Polaron scattering in colossal magnetoresistive oxides

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Introduction

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 increase 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 these materials may provide the next generation of read/write heads for the magnetic data storage industry. Their half-metallic behavior provides fully spin-polarized electrons for use in magneto-electronics applications and for sensors in a variety of applications (e.g., in the automotive industry).

CMR can be strongly enhanced in systems with reduced dimensionality and thus 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. 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.

Methods and Materials

The measurements were performed on a single crystal of the double-layer compound $La_{1.2}Sr_{1.8}Mn_2O_7$, cleaved from a boule that was grown using the floating-zone technique. The x-ray data were taken on the 1-ID-C diffractometer at the Advanced Photon Source, mostly using a high-energy beam of 36 keV to provide enough penetration in transmission geometry. Additional measurements were taken in reflection geometry with 21 keV. A wide range of reciprocal space was explored, including the (*h*0*l*) and (*hhl*) planes. The crystal structure of $La_{1.2}Sr_{1.8}Mn_2O_7$ is body-centered tetragonal (space group *I*4/*mmm*) with $a \cong 3.87$ Å and $c \cong 20.15$ Å, and consists of MnO_2 bilayers separated by (La, Sr) sheets. Previous neutron scattering measurements [1] have evidenced two-dimensional ferromagnetic correlations, which peak in intensity at the combined metal insulator and Curie transition at $T_C = 112$ K and extend over a large temperature range above T_C.

Results

Our measurements 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 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. The sharp rod of scattering

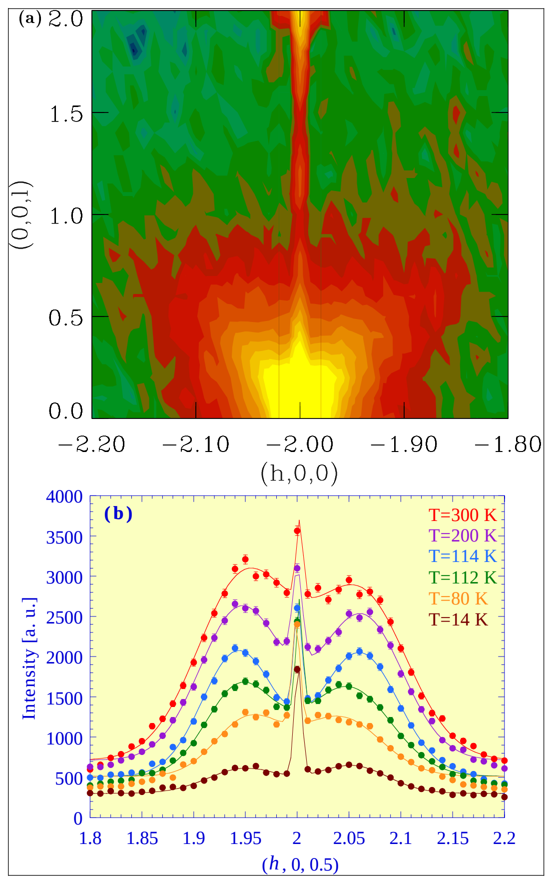

Figure 1: (a) Contour plot showing the lobe-shaped pattern of x-ray diffuse scattering around (2, 0, 0). (b) X-ray *h*-scans across the diffuse scattering at *l* = 0.5 at a series of temperatures.

along the [0, 0, l] direction is resolution limited in the [h, k, 0] plane and is associated with stacking faults. The h-scans across the diffuse scattering at l = 0.5 shown in Figure 1(b) clearly indicate that this diffuse scattering is anisotropic and strongly temperature dependent, with a dramatic response at T_C. Part of this diffuse scattering is due to acoustic phonons, but the sudden change at T_C cannot be due to conventional acoustic phonons, as confirmed by inelastic neutron measurements. A good description of the **q**-dependence of this diffuse scattering has been obtained in terms of Huang scattering, consistent with a Jahn-Teller type distortion around the Mn^{3+} ions.

The measurements also reveal the presence of broad incommensurate peaks in the paramagnetic phase, as shown by the contour plot of the x-ray intensity at 125 K in the (hk) plane at l = 18 in Figure 2(a). These peaks are characterized by a wave vector (±ε, 0, ±1) as measured from the nearest fundamental Bragg peak, where ε ≅ 0.3 (in terms of reciprocal lattice units [2π/a, 0, 2π/c]). The in-plane incommensurability is evident in the x-ray h-scans shown in Figure 2(b) at different temperatures. Note that this peak increases and then rapidly decreases in intensity as we cool through T_C. Figure 2(c) shows that the temperature dependence of the incommensurate peak intensity is remarkably similar to the Huang scattering derived from the x-ray scattering by subtracting the estimated thermal diffuse scattering. This indicates that both types of scattering are associated with the development of polarons above T_C. The incommensurate peak intensity falls slightly more rapidly than the Huang scattering with increasing temperature. This is consistent with ascribing the Huang scattering to individual polarons and the incommensurate peaks to polaron correlations, which become stronger with decreasing temperature. Below T_C we observe a "melting" of the polaron correlations occurring simultaneously with the collapse of the polarons themselves. These results have been confirmed using neutron scattering at the National Institute of Standards and Technology Center for Neutron Research, and provide important new insights into the relation of polarons to colossal magnetoresistance.

Discussion

Charge and orbital ordering have been observed at low temperature in a number of insulating, antiferromagnetic cubic manganites at small and large (x ≥ 0.5) doping, as well as in layered manganites with x = 0.5. However, short-range charge ordering in the paramagnetic phase of an optimally doped CMR ferromagnet is a novel feature observed here. In the present x = 0.4 system, the charge correlations are not strong enough to win the competition with the double-exchange interaction, and the charges delocalize at the ferromagnetic transition where the charge peaks collapse and the lattice strain relaxes. It is the delicate balance between double exchange, Coulomb repulsion, and the lattice strain field that dictates whether the material is a ferromagnetic metal or charge-ordered insulator at low temperatures.

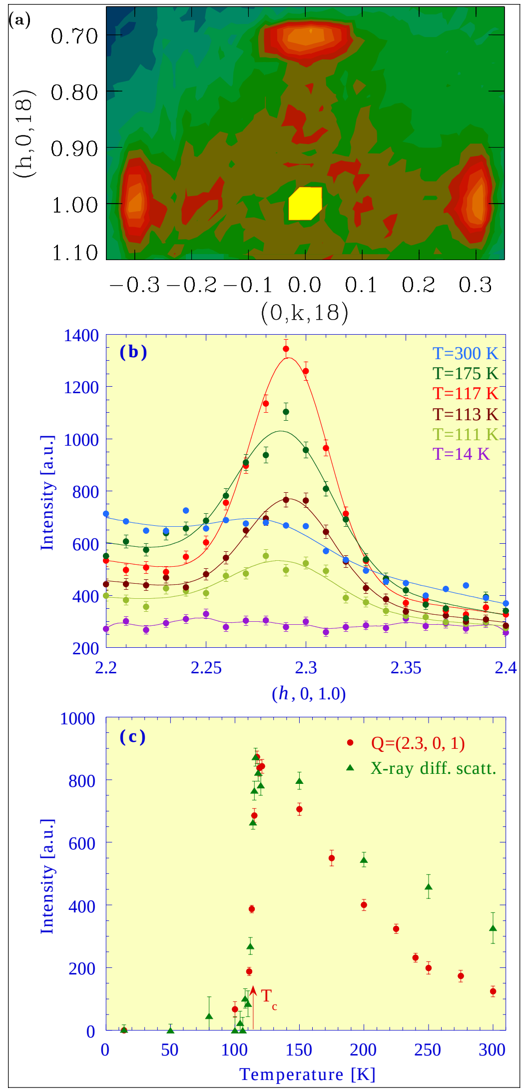

Figure 2: (a) Contour plot of the x-ray intensity in the (hk) plane at l = 18, collected at T=125 K. Three incommensurate peaks due to polaron ordering are observed, characterized by the wave vector (±ε, 0, l) or (0, ±ε, l). The expected fourth peak was not accessible experimentally. (b) X-ray h-scans through the incommensurate peak (2.3, 0, 1) at different temperatures. The higher scattering at small h is due to the proximity of the lobe-shaped diffuse scattering around the Bragg peak. (c) Temperature dependence of the x-ray intensity of the (2.3, 0, 1) incommensurate peak (red closed circles), and of the diffuse scattering after correction for the phonon contribution (green closed triangles).

Acknowledgments

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