



# Extrinsic magnetoresistance in $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ thick films

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## Abstract

Thick films of  $\text{La}_{0.66}\text{Ca}_{0.33}\text{MnO}_3$  have been obtained on alumina substrate by the “paint-on” method. They exhibit a clear extrinsic magnetoresistance (broad metal–insulator transition with peak temperature lower than Curie temperature, magnetoresistance at low temperatures and magnetic fields, etc.). This behavior seems to be related to the competition between tunneling magnetoresistance (TMR) at the grain boundaries and double exchange in the grains. An anomaly is observed in the resistance vs. applied magnetic field at low temperatures, which support the presence of TMR. EDAX analysis performed to the films shows elemental composition deviations near to the grain boundaries. © 2001 Published by Elsevier Science B.V.

**Keywords:** Manganites; Extrinsic magnetoresistance; Tunneling magnetoresistance

The appearance of the colossal magnetoresistance effects in manganites, governed by the double-exchange (DE) mechanism, is restricted to a narrow temperature range (near to the Curie temperature  $T_c$ ) and in a Tesla field range [1,2]. However, in granular samples (polycrystalline, powder and composites [3,4]), some of the following characteristics appear: temperature of metal–insulator (MI) transition ( $T_p$ ) can occur far below  $T_c$ , MI transition is broad, significant magnetoresistance (MR) ratio appears at low magnetic field and it rises with the decrease of temperature. This effect called extrinsic magnetoresistance (EM), [1,5] has been found also in artificial bicrystal junctions [6] and trilayer Junctions [7] and has been related to tunneling magnetoresistance (TMR) in grain boundaries (GB) [7,8]. Sharp changes in junction resistance ( $R$ ) related to moment reversal in electrodes are present. In irradiated films a hysteretic anomaly is reported in  $R$  vs. applied magnetic field ( $R(H)$ ) at about 3 kOe [9].

In this work we have prepared thick films of  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  on alumina substrate by a simple paint on method with the aim to study in detail the

characteristics of EM in this type of polycrystalline samples [10].

Ceramic  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  powder was prepared by standard solid-state reaction techniques, starting from the appropriate mixture of oxides. For more detail see Ref. [10]. The average thickness and grain size of the film was about 16 and 5  $\mu\text{m}$  respectively as estimated from electron microscopy measurements. EDAX analysis shows composition deviations in the grain boundaries region. XRD analysis of the film revealed a single crystalline phase. The transport measurements have been carried out using a standard four-probe DC method. The magnetic field (up to 10 kOe) is applied in the film plane. The DC magnetic susceptibility was measured using a SQUID magnetometer. The magnetization vs. temperature curves were obtained in zero-field cooled conditions. The resistance vs. applied magnetic field at low temperature was calculated using the  $R(T)$  curves measured at different values of magnetic fields.

In Fig. 1 are summarized the principal and typical magnetic and transport properties of our films. The magnetization vs. temperature curve was taking at 0.1 kOe. Typical EM is present [1]. The sample shows broad IM transition with  $T_p$  60 K below  $T_c$ . The little magnetic anomaly (a maximum) at 30 K is believed to be related to the residual  $\text{MnO}_2$  [12]. The shallow minimum in  $R(T)$  at 30 K has been reported in other works and considered

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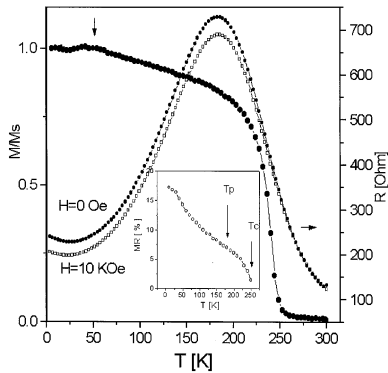


Fig. 1. Transport and magnetic properties of the film. The vertical arrow shows the magnetic anomaly with a maximum at 30 K. The inset shows the magnetoresistance at low temperature.

reminiscent of Kondo effects [11]. The inset shows a clear rise of MR ratio with the decrease of temperature. This dependence is very similar to the theoretically obtained for TMR in manganites [13] but with a different curvature between  $T_c$  and  $T_p$ . This fact could be associated to the DE contribution (for DE, MR peak is close to  $T_c$  [1]). The high magnetization (3.5 Bohr magnetons by Mn atom at 4.2 K) and the sharpness of magnetic transition of our samples, allow us to suppose that the core of the grains have  $T_c \approx 240$  K. However the EDAX results indicate that we can have small regions with depressed  $T_c = T_c \sim T_p$  near to GB. Broadness of the MI transition and the fact that  $T_p \ll T_c$  could be explained taking into account the competition between DE mechanism and TMR in the grain boundaries. DE occurs in the core of the grains and in zones with  $T_c$  below  $T_p$ . DE and TMR mechanism provides opposite trends in resistance versus temperature below  $T_c$  [1,13]. We think that the minimum observed in  $R(T)$  at 30 K, could be related to the fact that at low temperatures the TMR play the determinant role. Other important evidence of TMR in our samples is the presence of an anomaly in the  $R(H)$  dependence at low temperatures as it is shown in Fig. 2, it disappears at higher temperatures [7] (see inset of Fig. 2). This anomaly, is very similar to the reported for irradiated films [9], and it is also centered at 3 kOe. In this reference this anomaly was taken as a strong evidence that the microscopic spin disorder alone is able to induce similar behavior than that provided by TMR. The high magnetization of our samples must rule out this possibility.

In summary monophasic thick films of  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  have been obtained on alumina substrates, which show a clear extrinsic magnetoresistance behavior. The competition between double exchange mechanism in the core of the grains, the tunneling magnetoresistance in the grain boundaries and double exchange (shifted to lower temperatures) of the small zones in the grain surfaces, can

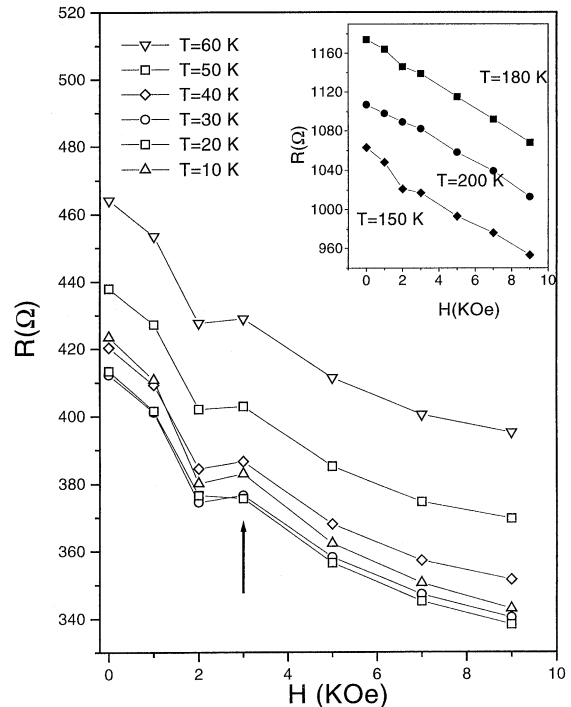


Fig. 2. Resistance versus applied magnetic field at different temperatures. The arrow shows an anomaly at 3 kOe that disappears at high temperatures (see inset).

explain the observed extrinsic magnetoresistance in our samples. The minimum shown at 30 K in the resistance vs temperature dependence, could be an indication that it is determined by tunneling magnetoresistance at low temperatures and not by Kondo-like effects. The anomaly observed in the resistance vs. applied magnetic field dependence at low temperatures, reinforce this hypothesis.

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