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Random Fields and Ordering in Antiferromagnetic Insulators

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Abstract

Neutron scattering measurements of the effect on the long range order and phase transitions of random magnetic fields are reviewed. The results at low temperatures in $\text{Rb}_2\text{Co}_x\text{Zn}_{1-x}\text{F}_4$ and $\text{Co}_x\text{Zn}_{1-x}\text{F}_2$ show that the long range order is not established when samples are cooled in a magnetic field, but they form a state with unusually long magnetic correlations. In $\text{Mn}_x\text{Zn}_{1-x}\text{F}_2$ a similar state is formed but only very close in temperature to T_N .

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Short title: Random Fields and Ordering

This paper is a brief qualitative report on neutron scattering measurements performed on randomly disordered antiferromagnets in an applied magnetic field. The applied uniform field produces a random staggered field and so the experiments provide information about the effect of a random staggered magnetic field on the phase transition and ordering of these antiferromagnets. Experiments have been performed on several systems; in $\text{Co}_x\text{Zn}_{1-x}\text{F}_2$, $\text{Mn}_x\text{Zn}_{1-x}\text{F}_2$ and $\text{KMn}_x\text{Ni}_{1-x}\text{F}_3$ the spins are coupled in a three dimensional arrangement, and the magnetic interactions are very Ising-like in $\text{Co}_x\text{Zn}_{1-x}\text{F}_2$ and Heisenberg-like in $\text{KMn}_x\text{Ni}_{1-x}\text{F}_3$, whereas in $\text{Rb}_2\text{Co}_x\text{Mg}_{1-x}\text{F}_4$ the spins interact with largely Ising-like interactions and are coupled in two-dimensional sheets. The experiments have been performed in collaboration with R. J. Birgeneau, W. J. L. Buyers, H. J. Guggenheim, M. Haken, H. Ikeda, S. Satija, G. Shirane and H. Yoshizawa and a preliminary account [1] has already been published.

It has been well known since the work of Imry and Ma [2] that site-random magnetic fields have drastic consequences on the phase transitions and ordering of magnetic systems. More specially a random field in systems with continuous symmetry is expected to destroy the long range order in all dimensions d less than 4, but in Ising systems the behavior is less certain. Imry and Ma showed that smooth domain walls will break up the long range order for dimensions d less than 2. There are, however, arguments based on expansions in $6-\epsilon$ dimensions [3,4] that the behavior of systems in a random field is

analogous to that of pure systems in $d=2$ dimensions, suggesting that in Ising systems the lower critical dimension is 3 rather than 2. More recently this suggestion gained further support from the work of Pytte et al. [5] who suggested that for dimensions $d < 3$ the long range order is unstable against the formation of rough domain walls and a similar conclusion by Kogon and Wallace [6] using supersymmetry arguments. The models used by these authors have, however, recently been criticized by Grinstein and Ma [7] who argue that the random field does indeed destroy the long range order only for dimensions $d < 2$.

In view of this controversy it is clearly essential to perform experiments. Fortunately, Fishman and Aharony [8] have pointed out that a random staggered field is produced when a uniform magnetic field is applied to a random antiferromagnet. This allows experiments to be performed in which the strength of the random staggered field can be altered by changing the magnitude of the applied uniform field.

The experiments on the two-dimensional system $\text{Rb}_2\text{Co}_{0.7}\text{Mg}_{0.3}\text{F}_4$ were performed at the Brookhaven National Laboratory High Flux Beam Reactor with a two-axis neutron scattering spectrometer. The results show that the width of the magnetic scattering increases with increasing magnetic field even though the width of the nuclear Bragg scattering is unchanged. This shows that the magnetic field destroys the long range magnetic order showing that the lower critical dimension for the random field problem is less than or equal to 2. In more detail the line shape is very unusual compared with that observed in more critical scattering problems. It has more extended wings than would be expected if the system broke up into

Gaussian domains, while on the other hand the wings are less extended than those given by Lorentzian critical scattering. Recent theories [5,6] have suggested that the line shape be of the form:

$$S(\underline{Q}) = \frac{A}{(\underline{K}^2 + \underline{q}^2)} + \frac{B}{(\underline{K}^2 + \underline{q}^2)^2}, \quad (1)$$

where $\underline{Q}$ the momentum transfer = $\underline{q} - \underline{G}$ and $\underline{G}$ is a reciprocal lattice vector. We have fitted this form to the experimental results and it does indeed give an excellent account of the experimental results. At high temperatures above T_N the Lorentzian squared term is negligible but it increases in importance as the temperature is reduced. At low temperatures we find that the correlation length, K , is proportional to the applied field, H , to the power 1.6 whereas the amplitude B is approximately proportional to K^2 . These results are broadly consistent with the behavior expected theoretically although there is still no detailed theory of the temperature dependence or of the magnitude of K , A , and B .

Similar experiments at the Brookhaven HFBR have been conducted on two $\text{Co}_x\text{Zn}_{1-x}\text{F}_2$ samples with $x=0.35$ and 0.26 and $T_N=13.25\text{K}$ and 6.7K respectively. At high temperatures above T_N the scattering was consistent with a Lorentzian line shape as it was for the more dilute sample for magnetic fields above 11.0 kG and temperatures down to 1.8 K . At high temperatures or high magnetic fields, the scattering is characteristic of a normal paramagnetic phase. At low temperatures and fields the scattering is unusual in several different aspects.

Firstly, the results are dependent on the immediate prior history of the sample: different results are obtained when the sample is cooled from above T_N to those obtained when the field is altered below T_N . Consistent results are obtained if the field is always altered when the sample is above T_N and this technique was used for the results described below.

As was found in $\text{Rb}_2\text{Co}_{0.7}\text{Mg}_{0.3}\text{F}_4$, the width of the scattered lineshape in the presence of an applied field was larger than consistent with long range magnetic order. This width steadily increased with increasing magnetic field showing that the long range order is destroyed by the random fields. The line shape was more sharply peaked than was consistent with a Lorentzian line shape and had larger tails than a Gaussian profile. It was consistent with the form given in Eqn. 1, namely a Lorentzian and Lorentzian squared, but could also be accounted for by a power law form:

$$\tilde{S}(Q) = \frac{A}{|q|^{2-\eta}} \quad (2)$$

with $-1 < \eta < 0$, but only the former results will be discussed. At low temperatures the inverse correlation length, K , is roughly proportional to the applied magnetic field H to the power 3.5 whereas the amplitude, B , is roughly proportional to $H^{3.1}$. On increasing the temperature K initially decreases slightly and then increases rapidly for temperatures greater than a critical temperature, $T_N(H)$, which decreases as H increases.

The experiments on $\text{Mn}_{0.78}\text{Zn}_{0.22}\text{F}_2$ were performed at the Chalk River Nuclear Laboratories and yielded results which are somewhat different from those on the $\text{Co}_x\text{Zn}_{1-x}\text{F}_2$ system. In this system it was found that a field of 40 kG produced no increase in the line width of the magnetic Bragg reflection at low temperatures. Consequently in this case the long range order is not destroyed at low temperatures on a scale of the experimental resolution; 1200 lattice spacings. The critical fluctuations are, however, different in the presence of the field from those measured without an applied field as also found by Belanger et al. [9]. In the absence of the magnetic field the scattering from the critical fluctuations could always be well described by a Lorentzian line shape with an inverse correlation length, K , which decreases as the temperature approached $T_N=48.7$ K.

In the presence of a field the scattering was very different especially for $T_N - T < 2$ K. The scattering was more divergent than that given by a Lorentzian form and furthermore was dependent on the sample history. This is clearly a very similar behavior to that obtained for the $\text{Co}_x\text{Zn}_{1-x}\text{F}_2$ sample but in this case is restricted to temperatures very close to T_N .

The results for the $\text{Co}_x\text{Zn}_{1-x}\text{F}_2$ and $\text{Mn}_x\text{Zn}_{1-x}\text{F}_2$ systems are of particular interest in view of the current theoretical controversy concerning the effect of random fields on the long range order in $d=3$ Ising systems. Unfortunately the situation is not completely clear experimentally. At first glance the results for the $\text{Co}_x\text{Zn}_{1-x}\text{F}_2$ system suggest that long range order is destroyed by the random field and that the lower critical dimension is at least 3. The observed long range order at low temperatures in

$\text{Mn}_{0.78}\text{Zn}_{0.22}\text{F}_2$ must then be explained as arising because the concentration $(1-x)$ is relatively small, and T_N is larger than for the Co samples. A larger magnetic field is then needed to produce a detectable effect on the scattering at low temperatures.

An alternative explanation is that the random field produces a large effect and anomalously shaped scattering only for temperatures relatively close to T_N , and that the true equilibrium state of the system at low temperatures is one of long range order, as found in $\text{Mn}_{0.78}\text{Zn}_{0.22}\text{F}_2$. In this case the $\text{Co}_x\text{Zn}_{1-x}\text{F}_2$ results can be understood if the "low temperature" results arise from "frozen-in" configurations of the state obtained close to T_N . The spin wave gap in the $\text{Co}_x\text{Zn}_{1-x}\text{F}_2$ system is much larger than $k_B T_N$, whereas in the $\text{Mn}_x\text{Zn}_{1-x}\text{F}_2$ system it is much smaller. Consequently the establishment of the equilibrium low temperature state may be more difficult in the Co salts than in the Mn salts. In this situation the lower critical dimension is less than 3, but the theory as yet fails to explain why the random field produces such a large and unusual effect on the critical fluctuations, or why there are such slow relaxation times.

The final set of experiments were performed on the $\text{KMn}_x\text{Ni}_{1-x}\text{F}_3$ system, at AERE, Harwell. In this system the magnetic interactions are isotropic and the spin wave energy gap is very small. On applying the magnetic field the domains in which the spin direction was aligned along the applied field underwent a spin-flop transition so that all the spins were aligned perpendicular to the magnetic field. In this case the field produces only a random transverse field which is not expected to destroy the long range order

as indeed was found experimentally. A uniform field in this case breaks the symmetry of the system and produces a spin-flop transition rather than a random field state. It is not possible using a uniform field to produce a random field with completely random directions as is required to test Imry and Ma's predictions for a Heisenberg system. These results are in agreement with the theoretical work of Aharony [10].

In conclusion the effect of random fields on the ordering and phase transitions of systems still presents a number of theoretical and experimental questions. Experimentally the results for two-dimensional Ising systems are qualitatively in accord with the theory which suggests that the long range order is destroyed by the random fields. The situation for three-dimensional systems with Ising interactions is less certain but in practice it is certain that an unusual state without long range order is obtained by cooling the $\text{Co}_x\text{Zn}_{1-x}\text{F}_2$ samples in a magnetic field. Whether this state is the true low temperature state or a metastable state only further experimental work can elucidate.

This paper reports only briefly and qualitatively on a substantial body of work to which many have contributed. Much of the work is currently being written up in detail and will be published in full in due course. I am grateful to my many collaborators for their help with this interesting project. Financial support was provided by the Division of Materials Sciences, U. S. Department of Energy under contract DE-AC02-76CH00016, by Atomic Energy of Canada Limited and by the Science and Engineering Research Council.

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