

A mean-field, effective medium theory of random binary alloys IV. The Heisenberg model with competing interactions and the mixed phase

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Abstract. The Heisenberg model with competing interactions is studied within a mean field, effective medium approach. The phase diagram indicates the existence of a mixed or randomly canted phase just beyond the spin glass regime. Experimental evidence of such a phase in alloys like FeCr is studied.

Keywords. Random magnetic alloys; Heisenberg model; mixed phase.

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1. Introduction

In magnetic systems with competing interactions there is strong evidence of a spin glass or a mixed magnetic phase in the concentration range between the ferro to antiferromagnetic transition. For example, in the magnetic phase diagram of $\text{Fe}_c\text{Ni}_{0.85-c}\text{Mn}_{0.15}$ and $\text{Fe}_c\text{Ni}_{0.8-c}\text{Mn}_{0.2}$ alloys, the ground state in the transition region is either a spin glass or a mixture between ferromagnetic and spin glass (Menshikov *et al* 1981). In an earlier paper Chen and Lubensky (1977) indicated that the high concentration regime may be subdivided into two further regions depending on whether the perpendicular component of the spin (perpendicular to the direction in which symmetry is broken) is frozen or not. However, they mentioned this very briefly in passing, and certainly their phase diagram, with the mixed phase occupying the entire high concentration regime at $T = 0$, is nowhere near experimental observation. Mookerjee (1979) and Medvedev (1979a) using slightly different approaches first focussed on this phase. The former called it the randomly canted ferromagnetic phase while the latter used the name asperomagnet. Around the same time Villain (1979) introduced the idea of the semi-spin glass to explain experimental results. Despite different nomenclature the basic ideas behind all these works are the same. The mixed phase is perhaps a name more widely accepted. The existence of this phase has found ample experimental justification of late (Mookerjee and Roy 1983 and references therein), not the least of these are the neutron scattering experiments on $[\text{Zn}_c\text{CO}_{1-c}]_A[\text{COTi}]_B\text{O}_4$ compounds with $0.2 < c < 0.8$ by Hubsch and Gavaille (1982).

In this paper we study the phases of a random Heisenberg model with competing interactions, focussing specially on the location of the mixed phase on the T - c diagram. We disagree with parts of the analysis of Medvedev (1982b) which arise perhaps from the approximations introduced by him. His predicted phase diagram, *e.g.* for FeCr alloys seemed at that time to agree with the experimental data of Mitchell and Gott (1972) and

Rode (1979); but the recent more careful work of Burke and Rainford (1983a) shows that unlike earlier assertions these alloys do not have intersecting $T_C(c)$ and $T_N(c)$ curves, but rather T_C and T_N precipitously drop near 19 and 16 at. % of Fe respectively, and between these concentrations we have a spin glass phase. In the region 20 to 30 at. % of Fe we have a behaviour which is very much like the mixed phase of AuFe (Loegl 1975), whereas between 16 and 9 at. % of Fe the behaviour is like an antiferromagnetic mixed phase (Burke and Rainford 1983b). Our analysis however, gives a phase diagram picture which is in good qualitative agreement with the experimental results.

Our approach will be a mean-field one. The question may arise: what is the necessity of going through another mean field approximation when several exist? Whenever any new attempt is made to look at spin glasses from this viewpoint, the statement is continually made these days that the Sherrington-Kirkpatrick type of approach or its sophistications by Parisi and others is the complete answer, at least in the mean field approximation. It is time to reflect why this statement is not completely correct. At the basis of these approaches SKP and crucial to it (as it then allows neglect of otherwise intractable series of terms) is the assumption of an infinitely long-ranged interaction which is *independent* of the distance separating the interacting centres: hence *infinitely weak*. No realistic interaction in any real system can fit this model. The RKKY, for instance, is long ranged, but decays faster than R^{-3} with distance R so that even qualitatively the model may be singular:

(i) in any configuration averaging in this model, very unlikely configurations are given great weightage. Configuration averaging of the free energy is the first step in the SKP approach (Kumar 1983).

(ii) Such an interaction matrix independent of the distance R has no localised eigenvalues. Any realistic matrix decaying faster than R^{-3} has localised eigenvalues (Economou and Antoniou 1977) and this is crucial to the understanding of a large body of experimental results in real spin glasses (Mookerjee and Chowdhury 1983).

It is thus, we believe, imperative to examine alternative approaches. Approaches which do not assume an infinitely long-ranged, infinitely weak interaction. One such approach, which we shall suitably enlarge and modify, is that of Klein (1968). Our main interest will be to focus again on the mixed phase. There seem considerable differences between the results of Chen and Lubensky's (1977) SKP type of approach and those of ours. The q_{\parallel} and q_{\perp} , m equations do not decouple in our approach for dimensions ≥ 2 (neither do they in that of Medvedev). Unlike the former, our phase diagram shows both the mixed phase as well as the ferromagnetic phase the latter at very high concentrations of the ferromagnetic constituent. In fact, the phase diagram of Chen and Lubensky with its straight line phase boundaries, is too crude for even reasonable qualitative comparison with experiment. Another, and more recent prediction about the mixed phase arising out of the SKP approach is that of Gabay and Toulouse (1981). They predict *two* low temperature transitions, which some experimentalists are at pains to 'prove' (Campbell *et al* 1983). Their approach is based on the mathematically beautiful but physically abstruse replica symmetry breaking ideas of Parisi. The physical significance of these transitions is difficult to visualise in realistic terms. Alternative explanations based on anisotropic interactions are also being put forward. This adds to our belief that alternative approaches should also be tapped. Even if the final result is not dissimilar to the SKP model, the exercise will be useful in view of the above mentioned serious reservations.

2. Formalism

We shall start with a random Heisenberg model with isotropic exchange interaction which depends on the occupation of the sites by different kind of magnetic atoms of the alloy.

The Hamiltonian is

$$H = -\frac{1}{2} \sum_k \sum_{k \neq j} J(|\mathbf{r}_k - \mathbf{r}_j|) \mathbf{S}_j \cdot \mathbf{S}_k. \quad (1)$$

J decays rapidly with $|\mathbf{r}_k - \mathbf{r}_j|$ but need not necessarily be the nearest neighbour only. The interaction is random depending upon the types of atoms occupying the sites \mathbf{r}_k and \mathbf{r}_j . In binary alloys without short ranged chemical ordering it could be J^{AA} , J^{AB} or J^{BB} with probabilities c_A^2 , c_B^2 and $2c_A c_B$. We shall neglect all anisotropies arising out of dipolar or Dzyloshinskii interactions. In a large class of alloys such anisotropies are indeed large and subsequently we shall have to study the stabilities of our phases against these anisotropies.

The free energy within the mean field approximation is

$$F = \frac{1}{2} \left[\sum_{ij \in AA} J_{ij}^{AA} \sigma_i^A \cdot \sigma_j^A + \sum_{ij \in AB} J_{ij}^{AB} (\sigma_i^A \cdot \sigma_j^B + \sigma_i^B \cdot \sigma_j^A) + \sum_{ij \in BB} J_{ij}^{BB} \sigma_i^B \cdot \sigma_j^B \right] - k_B T \left[\sum_{i \in A} \ln L_S(\beta h_i^A) + \sum_{i \in B} \ln L_S(\beta h_i^B) \right], \quad (2)$$

where σ_i^α ($\alpha = A, B$) are the random, local magnetisations $\langle \mathbf{S}_i \rangle$ and \mathbf{h}_i^α are the local, random molecular fields

$$\begin{aligned} \mathbf{h}_i^A &= \sum_j J_{ij}^{AA} \sigma_j^A + \sum_j J_{ij}^{AB} \sigma_j^B, \\ \mathbf{h}_i^B &= \sum_j J_{ij}^{BA} \sigma_j^A + \sum_j J_{ij}^{BB} \sigma_j^B. \end{aligned} \quad (3)$$

Note that the local molecular fields also depend on the type of atom occupying the site under consideration. In canonical spin glasses B is non-magnetic and there is no local magnetisation at the B sites.

We shall not configuration average at this stage, but obtain the stable phases by minimising the free energy with respect to the local magnetisations. This leads to

$$\begin{aligned} \sigma_{k\mu}^\alpha &= (h_{k\mu}^\alpha / h_k^\alpha) [L_S^{-1}(\beta h_k^\alpha) \partial L_S^\alpha(\beta h_k^\alpha) / \partial h_k^\alpha] = F_{k,\mu,S}^\alpha, \\ h_k^\alpha &= \left[\sum_\mu h_{k\mu}^{\alpha 2} \right]^{1/2}. \end{aligned} \quad (4)$$

Here μ labels the component (x, y, z), α labels the type of atom A or B occupying the site \mathbf{r}_k labelled by k . $F_S(x)$ is either the Langevin or the Brillouin functions. The local magnetisation are random, and we can introduce the following configuration averaged order parameters:

$$\begin{aligned} m_\mu^\alpha &= \int \sigma_{k\mu}^\alpha(\{h^\alpha\}) \Pr(\{\mathbf{h}^\alpha\}) \prod_\eta dh_{k\eta}^\alpha \\ q_{\mu\nu}^\alpha &= \int \sigma_{k\mu}^\alpha(\{h^\alpha\}) \sigma_{k\nu}^\alpha(\{h^\alpha\}) \Pr(\{\mathbf{h}^\alpha\}) \prod_\eta dh_{k\eta}^\alpha \end{aligned} \quad (5)$$

Averaging is done over probability distributions of the local molecular fields. Our main problem is now to calculate the probability distribution of the local molecular fields \mathbf{h}^A and \mathbf{h}^B . Because of statistical homogeneity in space these are not site dependent. The method adopted is very similar to that of Mookerjee (1979) and Mookerjee and Roy (1983) but modified to take into account several magnetic constituents.

It is easy to see from (3) that the local molecular field depends upon the relative positions of the A and B atoms with respect to the site \mathbf{r}_i under consideration. In other words, if we define a 'configuration' to be a particular arrangement of N_1 , A atoms and N_2 , B atoms on the $N = N_1 + N_2$ lattice sites, then the local molecular field randomly varies from configuration to configuration. We may write the probability distribution of \mathbf{h}_i as the radon transform

$$\text{Pr}_A(h_{i\mu}^A) = \langle\langle \delta[h_{i\mu}^A - \sum J^{AA}(|\mathbf{r}_i - \mathbf{R}_j|)\sigma_{j\mu}^A - \sum J^{AB}(|\mathbf{r}_i - \mathbf{S}_j|)\sigma_{j\mu}^B] \rangle\rangle_{\text{av}}. \quad (6)$$

The site \mathbf{r}_i is occupied by an A atom. Here $\mathbf{R}_j, \mathbf{S}_j$ denote the positions in a particular configuration of the A and B atoms on the lattice. In fact, in a given configuration the set $\{\mathbf{R}_j, \mathbf{S}_j\}$ is just some permutation of the lattice sites $\{\mathbf{r}_k\}$ ($\mathbf{R}_j, j = 1 \dots N_1; \mathbf{S}_j, j = 1 \dots N_2; \mathbf{r}_k, k = 1 \dots N = N_1 + N_2$). The average is taken over all different configurations.

We may now incorporate (3) and (4) into (6) and rewrite the δ -function in (6) as an exponential integral. We finally obtain:

$$\begin{aligned} \text{Pr}(h_{i\mu}^A) &= (2\pi)^{-3} \int d^3 k \exp(i\mathbf{k} \cdot \mathbf{h}_i^A) \sum_C P(\mathbf{C}) \int \dots \\ &\int \sum_{j \in A} dh_{j\mu}^A \delta[h_{j\mu}^A - \sum J_{ij}^{AA} \sigma_{j\mu}^A - \sum J_{ij}^{AB} \sigma_{j\mu}^B] \\ &\int \dots \int \prod_{j \in B} dh_{j\mu}^B \delta[h_{j\mu}^B - \sum J_{ij}^{BA} \sigma_{j\mu}^A - \sum J_{ij}^{BB} \sigma_{j\mu}^B] \exp[-ik(\sum J_{ij}^{AA} F_{j\mu,s}^A \\ &\quad + \sum J_{ij}^{AB} F_{j\mu,s}^B)]. \end{aligned} \quad (7)$$

\mathbf{C} is a configuration and $P(\mathbf{C})$ is its probability. We shall assume that all configurations are equally probable. As it stands, the complicated integral equation (7) is not tractable. It is at this point that we introduce our only crucial approximation other than the mean field. This approximation was first introduced by Klein (1968) and discussed in detail by Mookerjee (1978, 1979). It entails replacement of the δ -functions on the right side of (7) by their configuration averages. It is explicit from (7) that the distribution of \mathbf{h}_i^A involves the other \mathbf{h}_j^A at all sites, but through our approximation we replace the contribution of these other random variables by a configuration averaged effective contribution. In this sense the approximation is akin to the effective medium approximations.

Within this approximation:

$$\begin{aligned} \text{Pr}_A(h_{i\mu}^A) &= (2\pi)^{-3} \int d^3 \mathbf{k} \exp(i\mathbf{k} \cdot \mathbf{h}_i^A) \sum_C P(\mathbf{C}) \int \dots \int \prod dh_{j\gamma}^A dh_{j\gamma}^B \text{Pr}_A(h_{j\gamma}^A) \text{Pr}_B(h_{j\gamma}^B) \\ &\prod_{j\mu} \exp[-ik_{\mu}(J^{AA}(|\mathbf{r}_i - \mathbf{R}_j|) F_{j\mu,s}^A + J^{AB}(|\mathbf{r}_i - \mathbf{S}_j|) F_{j\mu,s}^B)]. \end{aligned} \quad (8)$$

Let us focus on the last exponential term on the right side. As we go over the configurations \mathbf{C} , the factors $J^{AA}(|\mathbf{r}_i - \mathbf{R}_j|)$ and $J^{AB}(|\mathbf{r}_i - \mathbf{S}_j|)$ goes over the values

$J^{AA}(|\mathbf{r}_i - \mathbf{r}_k|)$ and $J^{AB}(\mathbf{r}_i - \mathbf{r}_k)$ as A and B occupy the various lattice sites \mathbf{r}_k . In our model these are different values depending on the distance $|\mathbf{r}_i - \mathbf{r}_k|$, unlike the SKP model where there are only two possibilities J^{AA} and J^{AB} . Thus as we go over all configurations \mathbf{C} with equal probability, the factor $\exp[-ik_\mu J^{a\beta}(|\mathbf{r}_i - \mathbf{R}_j|)F_{j\mu,s}^\beta]$ takes on N different values $\exp[-ik_\mu J^{a\beta}(|\mathbf{r}_i - \mathbf{r}_k|)F_{k\mu,s}^\beta]$. We may thus carry out the configuration averaging in (8) to obtain:

$$\begin{aligned} \Pr_A(h_{i\mu}^A) &= (2\pi)^{-3} \int d^3\mathbf{k} \exp(i\mathbf{k} \cdot \mathbf{h}_i^A) \int \prod dh_{j\gamma}^A dh_{j\gamma}^B \Pr(h_{j\gamma}^A) \Pr(h_{j\gamma}^B) \\ &\left\{ \frac{1}{N} \sum_k \exp[-ik_\mu J^{AA}(|\mathbf{r}_i - \mathbf{r}_k|)F_{k\mu,s}^A] \right\}^{N_1} \left\{ \frac{1}{N} \sum_k \exp[-ik_\mu J^{AB} \right. \\ &\left. (|\mathbf{r}_i - \mathbf{r}_k|)F_{k\mu,s}^B] \right\}^{N_2}. \end{aligned} \quad (9)$$

As discussed earlier (Medvedev 1979; Mookerjee 1978, 1979) in writing (9) from (8) we include the 'multiple occupancy' terms. However, in the thermodynamic limit the contribution of these terms is a negligible fraction of the whole.

A simple rearrangement then allows us to write (9) as:

$$\begin{aligned} \Pr(h_{i\mu}^A) &= (2\pi)^{-3} \int d^3\mathbf{k} \exp(i\mathbf{k} \cdot \mathbf{h}_i^A) [1 - G_1/N]^{N_1} [1 - G_2/N]^{N_2}, \\ \text{with} \quad G_1 &= \sum_R \int dh_{j\mu}^A [1 - \exp\{-ik_\mu J^{AA}(\mathbf{R})F_{j\mu,s}^A(h_{j\mu}^A)\}] \Pr_A(h_{j\mu}^A) \\ G_2 &= \sum_R \int dh_{j\mu}^B [1 - \exp\{-ik_\mu J^{AB}(\mathbf{R})F_{j\mu,s}^B(h_{j\mu}^B)\}] \Pr_B(h_{j\mu}^B). \end{aligned} \quad (10)$$

The next step involves expanding the exponentials in (10) and arguing that for most fast decaying interactions where $J(R_{ij}) = A(R_{ij})R_{ij}^{-n}$ when A is a bounded function, if $j(R_{ij}) = J(R_{ij})/A$ then the spatial moments $I_n = \sum [J(R_{ij})]^n$ rapidly decrease with n . So that in the expansion we may neglect all I_n for $n \geq 3$. Within this approximation we may carry out the integral over \mathbf{k} to obtain:

$$\Pr_A(h_\mu^A) = P_0 \exp[-\sum \sum (h_\mu^A - I_{1\mu}^A) K_{\mu\gamma}^A (h_\gamma^A - I_{1\gamma}^A)], \quad (12)$$

$$\begin{aligned} \text{with} \quad I_{1\mu}^A &= c_A I_1^{AA} m_\mu^A + c_B I_1^{AB} m_\mu^B, \quad I_{2\mu\gamma}^A = c_A I_2^{AA} q_{\mu\gamma}^A + c_B I_2^{AB} q_{\mu\gamma}^B, \\ K_{\mu\gamma}^A &= U_{\mu\eta}^T I_{2\eta\tau}^A U_{\tau\gamma} \quad \text{where} \quad U_{\mu\eta}^T q_{\eta\tau}^A U_{\tau\gamma} = q_{\mu\gamma}^A \delta_{\mu\gamma}, \\ I_1^{AA} &= \sum_R J^{AA}(\mathbf{R}), \quad I_1^{AB} = \sum_R J^{AB}(\mathbf{R}), \quad I_2^{AA} = \sum_R [J_2^{AA}(\mathbf{R})]^2, \\ I_2^{AB} &= \sum_R [J_2^{AB}(\mathbf{R})]^2. \end{aligned}$$

The probability distribution of $\Pr_B(h_\mu^B)$ is found in an exactly analogous manner. Note that in the above we have dropped the suffix i from \mathbf{h} , this is because the probability density is independent of the site because of statistical homogeneity.

In the absence of anisotropic interactions we have:

$$m_z = m, \quad m_x = m_y = 0, \quad q_{xx} = q_{yy} = \frac{1}{2}q_\perp, \quad q_{zz} = q_\parallel \quad \text{and} \quad q_{xy} = q_{yz} = q_{zx} = 0. \quad (13)$$

The distribution (12) then reduces to a simple Gaussian

$$\text{Pr}_A(h_\mu^A) = [(2\pi)^3 I_{2\perp}^A (I_{2\parallel}^A)^{1/2}]^{-1} \exp[-(h_x^{A2} + h_y^{A2})/2I_{2\perp}^A - (h_z^A - I_1^A)^2/2I_{2\parallel}^A]. \tag{14}$$

with $I_{2\parallel}^A$ and $I_{2\perp}^A$ being the same as (12) expressed in terms of q_\parallel^A and q_\perp^A . The expression for $\text{Pr}_B(h_\mu^B)$ is exactly similar with the A 's replaced by B 's. The order parameter equations are then from (4):

$$\begin{aligned} m^\alpha &= \int \int \int dh_x^\alpha dh_y^\alpha dh_z^\alpha (h_z^\alpha/h^\alpha) [L_s^{\alpha-1}(\beta h^\alpha) \delta L_s^\alpha(\beta h^\alpha)/\partial h^\alpha] \text{Pr}_\alpha(\{h^\alpha\}), \\ q_\parallel^\alpha &= \int \int \int dh_x^\alpha dh_y^\alpha dh_z^\alpha (h_z^\alpha/h^\alpha)^2 [L_s^{\alpha-1}(\beta h^\alpha) \partial L_s^\alpha(\beta h^\alpha)/\partial h^\alpha]^2 \text{Pr}_\alpha(\{h^\alpha\}), \\ q_\perp^\alpha &= \int \int \int dh_x^\alpha dh_y^\alpha dh_z^\alpha [(h_x^{\alpha2} + h_y^{\alpha2})/h^{\alpha2}] [L_s^{\alpha-1}(\beta h^\alpha) \partial L_s^\alpha(\beta h^\alpha)/\partial h^\alpha]^2 \\ &\quad \text{Pr}_\alpha(\{h^\alpha\}), \end{aligned} \tag{15}$$

where α is either A or B and $h^\alpha = ((h_x^\alpha)^2 + (h_y^\alpha)^2 + (h_z^\alpha)^2)^{1/2}$.

3. The paramagnetic boundary

In the neighbourhood of the paramagnetic to the ordered phase transition all the six m 's and q 's are small. We may expand the right side of (15) in terms of these variables and neglect the higher order terms. Using $L_s(x) = x - a_s x^3$ where $a_s = 1/3$ for the Langevin function for classical spins, we may convert the set of integral equations in (15) to a set of coupled algebraic equations in the six variables.

These equations contain as their possible solutions an inhomogeneous situation where the A and the B sublattices of atoms order at different temperatures (Mookerjee and Roy 1983b). In such a situation we may also have the two sublattices ordering differently. However, in a completely random alloy, because of statistical homogeneity we do not expect this. Experimental evidence also points to unique Curie, Néel and spin glass temperatures, where the entire system orders in a particular way. The experimentally measured magnetisation is a bulk average $m = c_A m^A + c_B m^B$ and it is this which vanishes at the Curie temperature. We must therefore seek solutions of the form

$$m^B = \eta(T, c)m^A, \quad q_\parallel^B = \mu_1(T, c)q_\parallel^A, \quad q_\perp^B = \mu_2(T, c)q_\perp^A$$

Substituting this into (15) we get six equations for the six variables $m^A, q_\parallel^A, q_\perp^A, \eta, \mu_1$ and μ_2 . A possible solution is all of these to be zero. This corresponds to a paramagnetic situation.

In the spin glass regime $m^A = m^B = 0$ and because of directional isotropy of the situation we also have $q_\parallel^A = \frac{1}{2}q_\perp^A$, and $\mu_1 = \mu_2 = \mu$. If we incorporate this into the equations obtained from (15), we may obtain another solution which is not all zero. At the spin glass-paramagnetic boundary this other solution also vanishes. This condition gives:

$$9T_\theta^2 - (T_{\theta 1}^2 + \mu^2 T_{\theta 2}^2) = 0 \quad 9\mu T_\theta^2 - (T_{\theta 3}^2 + \mu T_{\theta 4}^2) = 0,$$

where

$$\begin{aligned} k_B^2 T_{\theta 1}^2 &= c_A \sum [J^{AA}(R)]^2, & k_B^2 T_{\theta 2}^2 &= c_B \sum [J^{AB}(R)]^2, \\ k_B^2 T_{\theta 3}^2 &= c_A \sum [J^{AB}(R)]^2, & k_B^2 T_{\theta 4}^2 &= c_B \sum [J^{BB}(R)]^2 \end{aligned}$$

Eliminating μ we obtain,

$$T_\theta^2 = (1/18)[T_{\theta 1}^2 + T_{\theta 4}^2] \pm [(T_{\theta 1}^2 - T_{\theta 4}^2)^2 + 4T_{\theta 2}^2 T_{\theta 3}^2]^{1/2}. \quad (16)$$

Near the paramagnetic-random uniaxial ferromagnetic phase boundary $q_1^z = 0$. Again we may obtain this boundary by examining where the second non-zero solution of the equations obtained from (15) vanish. This yields

$$T_C = (1/6)[(T_{11} + T_{22}) \pm [(T_{11} - T_{22})^2 + 4T_{12}T_{21}]^{1/2}], \quad (17)$$

where

$$k_B T_{11} = c_A \sum J^{AA}(R), \quad k_B T_{12} = c_B \sum J^{AB}(R), \\ k_B T_{21} = c_A \sum J^{AB}(R), \quad k_B T_{22} = c_B \sum J^{BB}(R)$$

To obtain the Néel temperature we rewrite the Hamiltonian as

$$H = -\frac{1}{2} \sum \sum (-\tilde{J}_{ij}) \tilde{S}_i \cdot \tilde{S}_j$$

where \tilde{S}_i are the staggered spins and \tilde{J}_{ij} the staggered interaction. We may now carry out the entire analysis

$$T_N = (1/6)\{(\tilde{T}_{11} + \tilde{T}_{22}) \pm [(\tilde{T}_{11} - \tilde{T}_{22})^2 + 4\tilde{T}_{12}\tilde{T}_{21}]^{1/2}\}, \quad (18)$$

where the $\tilde{T}_{11}, \tilde{T}_{12}$ etc are defined exactly as in (17) with the staggered interactions replacing the $J^{aa}(R)$.

The choice of the roots in (16)–(18) is unique. For the Curie and Néel temperatures only the positive root has physical significance. For the spin glass temperature only the greater positive root is of significance. It is interesting to note that for only nearest neighbour interaction the Néel temperature is the negative solution of (17) with the sign reversed.

The phase diagram is to be understood as follows: in those concentration regimes where $T_C > \text{both } T_\theta \text{ and } T_N$ we have transition from the paramagnetic to the uniaxial random ferromagnetic phase (URF); while in those regimes where $T_N > \text{both } T_\theta \text{ and } T_C$ the transition occurs from the paramagnetic to the antiferromagnetic phase. If there exists a concentration regime where $T_\theta > \text{both } T_N \text{ and } T_C$ we have a transition from the paramagnetic to the spin glass. In no case do we find a direct transition from the paramagnetic to the mixed phase. To study the possibility of the mixed or randomly canted phase (Mookerjee 1979; Mookerjee and Roy 1983a) or the re-entrant spin glass phase (if there be any) we have to study the low temperature phases away from the paramagnetic boundary.

In case the T_C and T_N curves intersect above T_θ for some concentration range, then we are in for trouble. Because this would predict a transition from the ferromagnetic to the antiferromagnetic phase with a variation of concentration. This transition cannot be of the second order as it is always accompanied by a finite change in the free energy. First order transitions cannot be described in the manner we have proceeded. Medvedev and Zaborov (1982) seem to suggest that there is no direct transition from the ferro to the antiferromagnetic phase with variation of concentration, but rather several types of canted phases in between. However, this argument would be on a firmer basis if we come across systems which show this kind of behaviour. As of present, we are not aware of any experimental evidence of this.

Near the intersection points of T_θ with T_C and T_N , as in the case of canonical spin glasses (Mookerjee and Roy 1983) we obtain another type of solution where both q_{\parallel}^z

and q_1^α are $\neq 0$, with $q_1^\alpha + q_1^\alpha = 1$. This is the randomly canted ordered phases (as $m^\alpha \neq 0$) or the mixed phase where there is long ranged order in the z -direction as well as spin freezing in the transverse plane. This we wish to study in greater detail.

4. The $T = 0$ Phases

Near the $T = 0$ line, (15) again simplify. In this region $L_s(\beta h) = 1$ and we have in cylindrical coordinates

$$m^\alpha = (2\pi I_{21}^\alpha I_{21}^{\alpha 2})^{-1/2} \int_0^\infty \int_{-\infty}^\infty \exp[-(r^2/2I_{21}^\alpha) - (z - I_1^\alpha)^2/2I_{21}^\alpha] (zr/z^2 + r^2)^{1/2} dr dz.$$

At the spin-glass, mixed phase boundary $m^\alpha = 0$, $q_1^\alpha = \frac{1}{2}q_1^\alpha = 1/3$, $\mu_1 = \mu_2 = 1$. This yields the equation for this boundary at $c = c_0$ at $T = 0$

$$A(c_0)T_{11} + A(c_0)B(c_0)[T_{12}T_{21}/(I_{21}^B - B(c_0)T_{22})] - I_{21}^A = 0. \quad (19)$$

Other phase boundaries cannot be analytically obtained and a numerical iterative solution of (15) has to be performed.

$$A(c) = (2\pi I_{21}^A I_{21}^{A 2})^{-1/2} \iint \exp[r^2/I_{21}^A - z^2/2I_{21}^A] (z^2r/(z^2 + r^2))^{1/2} dr dz$$

$$B(c) = (2\pi I_{21}^B I_{21}^{B 2})^{-1/2} \iint \exp[r^2/I_{21}^B - z^2/2I_{21}^B] (z^2r/(z^2 + r^2))^{1/2} dr dz$$

These analytic results serve as important checks for the numerical calculations as well as starting point of the iterations.

5. Results and discussions

We display numerical results for a model case: with $I_1^{AA} = 1$, $I_1^{BB} = -1$ and $I_1^{AB} = 0.8$ (in arbitrary units with $k_B = 1$). Further we shall consider ratios $(I_2^{AA})^{1/2}/I_1^{AA} = \gamma$ between 0.45 and 0.9. We have discussed earlier (Mookerjee 1979; Mookerjee and Roy 1983b) that this ratio γ is determined both by the range and nature of the interaction and by the underlying lattice structure of the alloy.

The question may be raised: Why look at model cases with arbitrary values given to the six parameters I_1^{AA} and I_2^{AA} ? Why not determine these for a real alloy and compare them with experimental results? These parameters enter the expressions for the Curie, Néel and spin glass temperatures and provided we know these for a large number of concentrations we may determine all six parameters for a given system. In fact rough estimates of I_1^{AA} are available for alloys like FeCr. However, the published literature does not quote sufficient number of values of the transition temperatures to enable us to determine I_2^{AA} with any degree of confidence. The values chosen here are crude guesses which seem to be consistent with the scanty data we could gather.

In figure 1 we show the phase diagram for $\gamma = 0.45$. For this ratio T_g lies totally below

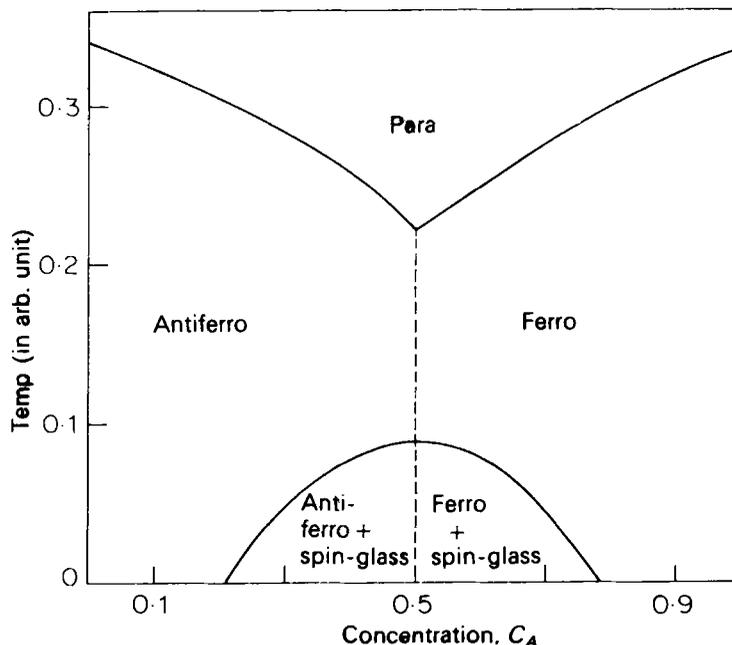


Figure 1. Phase diagram for $I_1^{AA} = 1$, $I_1^{AB} = 0.8$, $I_1^{BB} = -1$, $\gamma = 0.45$.

T_C and T_N curves, so that there is no direct paramagnetic-spin glass transition. Also at $T = 0$, there is no spin glass solution, rather a mixed phase stretches from $0.22 < c < 0.78$ giving way to uniaxial ordered phases at higher and lower concentrations. As mentioned before, the ferro to antiferromagnetic transition with concentration variation at higher temperatures is most probably a first order one. Medvedev and Zoborov (1982) suggest that in the absence of anisotropy the directions of broken symmetry in the ferromagnetic and the antiferromagnetic phases at the two concentration limits are different (in fact, perpendicular). This leads to a series of ordered canted phases (in contrast to randomly canted ones) in intermediate concentration regimes. No justification seems to be available in their paper and it is difficult to accept without further clarification, his basic assumption.

In figure 2 we show the phase diagram for $\gamma = 0.75$. Now for a concentration range there is a transition from the paramagnetic to the spin glass. At $T = 0$, the spin glass solution stretches from 0.3 to 0.7, while the rest of the concentration range is occupied by the mixed phases giving way to the uniaxial ordered phases in the concentration wings. As in canonical spin glasses (Mookerjee and Roy 1983a) careful numerical calculations indicate a precipitous fall of T_C and T_N as one approaches the spin glass region, so that there is strictly no true re-entrant spin glass phase at low temperatures. What actually seems to occur at concentrations just above the percolation concentration is a transition, at low temperatures, from the ordered to a mixed phase. Experimentally, this picture seems to be confirmed for FeCr alloys (Burke and Rainford 1983b).

To illustrate the effect of I_1^{AB} , we have shown in figure 3 a case with $I_1^{AB}/I_1^{AA} = 5$. Here both T_C and T_N initially increase on alloying and for $\gamma > 0.7$, T_θ lies above T_C and T_N for a

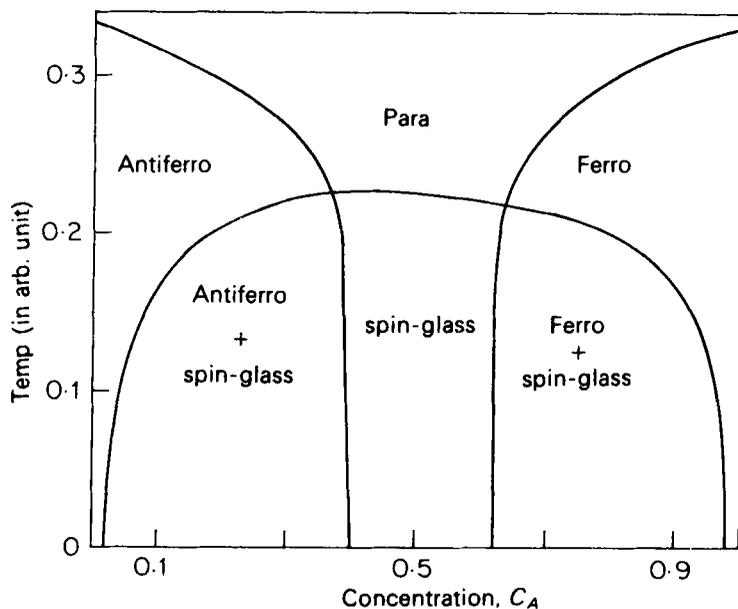


Figure 2. Phase diagram for $I_1^{AA} = 1$, $I_1^{AB} = 0.8$, $I_1^{BB} = -1$, $\gamma = 0.75$.

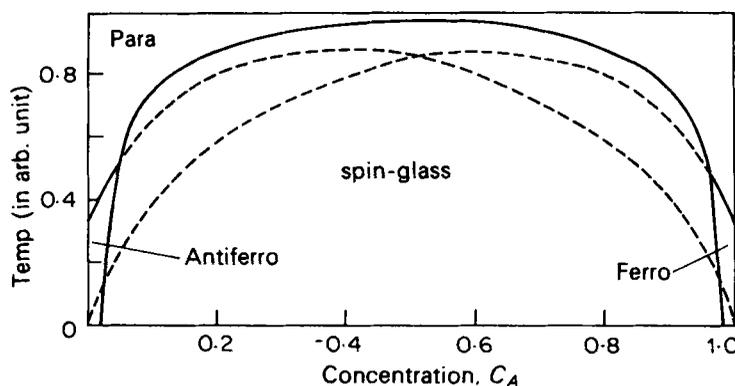


Figure 3. Phase diagram for $I_1^{AA} = 1$, $I_1^{AB} = 5$, $I_1^{BB} = -1$, $\gamma = 0.8$.

large concentration range. Otherwise qualitatively no feature is different. This is also in agreement to the picture indicated by Medvedev.

Our prediction thus, is that for the given $I_1^{aa'}$, for $\gamma > 0.7$ there is a spin glass phase, while for $\gamma < 0.7$ the low temperature phase is a mixed phase. For $\gamma > 0.7$ beyond the percolation concentrations there is always a transition from the uniaxial ordered phase to a mixed phase at low temperatures below another critical concentration. This picture is in qualitative agreement with that of Medvedev.

Before conclusion we wish to comment on several aspects of the kind of treatment we have followed. First, if we are to make quantitative calculations we have to go beyond

the simple mean field approximation and at least include the effect of the reaction field. For canonical spin glasses, this has been done within an approach very similar to ours by Klein *et al* (1979) on the Ising model. The internal local molecular field distribution in their approach cannot be analytically expressed, but careful analysis showed that the distribution is still roughly Gaussian for $c > 0.05$, but with a Lorentzian centre for $c < 0.05$. We may carry out a similar calculation for the Heisenberg model with competing interactions, but nothing from the calculations on the canonical spin glasses indicate that we will obtain any new qualitative feature. In none of these approaches do we obtain a distribution with a hole at the centre at $T = 0$. Such a 'hole' is necessary to give correct behaviour to internal energy and entropy at $T = 0$. Mookerjee and Chowdhury (1984) have recently argued that even for the replica approaches, unless we have $\chi(T) \propto A/T^2$, there should be no holes either. A hole does arise in the percolation model (Mookerjee and Chowdhury 1983) but this model does not envisage the transition as a cooperative phase transition but rather a dynamical freezing with a distribution of relaxation times. A large body of experimental evidence does in fact point to the fact that these transitions to the spin-glass or the mixed phase is, in fact, a dynamical freezing phenomenon. This next level of sophistication: *i.e.* development of the percolation model for the mixed phase is currently been looked at by these authors and a preliminary report submitted (Roy *et al* 1984).

Secondly, what about the stability of the so-called 'phases'? Here, it is important to emphasize our approach. When we minimise the free energy, it is *before* configuration averaging. The free energy is minimised as a function of an *infinite* number of variables: the 'local' magnetisations. For an ordered alloy we minimise the free energy with respect to only *one* order parameter m and in the ordered phase the free energy has *two* minima, one for $m = m_0$ and another for $m = -m_0$. In our case, in the spin glass phase the free energy has an infinite number of minima, one for each configuration of the $\{\sigma_i\}$. This picture is essential to the description of the spin glass and is lost when we configurationally average the free energy by the replica tricks and express the result as a function of only *two* order parameters m and q , as in the SK solution. It then requires the entire paraphernalia of the Parisi approach to restore this. Subsequent to the minimisation, we characterise the phases by the entire distribution of the local magnetisations (equivalently, the distribution of the local molecular fields which govern the local magnetisations). If the distribution is a δ -peak centred at zero we ascribe this to the paramagnet, while a wide distribution peaked at the centre zero is ascribed to the spin glass and that peaked at a non-zero value to the ferromagnet. The parameters $m, q_{\parallel}, q_{\perp}$ go into the description of the probability distribution. This viewpoint was introduced prior to the Edwards-Anderson approaches by Klein (1968).

Finally, a comment regarding the predictions of the Sherrington-Kirkpatrick-Parisi approaches. Gabay and Toulouse (1981) predicted two low temperature phase transitions in the concentration regime just beyond the percolation concentration. One is analogous to our mixed phase, accompanied by weak irreversibility, anomalies in magnetisation, susceptibility, neutron scattering intensity, etc. While at low temperatures they predict a second transition. This is intimately related to the kind of replica symmetry breaking. Campbell *et al* (1983) interpret their observation of strong irreversibility at temperatures much lower than the m and χ anomalies in the light of this prediction. Our approach, however, does not indicate any such second transition. However, the onset of strong irreversibility may be explained without taking recourse to any physically obscure replica symmetry ideas. In any random system, at any finite

temperature, apart from an infinite cluster of ferromagnetic spins (below T_C), there always exist finite clusters whose boundaries are closed sets of frustrated bonds which effectively magnetically isolate this cluster from the rest of the system. If the activation barriers, because of intraccluster exchange, intercluster dipolar couplings, anisotropies, etc seen by these finite clusters are much larger than those hindering formation of Bloch walls within the infinite ferromagnetic cluster, at low enough temperatures these clusters will appear frozen and consequently give rise to strong hysteresis effects.

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