Magnetic transition and electronic transport in colossal magnetoresistance perovskites

S. E. Lofland and S. M. Bhagat
Department of Physics, University of Maryland, College Park, Maryland 20742-4111

K. Ghosh and R. L. Greene
Center for Superconductivity Research, University of Maryland, College Park, Maryland 20742-4111

S. G. Karabashev, D. A. Shulyatev, A. A. Arsenov, and Y. Mukovskii
Moscow State Steel and Alloys Institute, Leninskii Prospect 4, Moscow 117936, Russia
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We present here the results of a careful study of the magnetic transition, as well as, the dc resistivity in the same specimen of a single crystal of $La_{0.8}Sr_{0.2}MnO_3$. We find that the Curie temperature of the specimen is far below the point where the temperature coefficient of the resistivity changes sign. [S0163-1829(97)05245-4]

Numerous experimental studies[1,2] assert that the temperature (T_{MI}) of the so-called metal-insulator (MI) transition (equivalently designated the peak temperature, T_p) observed in the colossal magnetoresistance manganites, such as $La_{0.8}Sr_{0.2}MnO_3$, coincides with that marking the onset of ferromagnetism (T_C). Many models[3–5] are also predicated on a close coincidence of these phenomena since the onset of spin alignment is invoked to account for the sharp drop in resistivity ρ at the MI transition. A few reports[6,7] have, however, suggested that T_{MI} may not be the same as T_C. It is clear that if T_{MI} is significantly different from T_C, an adequate theory of the transport properties of these interesting compounds must reflect this distinction. Since T_C, in particular, is likely to be rather sensitive to the homogeneity, strain distribution, and local chemistry of the material, it is imperative to use samples of the highest quality to settle this question unequivocally. With this in view, we have carried out a careful study of both the magnetic transition as well as the dc resistivity on the *same specimen* of a single crystal of $La_{0.8}Sr_{0.2}MnO_3$. As we shall see, T_{MI} is, in fact, several K higher than T_C so that on lowering T the resistivity registers a significant drop well before the onset of spin alignment. Equivalently, $d\rho/dT$ is positive, that is, ''metal-like'' on both ''sides'' of the paramagnetic-ferromagnetic (PM-FM) transition. Whereas spin alignment must play a role in the transport properties of the manganites, it is clear that one requires additional considerations to account for the present observations.

A single crystal of $La_{0.8}Sr_{0.2}MnO_3$ was made by a floating zone technique. The process has been described in adequate detail in a previous report.[8] From the resulting 4 mm diameter rod, a 0.2 mm-thick disk was cut. Its surface was subsequently polished with 0.05 μm alumina powder. Energy-dispersive x-ray analysis and x-ray diffraction attest to the stoichiometry and structural integrity. However, the ferromagnetic resonance data (Fig. 4 below), which are among the most demanding of sample quality, show that defects and inhomogeneities are not entirely absent.

The magnetic transition was studied in detail by several techniques. First, the low-field (10 Oe), low-frequency (135 Hz) ac susceptibility (χ_{ac}) was measured on a locally built mutual-inductance susceptometer. The magnetization M was measured in a superconducting quantum interference device (SQUID) magnetometer for a field H of up to 10 kOe applied in the plane of the sample. Again, the spontaneous magnetization was accessed by using a ferromagnetic antiresonance (FMAR) method[9] at 10 GHz. Further, ferromagnetic resonance (FMR) spectra were taken at 10 GHz by exposing the center of the disk face using a microwave microscope described in detail elsewhere.[10] Spectra were obtained with the field either parallel or normal to the plane of the disk. Small corrections to the resonance equations due to the finite demagnetizing field were made from the determination of the local demagnetizing field H_D. H_D was found by placing a paramagnetic marker[11] (a grain of diphenyl-picryl-hydrazyl) atop the spot of interest and measuring the shift in the paramagnetic resonance field compared to that of a grain placed elsewhere in the same cavity, well away from the sample. H_D also served as an additional check on the T dependence of M since $H_D = DM$ where D is the effective demagnetization factor at the center of the disk surface.

Figure 1 shows the temperature variation of the ac susceptibility. As expected for a soft ferromagnetic sample with a well-defined demagnetization factor, the curve is indeed

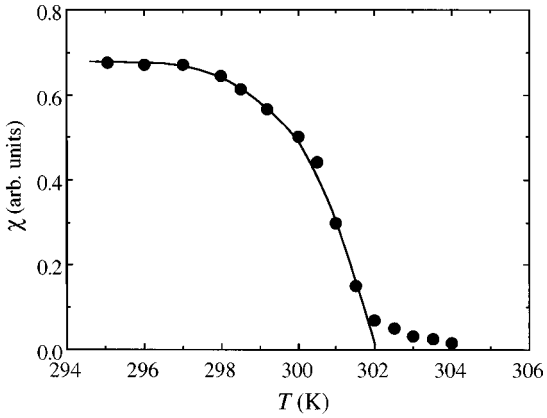

FIG. 1. ac susceptibility vs T for a $La_{0.8}Sr_{0.2}MnO_3$ crystal. The line serves as a guide to the eye. Note the highly expanded temperature scale.

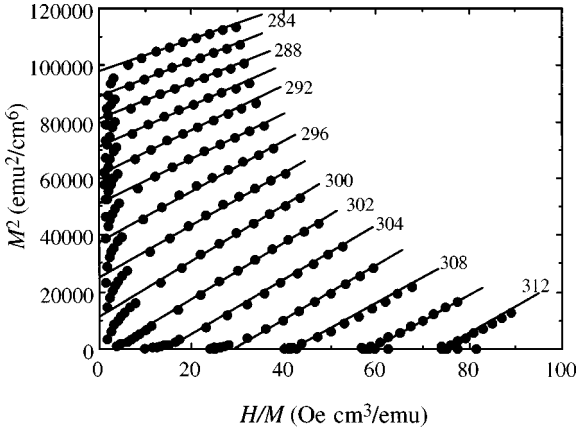

FIG. 2. Arrott plot of magnetic isotherms obtained from dc magnetization measurements on the $La_{0.8}Sr_{0.2}MnO_3$ crystal of Fig. 1. Here the exponents β and γ were given their mean-field values, 0.5 and 1, respectively. Concomitantly, T_C is about 301 K. The numbers designate the temperature of the isotherm.

sharp (<3 K wide). The data in Fig. 1 suggest that $T_C = 302 \pm 2$ K.

A conventional method for accessing T_C is to present the magnetic isotherms as a generalized Arrott plot. One displays $M^{1/\beta}$ as a function of $(H/M)^{1/\gamma}$ where β and γ are the critical exponents and looks for the temperature whose isotherm intersects the origin. In the simplest case, β and γ are given their mean-field values, 0.5 and 1, respectively. The results obtained from the SQUID measurements of dc magnetization M are shown in Fig. 2 and mark $300 < T_C < 302$ K. To encompass other possibilities β (γ) was varied between 0.33 and 0.5 (1.33 and 1). The concomitant T_C values were found to lie between 299 and 302 K.

The magnetization values obtained[9,10] from the three resonance methods, FMAR, FMR (in both the parallel and perpendicular geometries) as well as the paramagnetic marker technique are all in accord (Fig. 3). The full line shows the fit to the scaling law, $M \propto (T_C - T)^{\beta}$ where $T_C = 304 \pm 3$ K and $\beta = 0.34 \pm 0.05$.

Thus, all the magnetic measurements (χ_{ac}, dc magnetization, resonance) *agree with one another* and yield $T_C = 302$

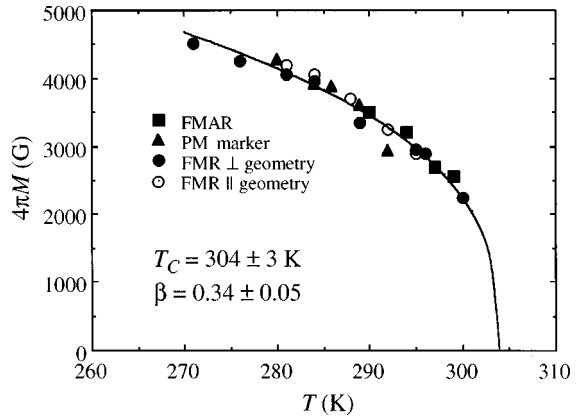

FIG. 3. $4\pi M$ vs T for the $La_{0.8}Sr_{0.2}MnO_3$ crystal of Figs. 1 and 2 determined from microwave measurements (see text and Ref. 9). The full line represents a least-squares fit to the scaling law $\propto (T_C - T)^{\beta}$ with $T_C = 304$ K and $\beta = 0.34$.

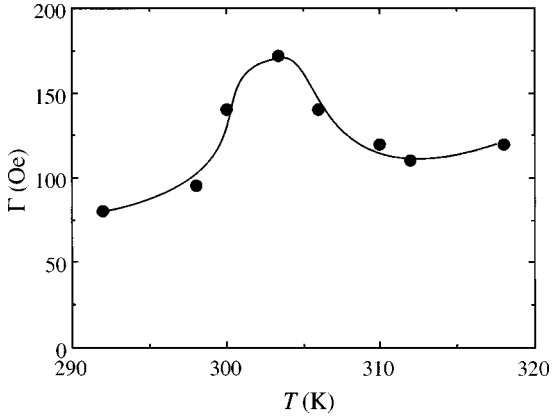

FIG. 4. FMR linewidth Γ vs T for the $La_{0.8}Sr_{0.2}MnO_3$ crystal. The line is to serve as a guide to the eyes. The peak in Γ indicates a small (~ 3 K) distribution in T_C values (cf. Ref. 13). Even so, the linewidth is small (<100 Oe) away from the peak.

± 2 K.[12] However, it must be pointed out that the FMR linewidth Γ (Fig. 4) has a peak near 304 K. As noted earlier,[13] this is symptomatic of a sample with a T_C distribution. In Ref. 13, it was shown that the temperature at which the linewidth is a maximum corresponds to the average T_C while the width reflects the spread in T_C values, which in this case amounts to about 3 K. This is quite small compared to that of most other bulk samples.[13] Although one would have preferred no such T_C distribution, the present crystal is good as is also evidenced by the fact that at temperatures away from the peak, the line is rather narrow (<100 Oe). The T_C spread is the most likely cause for our inability to obtain a more precise value for β.

With the magnetic transition temperature fixed, the transport characteristics are now discussed. Shown in Fig. 5 is the resistivity obtained by a four-probe method. The resistivity reaches a peak value of about 3.5 mΩ cm and drops to about 70 $\mu\Omega$ cm at 5 K. The relatively small resistivity at low T is another indication that the crystal is of high quality. At high T (>340 K), the resistivity enters the ''insulating'' regime and becomes semiconductorlike; $\rho = \rho_0 \exp(E_a/kT)$ with $E_a \sim 40$ meV, in accord with data taken on similar crystals.[14,15] However, one must note that the present estimate of E_a may be low because the data were taken for only

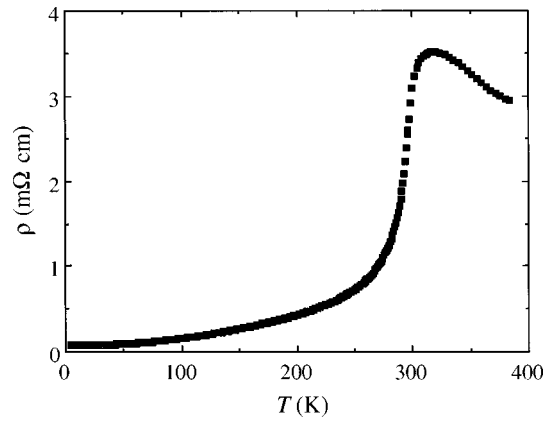

FIG. 5. Resistivity vs T for the $La_{0.8}Sr_{0.2}MnO_3$ crystal. Note that at 5 K $\rho = 70$ $\mu\Omega$ cm, attesting to the high quality of the sample.

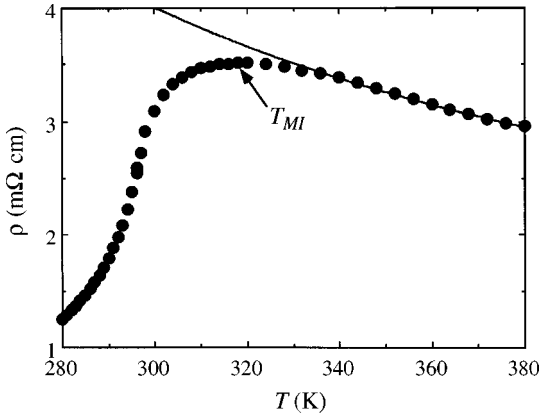

FIG. 6. Resistivity vs T for the $La_{0.8}Sr_{0.2}MnO_3$ crystal near $T_{MI}=318\pm2$ K. The full line represents $\rho=\rho_0\exp(E_a/kT)$ with $E_a\sim40$ meV and deviates from the data near 335 K. Note that $T_C=302\pm2$ K (Figs. 1–3) is well below T_{MI}.

a narrow temperature range above T_{MI}. As usual, T_{MI} (or T_p) marks the temperature where $d\rho/dT$ changes sign.

Figure 6 shows the resistivity in the region near T_C. Note that T_{MI} occurs at 318 ± 2 K, well above T_C, and $d\rho/dT$ is ''metallic'' as the magnetic transition is traversed. It is useful to point out that ρ deviates from the semiconducting (insulating) behavior at $T<335$ K (Fig. 5, full line). Since the appearance of metallic behavior is supposed to be connected to that of spin alignment, the correlation must occur at temperatures significantly above that for the onset of long-range order (LRO), i.e., T_C.

Our recent ESR studies[16] on powders taken from this crystal indicate that the Curie-Weiss temperature θ_p is about 354 K. One can hypothesize that the MI transition is brought about by the presence of short-range magnetic correlations that occur in the temperature range $T_C<T<\theta_p$. At first sight, this may seem to conflict with the claim that LRO is a requisite for delocalization since, in principle, both magnetization and resistivity are static measurements. However, one must bear in mind that resistivity is a dynamic property in that there is a characteristic hopping (or relaxation) time. If the transport time becomes comparable to the lifetime of the magnetic correlations, short-range order will appear to be static to the charge carriers and delocalization can take place without LRO.

In conclusion, we have carefully analyzed the magnetic (PM-FM) and transport (MI) transitions of a single crystal of $La_{0.8}Sr_{0.2}MnO_3$. Its Curie temperature is 302 ± 2 K while the metal-insulator transition (resistivity peak) occurs at 318 ± 2 K. This difference should not be neglected in any complete theory of the CMR materials.

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