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Fitting of transport measurements in polycrystalline $La_{2/3}Ca_{1/3}MnO_3$

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Abstract

Using a previous qualitative explanation to describe the transport properties of polycrystalline $La_{2/3}Ca_{1/3}MnO_3$ thick films, we achieved a good fit of the temperature dependence of the resistance $R(T)$. Depending on the sample, we have observed different metal–insulator (MI) transitions while the magnetic behavior is always similar. Small regions of depleted T_c adjacent to the grain boundary could have an important resistance contribution without affecting the magnetic properties in an appreciable manner. In this work, we achieve a quantitative explanation for the different transport behaviors that we have observed experimentally. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Manganites; Metal–insulator transition; Extrinsic magnetoresistance

The effect of grain boundaries (GB) on the transport properties of manganites has been extensively studied [1–4], fundamentally because some specific properties of polycrystalline samples make them attractive for applications usually requiring large MR effects at small applied magnetic fields. Several models have been proposed in order to describe the GB influence on the transport properties of manganites [1,2,5–8]. In particular, it has been previously suggested [2,7,8], that the presence of small regions of depleted T_c located around the grain boundary could play an important role in the behavior of polycrystalline samples.

Depending on the sample, we have observed [7,8] that the temperature of maximum resistance (T_p) can be equal or significantly lower than T_c (shift up to 60 K). In this work, we propose a quantitative explanation for the different MI transitions and obtain good fits of the transport properties of polycrystalline $La_{2/3}Ca_{1/3}MnO_3$ thick films over the whole temperature range. In order to explain the transport behavior of our samples we propose that regions of depleted T_c adjacent to the GB could play an important role in the characteristics of the MI transition.

To clarify the role of the GB in the transport properties of polycrystalline samples, we have grown thick films under different conditions [9] using the standard "paint-on" method. The transport measurements were carried out using the four-probe method and DC magnetic susceptibility was measured using a SQUID magnetometer.

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Fig. 1 shows resistance and magnetization vs temperature curves for three characteristic thick films. The magnetization curves were measured at 0.1 kOe and the $R(T)$ curves were obtained at zero-applied magnetic field. Fig. 1(a) shows a broad MI transition with a maximum resistance at T_p, 60 K below T_c. The sharpness of the magnetic transition allows us to suppose that the cores of the grains have $T_c \approx 250$ K (the sample Curie temperature) while T_p is around 190 K. Fig. 1(c) shows a very different behavior characterized by a lower resistance due to a better connectivity between grains [9]. In this case, T_p is near T_c. Fig. 1 (b) is an intermediate case. The transport curve of Fig. 1(b) shows two small $R(T)$ maxima centered at two different temperatures. We suppose that the sample of Fig. 1(b) have two similar resistance contribution, one from the core of the grains and the other from regions of depleted T_c. This behavior, showing two maxima in $R(T)$, has been observed before in artificial GB junctions grown on bicrystalline substrates [2]. The magnetic properties of the three samples are similar but the $R(T)$ curves are quite different. This suggests the importance of the quality of the GB for the transport properties.

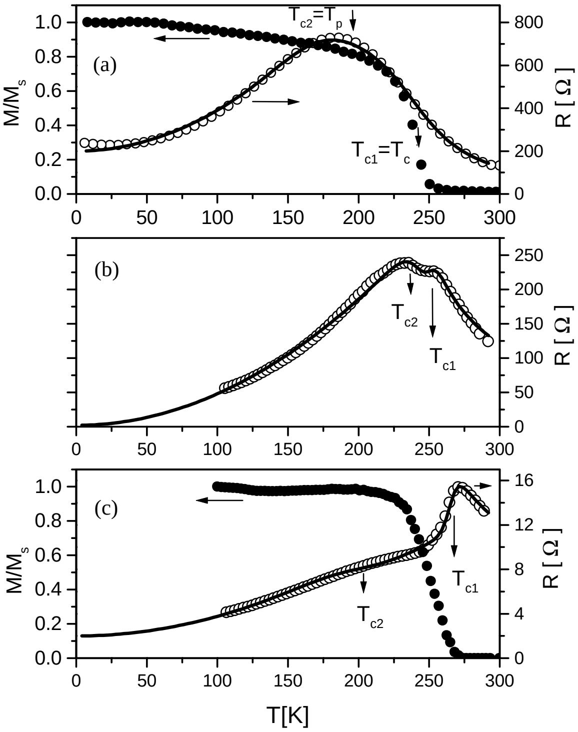

Fig. 1. Transport and magnetic properties (circles) of three typical samples. We also show the theoretical $R(T)$ curves (lines). Parameters used in the fitting: Fig. 1(a) $A_1 = A_2 = 100\,\Omega$ (obtained at $T \to 0$), $T_{c1} = T_c = 240$ K, $T_{c2} = T_p = 200$ K (directly determined from the experimental data), $R_1 = 0.7\,\Omega$, $R_2 = 3\,\Omega$, $E_{g1}/k_B = 750$ K, $E_{g2}/k_B = 750$ K, $B_1 = 0.005\,\Omega$, $B_2 = 0.014\,\Omega$, $U_1/k_B = 4000$, $U_2/k_B = 1800$; Fig. 1(b) $T_{c1} = 252$ K, $T_{c2} = 245$ K (directly determined from the experimental data), $A_1 = 0.1\,\Omega$, $A_2 = 1.9\,\Omega$, $R_1 = 3.3\,\Omega$, $R_2 = 1.0\,\Omega$, $E_{g1}/k_B = 1046$ K, $E_{g2}/k_B = 766$ K, $B_1 = 0.0022\,\Omega$, $B_2 = 0.0024\,\Omega$, $U_1/k_B = 20\,000$, $U_2/k_B = 10\,000$; Fig. 1(c) $T_{c1} = 266$ K, $T_{c2} = 207$ K (directly determined from the experimental data), $A_1 = 0.1\,\Omega$, $A_2 = 1.9\,\Omega$, $R_1 = 0.94\,\Omega$, $R_2 = 0.001\,\Omega$, $E_{g1}/k_B = 766$ K, $E_{g2}/k_B = 366$ K, $B_1 = 1.6 \times 10^{-4}\,\Omega$, $B_2 = 1.8 \times 10^{-5}\,\Omega$, $U_1/k_B = 25\,000$, $U_2/k_B = 2500$.

In order to fit the $R(T)$ curves over the whole temperature range, we assumed that near the magnetic transition the sample contains both paramagnetic (PM) and ferromagnetic (FM) regions. Following Ref. [10] we defined f as the volume fraction of FM domains. The corresponding volume fraction of paramagnetic regions is represented by $1-f$. Clearly, $f = 0$ at $T \gg T_c$; $f = 1$ at $T \ll T_c$ and $0 < f < 1$ at intermediate temperatures. An appropriate empirical expression for f is [10]

$$f = \frac{1}{1 + e^{-U(1-T/T_c)/k_B T}}, \tag{1}$$

where U is a temperature independent constant and T_c is the Curie temperature. In the following we will model the resistance of the PM regions as $R_{PM} = R_1 e^{E_g/k_B T}$ (polaronic dependence) and the resistance of the FM domains as $R_{FM} = A + BT^2$ (metallic behavior). The total resistance of our model is well described by

$$\begin{aligned} R(T) = {} & [(1-f_c)R_1 e^{E_{g1}/k_B T} + f_c(A_1 + B_1 T^2)] \\ & + [(1-f_s)R_2 e^{E_{g2}/k_B T} + f_s(A_2 + B_2 T^2)], \end{aligned} \tag{2}$$

where the first two terms describe the contribution of the core of the grains, and the last two contain the contributions from the regions of depleted T_c located around the GB. The volume fraction f depends on T_c, so it must stand for each magnetic phase T_{c1} and T_{c2}. We use the function f_c for the volume fraction of the core of the grains and f_s for the regions of depleted T_c. We have a large number of fitting parameters, but all the terms have a strong and opposite temperature dependence that allows a non-trivial and physically interpretable fitting.

Fig. 1 shows the theoretical curves (straight lines) that fit the experimental results over the whole temperature range, using Eqs. (1) and (2). The fitting parameters are reported in the figure caption. In general, from Fig. 1(a) to (c) R_1, R_2, B_1 and B_2 decrease according to the decreasing sample resistance. The analysis of the fitting parameters (Fig. 1(a)) shows that, the regions of depleted T_c dominate the transport properties and shift T_p from T_c; it seems like the temperature of the MI transition was different from T_c. Fig. 1(b) is an intermediate case where both contributions are comparable and two $R(T)$ maxima appear. In Fig. 1(c) the resistance contribution from the regions of depleted T_c is very small, and only one maximum appears, there is a correlation between the MI and the magnetic transition. The last case corresponds to a good connectivity between the grains that allows the correlation between the magnetic and transport properties.

In summary, we have explained the different MI transitions observed in samples with different grain connectivity in our thick films. We found that the core of the grains, although determine the magnetic properties of the sample, in some cases do not have an appreciable contribution to the transport properties as in Fig. 1(a). We suggest that in samples where T_p is very different from T_c, there are regions of depleted T_c adjacent to the GB that dominate the transport properties and induce an appreciable shift in the maximum of the $R(T)$ curves. When both resistance contributions are comparable, the $R(T)$ curves may have two maxima, as it is shown in Fig. 1(b).

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