Magnetic neutron diffraction of MnO thin films

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We report on magnetic neutron diffraction carried out on various epitaxial MnO (111) thin films grown on sapphire and MgO substrates. In all samples, of masses between 5 and 50 µg, magnetic Bragg peaks have been observed. The films exhibit what appears to be continuous phase-transitions in contrast to the strongly discontinuous transition exhibited by bulk samples [1]. In addition, the Néel temperature of films prepared on sapphire substrates is strongly enhanced above that of the bulk whilst that of the film grown on MgO is depressed. The possibility to measure magnetic excitations in such thin film systems is discussed in the light of promising test results obtained from an inelastic magnetic neutron scattering experiment on the IN8 spectrometer.

Transition metal mono-oxides are well known magnetic materials. The determination of their bulk magnetic structures was amongst the first experiments to demonstrate the power of magnetic neutron diffraction [1]. Bulk MnO exhibits a first-order phase-transition to an antiferromagnetic phase at a Néel temperature of 118 K. The magnetic phase-transition is accompanied by a lowering of the point group symmetry from cubic to trigonal on account of the type 2 antiferromagnetic order. The discontinuous character of the transition has been discussed within the framework of renormalisation group analysis [2]. This same analysis predicts a crossover to a continuous, second order, phase-transition on reducing the dimensionality of the order parameter. In bulk MnO the dimension of the order parameter is 8, due to the fourfold symmetry of the propagation vector. By preparing thin films one magnetic propagation vector could be favored, bringing the dimensionality of the order parameter down to 2.

Thin, epitaxial films of MnO have been successfully grown using a pulsed laser deposition technique [3]. The films used in these experiments were grown on two different substrates: sapphire, Al_2O_3, (0001) and MgO (111). The MnO films grow along the (111) direction, and were prepared with thickness varying from 200 Å up to 5000 Å. Non-resonant magnetic x-ray scattering, performed at the magnetic scattering beamline (ID20) of the European Synchrotron Radiation Facility, was able to establish that the films order antiferromagnetically with a similar magnetic structure to that observed in bulk MnO.

A first aim of our magnetic neutron scattering study, reported here, was to see whether comparable magnetic diffraction peaks from samples with a total mass of as small as 5µg - 50µg could be observed using neutrons. On the IN20 spectrometer, using the three-axis mode to keep the background low, we were able to measure magnetic peaks from three films with thickness of 530 Å (sapphire substrate), 2500 Å (MgO substrate) and 3500 Å (sapphire substrate). Count rates varied from 20 cts/sec to 120 cts/sec with increasing thickness. This surprisingly high count-rate allowed a systematic study of the thermal evolution of the antiferromagnetic order parameter in all three films.

Figure 1 shows the temperature dependence for a 3500 Å MnO thin film grown on sapphire. A continuous phase-transition with a Néel temperature of 149 K, which is much higher than the 118 K in bulk MnO, has been observed. The 530 Å film shows the same continuous phase-transition with a Néel temperature also close to 149K. This suggests that, for a given substrate and preparation conditions, the change in Néel temperature may be independent of film thickness over this range. On changing the substrate to MgO, Fig. 2, with a film thickness of 2500 Å (i.e. intermediate between the 530 Å and 3500 Å films), the Néel temperature *falls* below the bulk value to around 100 K whilst the transition remains continuous.

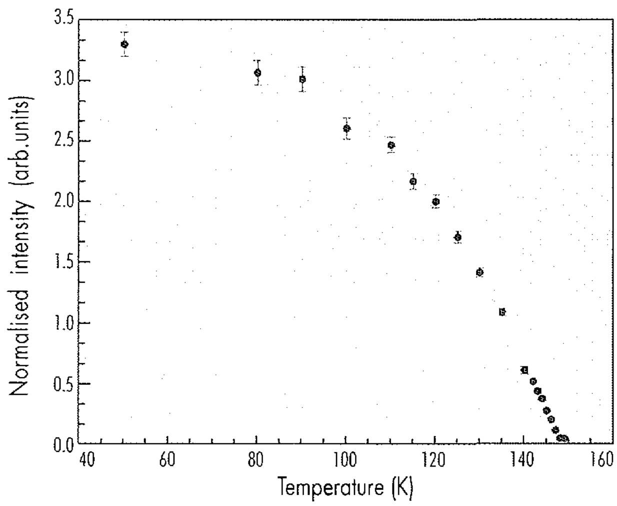

Figure 1: *Temperature dependence of the intensity of the (1/2 1/2 1/2) magnetic Bragg reflection of the MnO film (3500 Å) grown on sapphire Al_2O_3. The transition is continuous with a Néel temperature of 149 K, which is much higher than that in bulk MnO (T_N = 118 K). A MnO thin film of 530 Å grown on sapphire was also studied and the same temperature dependence was observed.*

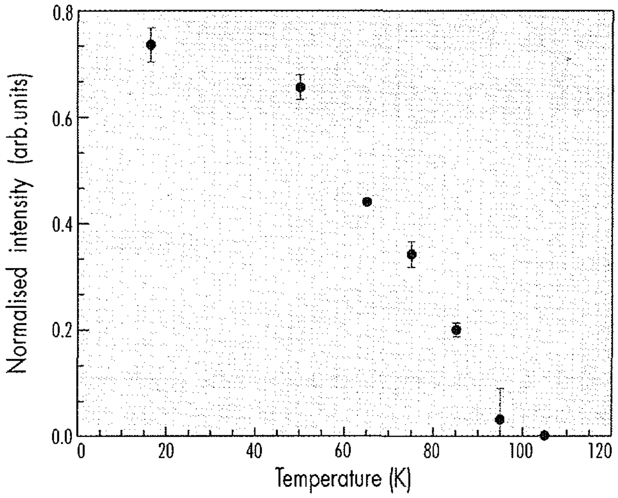

__Figure 2:__ Temperature dependence of the intensity of the (1/2 1/2 1/2) magnetic Bragg reflection of the MnO film (2500 Å) grown on MgO.The transition is continuous with a Néel temperature of about 100 K, which is now lower than in bulk MnO (T_N = 118 K).

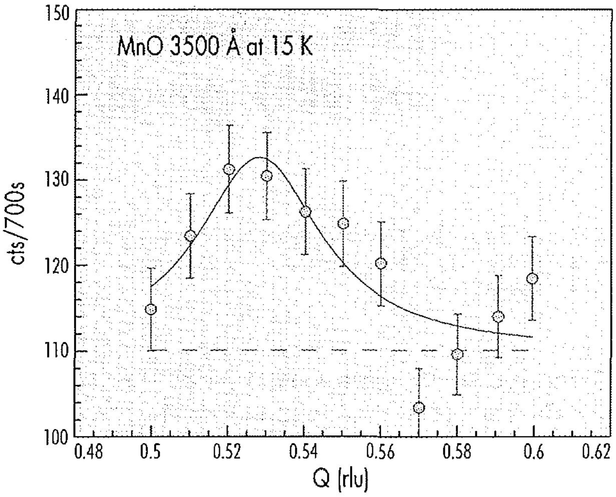

__Figure 3:__ Q-scan parallel to the surface normal (1 1 1) at fixed neutron loss energy of 8 meV, showing an enhancement at the position where magnons are expected.The straight line shows the background estimated from scattering above TN.

The results confirm the idea that the transition may be driven second order on going from bulk to film samples. However, the radically different transition-temperatures obtained, depending upon the substrate, indicate a significant impact of the substrate (assuming the preparation conditions of the films were sensibly similar). This strong role of the substrate on the Néel temperature implies that there is more than a simple reduction of dimensionality to consider. Indeed, dimensionality may not be a significant factor. For example, strain caused by substrate mismatch and the precise stoichiometry of the films could also play important roles. In this context, it is already established that changes of the Néel temperature observed in FeO_{1-x} compounds, may be related to their (controlled) departure from stoichiometry [4]. These initial experiments, which identify clear differences with the results of bulk MnO, open up interesting avenues of research.

In order to gain some information on the microscopic properties of the films, an attempt was made to characterise the magnetic excitation spectrum in the thickest film (3500 Å MnO on sapphire). A test experiment in the ordered phase at a sample temperature of 15 K was carried out on IN8. Figure 3 shows a Q-scan parallel to the surface normal at a fixed neutron energy loss of 8 meV. The small enhancement, observed around the scattering wave-vector (0.54 0.54 0.54), is close to the position where magnons have been observed in bulk MnO [5]. The straight line is an estimate of the background established using measurements at higher temperature (200 K). These promising results may open a way of studying the magnetic dynamics in two-dimensional systems.

Having demonstrated the feasibility of magnetic neutron-scattering experiments on such small quantities of material, there are clearly a number of open questions to be addressed for the MnO films. Amongst these are: (i) is it possible to quantify the roles of substrate lattice mismatch on T_N ? (ii) can one identify the moment direction within the (111) plane, and its absolute magnitude; are these parameters different from the bulk and do they vary as a function of film thickness and/or substrate ? (iii) finally, can one learn about the role of dimensionality on the dynamics ?

References

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