simple as that of the Ising model and has specific features of its own. Experimental distinction between the ferromagnetic phases: the 'uniaxial random' and 'canted random', is not available to this date. We cannot pronounce on the physical relevance of the two distinct phases.

Although we have analysed the spherical model, the analysis of models with generalised spins S is exactly similar, except that $(\coth\beta x-1/x)\,x^k/x$ is replaced by $x^k L_S^{-1}-1\,(\beta x)\;\partial L_S(\beta x)/\partial x$ which for small x gives $\simeq x^k\,(x-a_S\,x^3\ldots)$ where a_S is a known function of S. The phases and phase diagrams are exactly similar with only the specific numbers being different. A full calculation on the quantum Heisenberg model has not been carried out, but we do not expect the phase diagram to be different, although the exact shape of, say, the susceptibility is different. As far as the information sought in this paper is concerned, we do not expect anything new from the quantum calculations.

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