Polaronic Distortions and Their Correlations in the Bilayer Manganites

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Introduction

X-ray-diffraction studies of the colossal magnetoresistance (CMR) manganese oxides revealed diffuse (Huang) scattering due to polarons and their correlations in both quasi-2-dimensional layered (40% hole-doped)[1] and 3-dimensional perovskite manganites.[2] Such scattering is a direct manifestation of Jahn-Teller (JT) distortions of Mn^{3+}, which is necessary to explain the enhanced resistivity (ρ) above T_c and CMR phenomena. A more recent study has also elucidated the nature of these nanometer-scale polaron correlations in the 40% doped material.[3] The present study is concerned with the properties of the polarons in the 36% ($La_{1.28}Sr_{1.72}Mn_2O_7$; I4/mmm) and 44% ($La_{1.12}Sr_{1.88}Mn_2O_7$) hole-doped layered manganites, respectively. Unlike those in the intermediate doping regime (32%-42%), which are ferromagnetic (F) metals in their ground state, the 44% doped compound is antiferromagnetic (AF) below 140K and develops a ferromagnetic component below 90K. However, a truly metallic state is never reached.

Experimental Details

The high-energy-x-ray-diffraction study was carried out on the MU-CAT 6-ID beamline. Using a Si(111) monochromator, the incident energy was chosen to be 35 keV, below the La K edge, to optimize bulk penetration. Single crystals used in this study were grown at the Materials Science Division using a floating-zone technique described elsewhere.4 A few crystallites from a batch were characterized using an in-house rotating-anode source and a small crystal (44% doping) with a mosaic of ~0.075° was selected. The mosaic for the 36% doped material was superior and was observed to be ~0.027°. The crystals have plate-like morphology with the c axis perpendicular to the flat face. Count rate was recorded using a Ge solid-state detector. The samples were sealed in a Be can filled with He gas and cooled in a close-cycle He refrigerator.

Results

The results are summarized in Figs. 1 and 2. In the 44% doped material, two diffuse lobes and a central peak were observed (Fig. 2, inset) along [0.05 0 L] in the vicinity of the (0, 0, 12) and (0, 0, 14) Bragg peaks, in contrast to the behavior of the 36% and 40% hole-doped systems, where no central peak was found. The presence of an additional peak suggests a different mode of JT distortions in the 44% doped sample.

In the paramagnetic (P) phase, both lobes and the central peak are present (see Figs. 1 and 2). As the system enters the AF phase, the intensities of the lobes increase, reaching a maximum above T_c (Fig. 1, top left). The intensities then decrease through T_c, and become nearly T independent. This behavior qualitatively mimics the T dependence of ρ. The bottom-left panel shows the behavior of the short-range correlation peak, (0.3, 0, 1.0), among the polarons. The polarons remain strongly correlated at all T. The maximum correlation is observed above T_c in the AF phase, in agreement with the maximum in ρ in this region and a recent neutron-diffraction work on a sample with a 45% doping level.[5] In stark contrast, both diffuse scattering and polaronic correlations sharply diminishes below T_c in the 36% doped material when it becomes a F metal (right panels, Fig. 1). The latter behavior is similar to that observed in the 40% hole-doped material.[1] Interestingly, it is the central peak, observed in $La_{1.12}Sr_{1.88}Mn_2O_7$ that

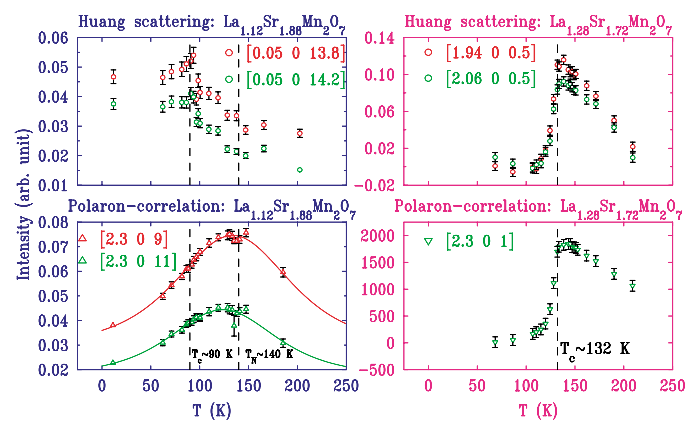

FIG. 1 Temperature dependence of diffuse scattering from polarons and their correlations. Blue (left) and magenta (right) panels are for the 44% and 36% hole-doped samples, respectively. Thermal diffuse scattering has been subtracted from the data shown in the right panels.

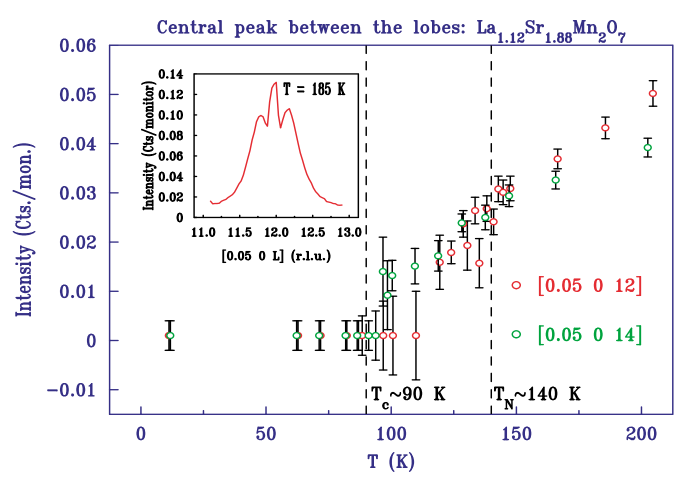

FIG. 2 Temperature dependence of the central diffuse peak. Inset: a scan showing the central peak in between the diffuse lobes.

becomes very weak above T_c and disappears in the F phase (Fig. 2), indicating a clear change in symmetry of the JT distortions at this transition.

Conclusions

We have shown the existence of polarons in the P, AF, and F phases of the 44% hole-doped bilayer manganite system, consistent with its nonmetallic behavior according to the resistivity measurements. The manifestation of a different JT symmetry prevailing in the P and AF phases of this compound is an intriguing new result. Theoretical work to elucidate the nature of this JT mode and its relation to both electronic transport and magnetic order is clearly needed. The behavior of the polarons in the 36% doped material is similar to those of the 40% doped compound studied[1] earlier.

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References

[1] L. Vasiliu-Doloc, S. Rosenkranz, R. Osborn, S.K. Sinha, J.W. Lynn, J. Mesot, O.H. Seeck, G. Preosti, A.J. Fedro, and J.F. Mitchell, Phys. Rev. Lett. **83**, 4393 (1999).

[2] S. Shimomura, N. Wakabayashi, H. Kuwahara, and Y. Tokura, Phys. Rev. Lett. **83**, 4389 (1999).

[3] B.J. Campbell, R. Osborn, D.N. Argyriou, L. Vasiliu-Doloc, J.F. Mitchell, S.K. Sinha, U. Ruett, C.D. Ling, Z. Islam, and J.W. Lynn, to be published.

[4] J.F. Mitchell, D.N. Argyriou, J.D. Jorgensen, D.G. Hinks, C.D. Potter, and S.D. Bader, Phys. Rev. B **55**, 63 (1997).

[5] M. Kubota, Y. Oohara, H. Yoshizawa, H. Fujioka, K. Shimizu, K. Hirota, Y. Moritomo, Y. Endoh, J. Phys. Soc. Jpn. **69**, 1986 (2000).