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LETTER TO THE EDITOR

Frustrated antiferromagnets in an external magnetic field

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Abstract. Magnetic properties of disordered Ising antiferromagnets with frustrated intrasublattice exchange interactions are studied. The theory explains satisfactorily some experimental results.

Some strongly disordered Ising antiferromagnets (AF) are currently under experimental investigation [1–4]. In [5–8], a mean-field-type theory of such magnets has been developed and AF in which the frustration of intersublattice exchange interactions exceeds that of the intrasublattice ones have been investigated in detail. It has been shown that the temperature $T_g(H)$ of the phase transition from the ergodic AF state to the non-ergodic one should increase with the magnetic field H up to a field H_0 where the line $T_g(H)$ crosses the Néel temperature line $T_N(H)$. At $H > H_0$, the temperature $T_g(H)$ falls with increasing H. A twofold increase of $T_g(H)$ has been observed in the Ising metamagnet $Fe_xMg_{1-x}Cl_2$ [1].

Recently, another frustrated AF, $Fe_xMn_{1-x}TiO_3$, has been studied [3, 4]. Some experimental results obtained for alloys with $x > 0.57$ differ from both the earlier observations for $Fe_xMg_{1-x}Cl_2$ and from theoretical predictions [5–8].

(i) The curve representing $T_g(H)$ has a sharp maximum at some field H_m lower than the crossing field H_0. At H_m, $T_g(H)$ touches the Néel temperature curve $T_N(H)$.

(ii) The measured ratio $T_g(H_m)/T_g(0) \simeq 6$ and is three times greater than the theoretical value.

(iii) The magnetic susceptibility has a minimum in the ergodic state. The greater the degree of frustration the more pronounced the minimum.

The aim of this letter is to show that all these features can be explained if it is taken into account that in $Fe_xMn_{1-x}TiO_3$ only the intrasublattice exchange interaction is frustrated. The reason that the frustration of the intersublattice interaction is small or zero is as follows. Both $FeTiO_3$ and $MnTiO_3$ are layered AF with antiferromagnetic interlayer interactions. The intralayer interaction is ferromagnetic in $FeTiO_3$ and antiferromagnetic in $MnTiO_3$. So it is natural to suppose that during alloying of $FeTiO_3$ with Mn the sign of the intersublattice interaction does not fluctuate and only the intrasublattice interaction is frustrated.

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We consider the system of spins distributed randomly over two sublattices and interacting according to the Ising Hamiltonian

$$\mathcal{H} = \sum_{i,j} J_{ij} S_{1i} S_{2j} - \sum_{\substack{p=1,2\\ i,j}} V_{ij} S_{pi} S_{pj} - H \sum_{\substack{p=1,2\\ i}} S_{pi}. \tag{1}$$

$p = 1, 2$ is the sublattice number, $S_{pi} = \pm 1$. As is usual in the infinite-ranged model [9], we suppose that the interaction energies J_{ij} and V_{ij} do not depend on the distance r_{ij} and are distributed normally with the mean values J_0/N, V_0/N and variances J^2/N, V^2/N, where N is the total number of spins in a sublattice.

The infinite-ranged model is generally accepted to describe the static properties of real spin glasses and re-entrant magnets. The reason for this is that correlations due to spatial dispersion have very slow time decay, and so the spin glass order parameter differs from zero for actually attainable times at least for Ising systems [10, 11]. We show in this letter that magnetic properties of the materials under discussion are qualitatively explained in the framework of the infinite-ranged model as well.

The equations of state for this model, which determine the sublattice magnetisations $m_{1,2}$, the Parisi functions $q_{1,2}(x)$ [12], the free energy etc, were obtained earlier [6, 7] using the replica trick and the Paris *ansatz*. In particular, the expression for the transition temperature T_g from the ergodic state to the broken replica symmetry (non-ergodic) phase is

$$(T_g^2 - V^2\langle\cosh^{-4} E_1\rangle)(T_g^2 - V^2\langle\cosh^{-4} E_2\rangle) - J^4\langle\cosh^{-4} E_1\rangle\langle\cosh^{-4} E_2\rangle = 0 \tag{2}$$

$$E_p = (1/T)[H_0 + V_0 m_p - J_0 m_{\bar{p}} + z(V^2 q_p + J^2 q_{\bar{p}})^{1/2}] \qquad p \neq \bar{p}$$

$$\langle u(z)\rangle = \int \frac{\mathrm{d}z}{\sqrt{(2\pi)}} \mathrm{e}^{-z^2/2} u(z) \tag{3}$$

where m_p and q_p should be obtained from the Sherrington–Kirkpatrick-type equations

$$m_p = \langle\tanh E_p\rangle \qquad q_p = \langle\tanh^2 E_p\rangle. \tag{4}$$

In the $J/V \to 0$ limit, equation (2) splits into two independent equations $T_g^2 = V^2\langle\cosh^{-4} E_p\rangle$. This means that the non-ergodicity appears in the sublattices independently. Without the external field $m_1 = -m_2$, $q_1 = q_2$, and so $T_{g1} = T_{g2}$. If the field is applied, $T_{g1} \neq T_{g2}$. It is clear that the irreversibility and other spin-glass-type effects appear at the higher of these temperatures, so it is the magnetic field dependence that should be compared with experiments.

For the sublattice with spins down at $H \to 0$ (sublattice one) the absolute value of the magnetisation $|m_1|$ is smaller than m_2 at all H, T values in the AF region of the phase diagram. This means that the effective field on spins of the first sublattice is smaller than that on spins of the second one, and so $T_{g1}(H) > T_{g2}(H)$. At the onset of the paramagnetic phase the temperatures $T_{gp}(H)$ coincide.

Similar arguments can be used to explain qualitatively the magnetic field dependences of T_{gp}. If we suppose that the metamagnetic transition point is not in the phase diagram region we consider now (the influence of the metamagnetic phase transition on the spin glass properties and vice versa have been considered in [7, 8]), $|m_1|$ decreases monotonically when H increases and goes to zero at some field $H = H_m$. Hence it is clear that $T_{g1}(H)$ increases with H for $H < H_m$. For $H > H_m$ the magnetisation m_1 is positive and increases with H, so $T_{g1}(H)$ falls.

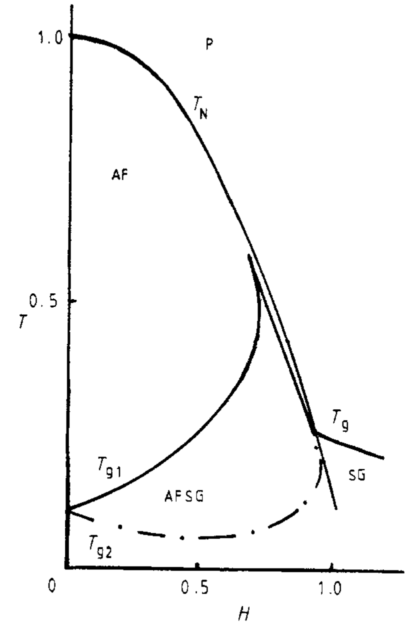

Figure 1. The phase H–T diagram of a frustrated antiferromagnet. The parameters are: $V_0 = J = 0$; $J_0 = 1$; $J_0/V = 1.7$. The phases are: P—paramagnetic; AF—antiferromagnetic ergodic; AFSG—antiferromagnetic non-ergodic; SG—non-ergodic phase without antiferromagnetic order. The chain curve denotes the temperature at which the second-sublattice non-ergodicity appears.

On the other hand, at the second sublattice $T_{g2}(H)$ decreases with increasing H for small H, and increases at larger fields $H > H_m$ up to a value of H when the antiferromagnetic ordering is destroyed.

Thus, the curve $T_{g1}(H)$ has a maximum in the antiferromagnetic phase on the $m_1(H, T) = 0$ line. The value $T_{g1}(H_m) = T_{gm}$ can be determined from

$$q_1 = \langle \tanh^2(V_z/T_{gm})\sqrt{q_1}\rangle \qquad T_{gm}^2 = V^2\langle \cosh^{-4}(V_z/T_{gm})\sqrt{q_1}\rangle \tag{5}$$

which has only one solution $q_1 = 0$, $T_{gm} = V$. This means that the transition considered is equivalent to the transition from the 'paramagnetic' state of the first sublattice ($m_1 = q_1 = 0$) to the 'pure' spin glass ($m_1 = 0$, $q_1 \neq 0$). So $T_{g1}(H)$ should have a cusp at $H = H_m$.

The H–T phase diagram at $J = 0$, $V_0 = 0$ and $T_N(0)/T_g(0) = 9.5$ obtained by numerical solution of (2) and (4) is given in figure 1. We should note that in all samples studied in [3, 4], the long-range magnetic ordering disappeared before the second-order phase transition was reached, which means that either V_0 is small or the first-order transition is depressed by the disorder [7, 8]. This is why it is the results for $V_0 = 0$ that are compared with experiment.

Comparison of figure 1 with figure 3 of reference [4] shows us that the general behaviour of the theoretical and experimental T_g-curves is the same: for $H < H_m$ as well as for $H > H_m$ the T_g-curve is within the antiferromagnetic phase. In contrast to what is found by experiment, $T_{g1}(H_m) \neq T_N(H_m)$ in the theoretical phase diagram, but the difference between these two values is very small, so it probably cannot be detected without special measurement techniques.

If $J = 0$, the increase of T_g in the magnetic field is much larger than in the case where $V = 0$ studied by us previously [6]: at $T_N(0)/T_g(0) = 9.5$, T_g has an approximately twofold increase at $V = 0$ and a sixfold increase if $J = 0$ (figure 1). The latter number does not differ essentially from the measured value.

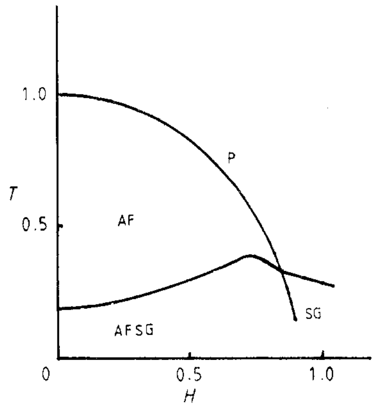

Figure 2. The H–T phase diagram of a frustrated antiferromagnet for $V_0 = 0$; $J_0 = 1$; $V^2/J^2 = 5$; $J_0/(V^2 + J^2)^{1/2} = 1.5$.

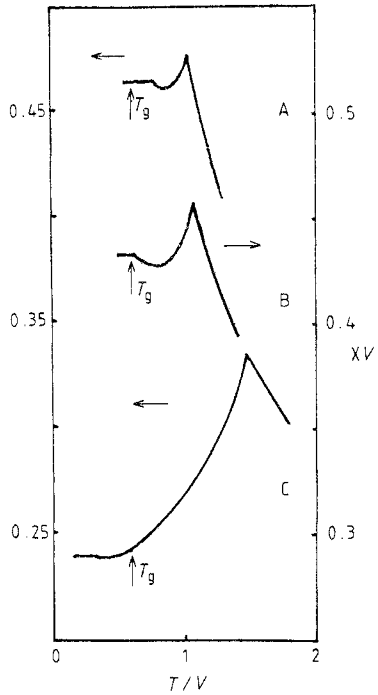

Figure 3. The temperature dependence of the magnetic susceptibility for several degrees of frustration $x = J_0/V$ ($V_0 = J = 0$): A: $x = 1.05$ ($T_N(0)/T_g(0) = 1.37$); B: $x = 1.1$ ($T_N(0)/T_g(0) = 1.66$); C: $x = 1.5$ ($T_N(0)/T_g(0) = 5.46$).

We would like to note also that at small fields $T_g(H) - T_g(0) \sim H^\alpha$, $\alpha = 2$ at $J \neq 0$ and $\alpha = 1$ at $J = 0$. This enables us to understand why the α-value measured for $Fe_xMn_{1-x}TiO_3$, $x > 0.57$ is much smaller than 2—which was unclear before and was considered in [13] to show a disagreement between the theory and experiment.

At non-zero (but small) J the maximum in the $T_{g1}(H)$ curve broadens and moves from the $T_N(H)$ curve into the body of the antiferromagnetic phase (figure 2). Strictly speaking, the second solution $T_{g2}(H)$ of (7) does not make sense at $J \neq 0$ as the non-ergodicity in this case appears in both sublattices simultaneously. But if $J \ll V$, the non-ergodicity in the second sublattice in the temperature interval $T_{g1}(H) > T > T_{g2}(H)$ is small:

$$\mathrm{d}q_2(x)/\mathrm{d}x \simeq (J^2/V^2)(\mathrm{d}q_1(x)/\mathrm{d}x).$$

The equations of state obtained earlier [7] enabled us to obtain a general expression for the magnetic susceptibility χ, valid for any H, T values. For the ergodic state, the formula for χ can be derived by differentiating (2) for m_p and q_p with respect to H at $H \to 0$ and has a rather simple form:

$$\chi = \pi/[1 + (J_0 - V_0)\pi]$$
$$\pi = \frac{1}{T}\left[g_1 + \frac{J^2 - V^2}{2T^2} g_2^2 \left(1 - \frac{J^2 - V^2}{2T^2} g_3\right)^{-1}\right]$$
$$g_k = \langle \mathrm{d}^k \tanh E_p / \mathrm{d}E_p^k \rangle|_{H \to 0}. \tag{6}$$

For $V_0 = V = 0$ this expression is equivalent to that given in [6].

We see that the V^2- and J^2-terms in (6) have opposite signs, i.e. the frustrations of the intersublattice and intrasublattice exchange interactions influence χ in different ways. This can be easily understood by considering the effect of the weak external magnetic field H on the variance δH_p of the effective field acting on spins of a subsystem p. It follows from (3) that δH_p is equal to

$$\delta H_p = (V^2 q_p + J^2 q_{\bar{p}})^{1/2} \qquad p \neq \bar{p}. \tag{7}$$

A weak magnetic field decreases $|m_1|$ and q_1 (note that the subscript one applies to the sublattice with spins directed down at $H = 0$) and increases m_2 and q_2, so the magnetic field contributions to $V^2 q_p$ and $J^2 q_{\bar{p}}$ have different signs.

Analytically, the $\chi(T)$ dependence in the ergodic state can be studied for strongly frustrated AF, when $b = (J_0 + V_0)/(J^2 + V^2)^{1/2} - 1 \ll 1$. In this case $T_g(0)/T_N(0) = 1 - b$, and the perturbation theory on b can be derived for all temperatures above T_g [5]. According to [5], it appears that at $V = 0$, $J \neq 0$, the susceptibility increases monotonically with the decrease of T in the ergodic phase. If $J = 0$, $V \neq 0$, the situation is quite different. In this case

$$\chi = (1/2J_0)[1 - (T_N/J_0)\tau(b - \tfrac{1}{3}\tau^2)/(b + \tau)]. \tag{8}$$

Here $\tau = 1 - T/T_N \ll 1$. It follows from (8) that χ changes non-monotonically with T for $T_g < T < T_N$ and has a minimum at $\tau = (\frac{3}{2}b^2)^{1/3} < b$. It is seen from figure 3 that for $J = 0$ such a dependence on $\chi(T)$ is found for any degree of frustration. This conclusion agrees with the experimental observations [3, 4]. Moreover, comparing figure 3 with figure 1 [4] we see that in both the theory and experiment the minimum in the $\chi(T)$ curves nearly disappears in the weakly frustrated magnets.

To study $\chi(T)$ below the T_g-point the general expressions of state derived in [7] should be solved, which is possible only near T_g, when $\tau_g = (T_g - T)/T_g \ll 1$. Using the procedure proposed in [6] we calculate the derivatives $D_\pm = \mathrm{d}\ln\chi/\mathrm{d}\ln T|_{T=T_g\pm 0}$. It appears that $D_- > 0$, $D_+ < 0$ at some values of J/V, which means that $\chi(T)$ has a cusp at $T = T_g$. There is a disagreement between theory and experiment at this point: according to the theory $\chi(T)$ changes very slightly below T_g and has a cusp at $T = T_g$, while experimentally the field-cooled susceptibility does not reveal an anomaly at $T = T_g$ and becomes temperature independent only considerably below T_g.

We would like to note in conclusion that our theory can explain the existence of a wide smooth maximum in the $T_g(H)$ curve observed for $Fe_{0.552}Mg_{0.448}Cl_2$ [1]. The reason for its existence is that when an $FeCl_2$ crystal with strong ferromagnetic in-plane interaction and weak antiferromagnetic inter-plane interaction is diluted by Mg atoms, the 're-entrant' phase transition is exhibited when the spin concentration is near to the in-plane percolation threshold. In this case the next-nearest-neighbour in-plane exchange interaction becomes important, but essentially does not exceed the inter-plane interaction. So, it is natural to suppose that for the magnets we discussed just now $J \leqslant V$. It can be seen from figure 2 that the theory predicts the existence of a smooth maximum in the T_g-curve for such a region of parameters.

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