POLARONS IN COLOSSAL MAGNETORESISTIVE MATERIALS

The magnetic properties of the lanthanum manganese oxide class of materials have attracted tremendous interest recently because of the dramatic increase in conductivity these systems exhibit when the magnetic moments order ferromagnetically, either by lowering the temperature or by applying a magnetic field. This huge change in the carrier mobility, which has been given the name "colossal magnetoresistance" (CMR), is both of scientific and technological interest. In particular, it is anticipated that related materials may provide the next generation of read/write heads for the magnetic data storage industry, while the "half-metallic" behavior provides fully spin polarized electrons for use in magneto-electronics applications, and for sensors in a variety of applications such as in the automotive industry.

CMR can be strongly enhanced in systems with reduced dimensionality and so there has been considerable interest in the two-layer Ruddlesden-Popper compounds, $La_{2-2x}Sr_{1+2x}Mn_2O_7$. The reduced dimensionality leads to significant extension of the temperature range over which magnetic correlations are important, and thereby allows a detailed examination of the link between local spin correlations and the resulting magnetotransport. We have therefore investigated the magnetic correlations in $La_{1.2}Sr_{1.8}Mn_2O_7$ using neutron scattering. Over a large temperature range above $T_C = 112$ K, we found evidence for two-dimensional magnetic correlations which peak in intensity at the transition. Although the in-plane correlations are predominantly ferromagnetic, a canting of spins in neighboring planes within the bilayers, at an angle that is dependent both on temperature and magnetic field, was observed [1,2].

One of the central questions in the field of manganites concerns the lattice involvement in the mechanism of CMR. While the relation between ferromagnetism and conductivity was explained in terms of double exchange, it is now clear that a full understanding of these materials must include the lattice degrees of freedom. In particular, the formation of lattice polarons above the Curie temperature has been inferred from a variety of measurements, but direct evidence has been lacking.

Neutron measurements carried out at NIST, and X-ray measurements at the Advanced Photon Source, have revealed charge localization in the paramagnetic-insulating phase of the layered $La_{1.2}Sr_{1.8}Mn_2O_7$ CMR material, with the associated diffuse polaron scattering that originates from the lattice distortions around the localized charges. Figure 1(a) shows two of the observed incommensurate superlattice peaks associated with the charge ordering, characterized by the wave vector (0.3, 0, 1). Polarized neutron scattering has shown that the incommensurate superlattice peaks are pure structural reflections, originating most likely from Mn^{3+}-Mn^{4+} charge correlations.

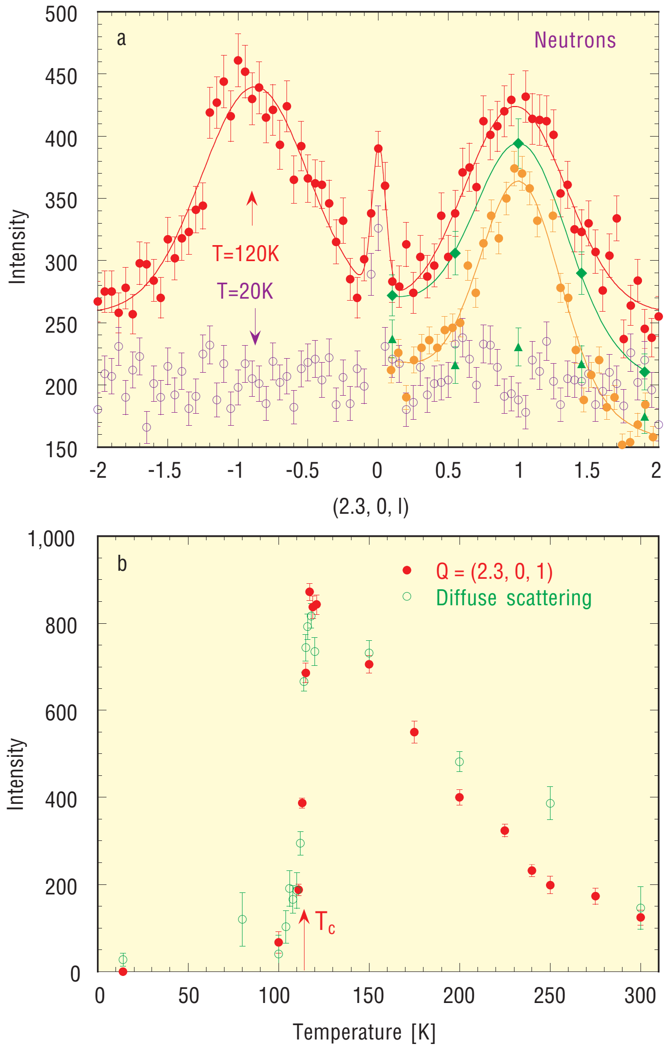

FIGURE 1. (a) *l*-scans through the charge ordering peaks at (2.3, 0, ±1) at T=120 K: energy-integrated (red), elastic (orange), non-spin-flip scattering measured with polarized neutrons (green diamonds), spin-flip scattering (green triangles). The orange and green data points have been scaled by appropriate factors. The *l*-scan at T=20 K (open purple circles) shows that the charge peaks have vanished. (b) Temperature-dependence of the intensity of the superlattice peak (2.3, 0, 1) (red), and of the diffuse scattering after correction for the phonon scattering (green), showing that the charge order and lattice polarons collapse at the Curie temperature.

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These correlations are quasi-static on a time scale $\tau \approx \hbar/2\Delta E \approx 1$ ps set by the energy resolution of the instrument. The superlattice peaks are broader than the *q* resolution in both *h* and *l* directions, showing that both the in-plane and out-of-plane charge correlations remain short range at all temperatures. The charge order melts as the insulator-to-metal transition is traversed and long-range ferromagnetic order is established [3], as shown in Fig. 1b. By similarity with the 3D perovskite manganites and with the cuprates, this scattering may originate from a charge-ordered stripe phase above T_C, which is destroyed when the double exchange mechanism drives the system metallic. The lattice strain induced by the localized charges gives rise to a four-fold, lobe-shaped pattern of diffuse scattering around the Bragg peaks. The upper panel in Fig. 2 shows a contour plot of the diffuse X-ray scattering in the [*h*, 0, *l*] plane around the (2, 0, 0) reflection. Only the $l > 0$ half is shown, but the pattern is symmetric with respect to $l = 0$, as proved by the neutron *l*-scans in the lower panel of Fig. 2. The sharp rod of scattering along the [0, 0, *l*] direction is resolution limited in the [*h*, *k*, 0] plane and is associated with stacking faults. Part of the lobe-shaped diffuse scattering is due to conventional acoustic phonons, while the temperature dependence of the additional, polaron scattering (green open circles in Fig. 1b), arises from static or quasi-static atomic displacements as revealed by elastic neutron scattering (see Fig. 2 bottom). The lattice strain caused by the polarons relaxes when the short-range charge order melts at the Curie temperature, providing compelling evidence of the role of polarons in the origin of CMR. Further work is now in progress to determine if the charge melting can be controlled by the application of magnetic or electric fields, which would open up completely new avenues for applications.

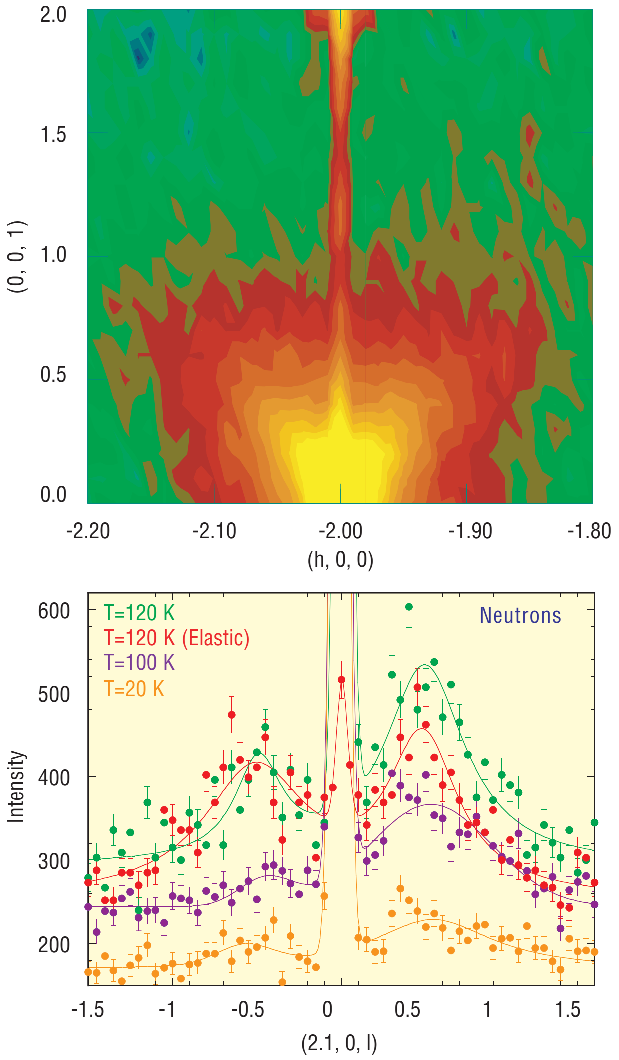

FIGURE 2. (Top) Contour plot showing the lobe-shaped pattern of diffuse scattering around (2, 0, 0). (Bottom) Neutron energy-integrated *l*-scans across the diffuse scattering in the upper panel, for T=120, 100, and 20 K. The scan at T=120 K (red circles) is an elastic scan, and has been scaled by an appropriate factor.

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