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THE PHASE DIAGRAM OF $\text{Rb}_2\text{Mn}_{0.7}\text{Mg}_{0.3}\text{F}_4$ IN A MAGNETIC FIELD

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Abstract. – The phase diagram of the quasi-two-dimensional disordered antiferromagnet, $\text{Rb}_2\text{Mn}_{0.7}\text{Mg}_{0.3}\text{F}_4$ in a uniform magnetic field along the unique c -axis has been measured using neutron scattering techniques. There are three distinct phases; at high temperatures the material is paramagnetic, and on cooling in fields above 1.7 T, it undergoes a transition to a spin-flopped phase with long range order perpendicular to the c -axis. The transition temperature decreases with decreasing magnetic field and is below 2 K at 1.75 T. At lower fields there is a phase in which different domain sizes are metastable and the metastability boundary decreases in temperature with increasing field and appears to meet the phase boundary for the spin-flopped phase at $T = 0$.

Rb_2MnF_4 is a quasi-two-dimensional antiferromagnet [1] in which the Mn ions interact with strong antiferromagnetic interactions to produce two-dimensionally ordered sheets perpendicular to the unique c -axis. There are much weaker exchange interactions between the sheets and in the antiferromagnetic state the effective interactions between the sheets largely cancel. The weak dipolar interactions between the spins cause them to align along the c -axis, in a three dimensional antiferromagnetic structure below 44 K. The introduction of substitutional Mg produces non-magnetic holes in the magnetic lattice and the magnetic ordering temperature is reduced.

The phase diagrams of both pure Rb_2MnF_4 and disordered $\text{Rb}_2\text{Mn}_{0.7}\text{Mg}_{0.3}\text{F}_4$ in the presence of a magnetic field aligned along the c -axis have been measured using neutron scattering techniques. The single crystals were aligned with the c -axis vertical and mounted in a vertical field, variable temperature, superconducting magnet. The neutron scattering experiments were performed with a triple crystal spectrometer at the Brookhaven HFBR. Pyrolytic graphite was used for the monochromator and analyser and the horizontal collimation from reactor to detector was $20'$, $10'$, $10'$, and $20'$. The incident neutron energy was 14 meV and a pyrolytic graphite filter was used to suppress neutrons reflected from higher order planes. The spectrometer was mostly used with the analyser set to detect elastically scattered neutrons.

Initially the phase diagram of pure Rb_2MnF_4 was measured by observing the scattering at a large number of points at different fields, H , and temperatures, T . The phase boundaries were determined by plotting either the field or temperature dependence of either the Bragg scattering observed for a wavevector transfer $Q = (1, 0, 0)$ or the critical scattering with

$Q = (1, 0.005, 0)$. The results are shown in figure 1, and show a bicritical point similar to that observed in the three dimensional antiferromagnet, MnF_2 [2]. This is in contradiction to earlier neutron scattering measurements on K_2MnF_4 [3], which suggested a different shape for the phase diagram. We do not understand the origin of this discrepancy.

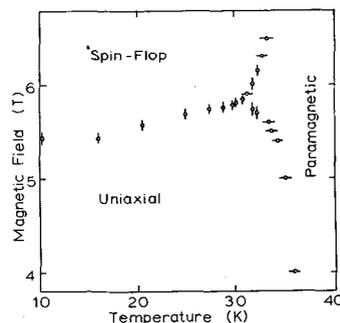


Fig. 1. – Phase Diagram of Rb_2MnF_4 .

The phase diagram for the disordered material is shown in figure 2. The boundary between the spin-flop phase and the paramagnetic phase was located by determining the temperature for the onset of long range order. Particularly at the smaller fields, the resolution limited peak from the long range order, was superposed on very intense diffuse scattering which was described by a Lorentzian squared form. The scattering observed for magnetic fields larger than 1.7 T was ergodic and dependent only on the field and temperature and not on the prior history of the sample.

At lower magnetic fields the behaviour was more complex and the results at low temperatures dependent on the prior history. When the material was

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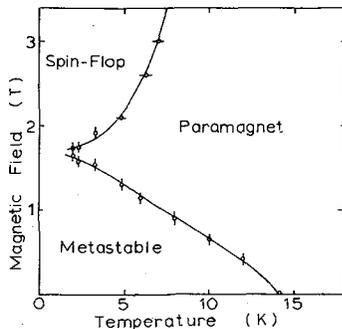


Fig. 2. - Phase Diagram of $\text{Rb}_2\text{Mn}_{0.7}\text{Mg}_{0.3}\text{F}_4$.

cooled in the field long range order was not obtained and the width of the scattering increased with increasing applied field. In contrast when the sample was cooled in zero field and the field increased some long range order was retained until the field or temperature was raised above a well defined boundary. Above this boundary the behaviour was ergodic but below the boundary a range of metastable states were all found to be stable and exhibited negligible time dependence over a 9 hour period. This behaviour is similar to that observed in the Ising-like, two dimensional antiferromagnet, $\text{Rb}_2\text{Co}_{0.7}\text{Mg}_{0.3}\text{F}_4$ [4], and also to that observed in three dimensional systems [5]. As shown in figure 2, the metastability boundary and the spin-flop boundary seem to be approaching the same field as the temperature approaches zero.

There is unfortunately not a detailed theory of the behaviour described by these results. Firstly the spin-flop phase of pure Rb_2MnF_4 is a nearly two-dimensional XY phase, and so might have been expected to show the power law behaviour characteristic of a Kosterlitz-Thouless phase rather than long range

order. Presumably the weak in-plane anisotropy, dipolar and three dimensional interactions cause a crossover to conventional long range order. In the disordered material the metastable phase is characteristic of an Ising model in a random field. As the strength of the field increases both the Ising anisotropy decreases and the random field increases. Both effects lead to a decrease in the tendency to order. When the field is sufficiently large that the anisotropy is effectively zero, there are very large fluctuations which may well destroy both the spin-flop ordering and the metastable phase. Possibly this is why the bicritical point, if it exists, is possibly at $T = 0$.

A more detailed account of these experiments including a detailed description of the form of the scattering will be published elsewhere.

Acknowledgments

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