First-Order Nature of the Ferromagnetic Transition in CMR Oxides

J. W. Lynn[1,2], C. P. Adams [1,2], V.N. Smolyaninova[1], K. Ghosh[1], R.L Greene[1], W. Ratcliff II[3],and S-W. Cheong[3]

[1]*Center for Superconductivity Research, University of Maryland, College Park, Maryland 20742.*

[2]*NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, Maryland*

[3]*Department of Physics, Rutgers University, Piscataway, NJ 08854*

The phenomenon of colossal magneto-resistance (CMR) in the manganese oxide class of materials (such as the $La_{1-x}Ca_xMnO_3$ prototype) has rekindled intense interest in these systems. In the manganites the charge, spin, and lattice degrees of freedom are strongly coupled together, leading to a delicate balance of interactions that gives rise to a rich variety of physical phenomena of current interest in condensed matter physics. These include a metal-insulator transition concomitant with ferromagnetic ordering, charge and orbital ordering, polaron formation, electronic phase separation, and spin and charge stripes. The manganites are also related to the high T_C cuprate oxides and relaxor ferroelectrics, with a commonality of many of the materials properties and underlying physical concepts. Recent progress in our understanding of these systems has provided insights into the manganites, and a deeper understanding of the fundamental properties of the manganites will surely elucidate the shared concepts underlying all three classes of materials. The CMR materials also offer potential in a number of technologies, such as for read/write heads, sensors, and spin-polarized electronics.

One of the fundamental aspects of the CMR materials to be elucidated is their remarkable sensitivity to small external perturbations, which produces (among other properties) the CMR effect itself. In our initial investigations of the ferromagnetic—paramagnetic transition[1] we found that the transition was unconventional, and could best be interpreted as a discontinuous transition between two-phases, the ferromagnetic-metallic state at low temperatures and the paramagnetic-insulating state at high temperatures. Subsequent work on single crystals[2] revealed that short-range polaron correlations form in the paramagnetic state, above the ferromagnetic transition, and these polarons trap the carriers and produce the insulating state.

The formation of these polarons appears to truncate the ferromagnetic transition, producing an anomalous temperature dependence to the spin stiffness $D(T)$, with $D(T_C)$ being finite rather than approaching zero. To investigate this behavior in detail we have measured the spin dynamics for an optimally doped x=3/8 sample, with a Curie temperature of 267.5 K. The observed $D(T)$ is shown in the figure, where we see that $D(T_C)$ is finite as expected. Moreover, when we replace O^{16} with O^{18}, T_C is reduced while the temperature dependence of the spin dynamics is unaffected. The original data[1] also fits on the same $D(T)$ presented in Fig. 1, but with a much lower ordering temperature (250 K).

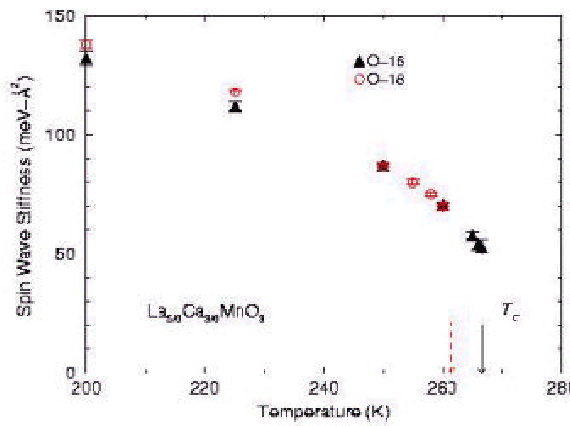

Figure 1: T dependence of the spin stiffness for optimally doped LCMO. The replacement of O^{16} with O^{18} changes T_C but has no effect on the spin dynamics.

These experimental neutron scattering results reveal that in the LCMO system T_C is not determined by the exchange interactions in the system, but rather by the formation of the (lattice + magnetic) polarons in the system. The effective T_C due the exchange interactions is ~280 K for x=3/8, but the polarons form at a lower T and truncate the ferromagnetism. At lower x the observed T_C is lower, and the concomitant anomaly in D(T) larger. In the Sr and Ba doped systems, on the other hand, T_C appears to be below the formation temperature for polarons, and the magnetic transition is then continuous and the spin dynamics conventional.

The next step in these studies is to determine what controls the formation of polarons, and why only short-range polaron correlations are observed in these (optimally doped) CMR systems.. Further work is in progress.

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[2]C. P. Adams, J. W. Lynn, Y. M. Mukovskii, A. A. Arsenov, and D. A. Shulyatev, Phys. Rev. Lett. **85**, 3954 (2000). J. W. Lynn, C. P. Adams, Y. M. Mukovskii, A. A. Arsenov, and D. A. Shulyatev, J. Appl. Phys. **89**, 6846 (2001); preprint.