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A structural transition induced by a magnetic field in $La_{1-x}Sr_xMnO_3$

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Abstract

There has been phenomenal interest recently in rare earth oxides of the type $R_{1-x}A_xMnO_3$ (R = rare earth, A = alkaline earth) because of their extraordinary electronic and magnetic properties. These include $La_{1-x}Sr_xMnO_3$ which exhibits a range of structural and magnetic transitions as a function of temperature, pressure and magnetic field. The feature of greatest interest being the Colossal Magnetoresistance that these materials exhibit near the Curie temperature. We have performed neutron diffraction studies on a single crystal of $La_{0.835}Sr_{0.165}MnO_3$. This has a ferromagnetic transition at 264 K and a structural transition from a rhombohedral to an orthorhombic structure at 296 K. Our studies show that by applying a magnetic field it is possible to reverse the structural transition below 296 K. By following a magnetic peak we observe an increased ferromagnetic alignment before this transition takes place.

Keywords: Lanthanum strontium manganite; Colossal magnetoresistance; Structural transition; Magnetic transition; Field-induced structural transition

1. Introduction

Rare earth oxides of the type $R_{1-x}A_xMnO_3$ (R = rare earth, A = alkaline earth) have recently become the focus of much investigation because of their unusual electronic and magnetic properties. The most extraordinary feature is the colossal magnetoresistance that these materials exhibit near the Curie point. Among these materials is $La_{1-x}Sr_xMnO_3$ which exhibits a range of structural and magnetic phase transitions as a function of temperature, pressure and magnetic field. The parent compound $LaMnO_3$ is an antiferromagnetic insulator containing Mn^{3+} ions. Doping a fraction, x, of the La sites with divalent Sr causes a similar amount of Mn^{3+} to become Mn^{4+}. For doping levels $x > 0.05$ the material has a ferromagnetic ground state with a Curie temperature (T_c) that depends on x. The origin of this ferromagnetic ground state is then described in terms of a 'double exchange' [1] interaction between Mn^{3+} and Mn^{4+} ions via an intermediate oxygen. $La_{1-x}Sr_xMnO_3$ also exhibits a structural transition from a high temperature

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rhombohedral phase to a low temperature orthorhombic phase. The temperature of this transition (T_s) is also dependent on the level of doping, x. Here we show that by choosing a suitable doping level it is possible to induce the structural transition by application of a magnetic field.

2. Experimental results and discussion

In order to induce the structural transition in a magnetic field we required a sample in which the structural and magnetic phase transitions are close together with the ferromagnetic transition occurring below the structural transition. Recently a study by Asamitsu et al. [2] of $La_{1-x}Sr_xMnO_3$ ($x = 0.17$) including resistivity and striction measurements suggested that in this range of doping the structural transition can be induced by application of a magnetic field. Based on these observations single crystals with $x = 0.165$ and $x = 0.17$ were grown in an infra red image furnace using the floating zone method. X-ray Laue photographs showed that the resulting samples were high quality single crystals. The crystals were then characterised in our laboratory by measuring the resistivity, AC susceptibility and magnetisation as a function of temperature. Fig. 1 shows the temperature dependence of the resistivity and inset magnetisation for single crystal $La_{0.835}Sr_{0.165}MnO_3$. The resistivity shows an increase at 296 K associated with the structural transition and a large decrease near the Curie point of 264 K. There is hysteresis in the structural transition showing its first-order nature. Measurements on the $x = 0.17$ sample found a T_c of 283 K and a T_s of 183 K. This meant our $x = 0.165$ sample had the same transition temperatures as the $x = 0.17$ sample of Asamitsu et al. From these measurements it was clear that the $x = 0.165$ sample satisfied the proposed requirements for observing the field induced structural transition.

Neutron-diffraction measurements were carried out at the reactor source at the Institut Laue–Langevin in Grenoble, France [3]. In a preliminary experiment on the single crystal diffractometer D10 extensive structural measurements were made and by following the intensity of the [4 4 5] reflection

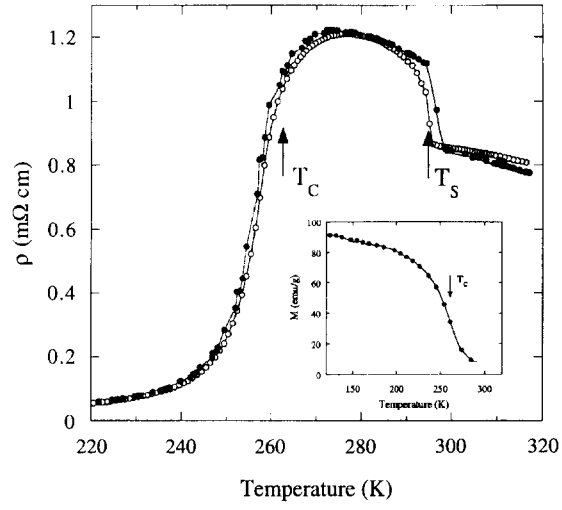

Fig. 1. Resistivity and inset magnetisation of single crystal $La_{0.835}Sr_{0.165}MnO_3$ as a function of temperature. The structural and magnetic phase transitions are marked with T_s and T_c, respectively. The resistivity was measured by a standard four-probe method, the open circles are for cooling and the filled circles are for warming. The magnetisation was measured using a VSM in a 1 T field.

the temperature of the structural phase transition was confirmed at 296 K. The [4 4 5] reflection is only present in the orthorhombic phase and so a measurement of its intensity gives a direct determination of the structural phase of the sample.

The field switching experiment was carried out on the polarised-neutron normal-beam diffractometer D3 which is equipped with a 4.6 Tesla cryomagnet. In this experiment the intensities of the [4 4 5] and [2 0 0] reflections were monitored as a function of temperature and magnetic field. The [2 0 0] is a weak nuclear reflection which shows a large increase in intensity at the onset of the ferromagnetic state. After the sample had been mounted and aligned it was cooled to 220 K by which time the transition to the orthorhombic phase was judged to be complete. The sample was then warmed to 290.5 K. At this temperature the sample is in the hysteretic region of the structural transition but is still in the orthorhombic phase.

At this temperature the magnetic field was gradually increased. Fig. 2 shows the intensity of the [4 4 5] reflection as a function of magnetic field. On

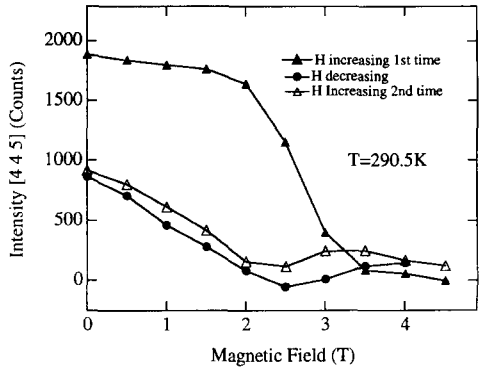

Fig. 2. Intensity of the [4 4 5] orthorhombic reflections as a function of magnetic field at 290.5 K. When the field is increased the first time, the intensity of the [4 4 5] reflection disappears at ~ 3 T showing the transition to the rhombohedral phase. On decreasing the field, the intensity of the [4 4 5] reflection increases only slightly indicating a partial transformation to the orthorhombic phase. On increasing the field for the second time the intensity follows the same path as the decreasing field and the sample goes fully into the rhombohedral phase at high fields.

increasing the field the first time the intensity of the [4 4 5] shows a decrease in intensity beginning at about 2 T. By about 3 T the intensity has disappeared showing that the magnetic field has induced the structural transition from the orthorhombic phase to the rhombohedral phase. On decreasing the field there is a slight increase in intensity below 2 T indicating that the sample partially transforms back into the orthorhombic phase but is unable to transform back completely at this temperature. On increasing the field a second time the sample reverts to being fully rhombohedral. A measurement of the [2 0 0] reflection showed an increase in intensity starting at about 0.5 T showing that there is an increase in ferromagnetic alignment before the structural transition takes place.

We have performed neutron scattering experiments in which we directly observe and confirm the bulk nature of the magnetic field induced phase transition in $La_{1-x}Sr_xMnO_3$ ($x = 0.165$). At the temperature at which the experiment was performed removal of the field does not produce a switch back and the transition is permanent. By varying the doping levels it is possible to adjust the structural and magnetic properties so that they are strongly coupled. As a result the structural phase transition can be induced by application of a magnetic field. When the field is applied there is an increased alignment of the magnetic moments before the structural transition takes place indicating that a ferromagnetic alignment of the spins encourages the structural phase transition from an orthorhombic to a rhombohedral phase in the presence of a magnetic field.

References

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