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Conduction and disorder in $LaMnO_3$-based materials

N G Bebenin[†] and V V Ustinov

Institute of Metal Physics, Ural Division of the Russian Academy of Sciences, GSP-170, Ekaterinburg 620219, Russia

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Abstract. We propose a simple theory of colossal magnetoresistance in which the metal–insulator transition in heavily doped lanthanum manganites is assumed to be caused by a shift of the mobility edge. The theory involves a small set of parameters which have clear physical meaning and can be easily found experimentally. The detailed comparison with experimental data available is made.

1. Introduction

The discovery of very large magnetoresistance in lanthanum manganites $La_{1-x}A_xMnO_3$, where A = Ca, Ba, Sr or vacancy, has attracted renewed interest to this class of materials. To distinguish this effect from the giant magnetoresistance in metallic multilayers, which is also intensively investigated, the magnetoresistance in the manganites is called 'colossal' magnetoresistance (CMR).

The parent material $LaMnO_3$ is an antiferromagnetic insulator. Doping with divalent ions or vacancies results in an increase of conductivity, onset of spontaneous magnetization and the CMR near the Curie temperature T_C. A great number of experiments on polycrystals, single crystals and thin films has been carried out to reveal the CMR dependence on temperature and magnetic field as well as on composition, details of sample preparation, etc, see recent reviews [1, 2]. It has been shown that the CMR is observed if conductivity of the sample is metallic below T_C and semiconductor-like in the paramagnetic region; in other words, the metal–insulator transition near T_C must occur for the CMR effect to exist.

The origin of the CMR remains unclear although the literature on the manganites is dominated by discussion on this subject. Traditionally the charge transfer in $LaMnO_3$-based compounds is considered on the basis of the Zener double exchange model [3]; this model is now being modified in order to take into account polaron effects, see for example [4]. Some authors pay attention to the fact that large magnetoresistance is typical to europium chalcogenides and interpret the results of measurements in terms of magnetic semiconductor physics [1, 5, 6]. It is to be noted that analysis of experiments is restricted everywhere by qualitative consideration without numerical estimations, so that what concept is more realistic is difficult to decide. Perhaps, it is partly due to the fact that the theoretical works remain very formal and their results cannot be used directly for interpretation of transport measurements.

[†] Corresponding author: N G Bebenin, Institute of Metal Physics, Ural Division of the Russian Academy of Sciences, 18, S Kovalevskaya Street, GSP-170, Ekaterinburg 620219, Russia. E-mail address: elph@ifm.e-burg.su

Under these circumstances any simple insights into the nature of conduction in the lanthanum manganites should help in establishing contact between theory and experiment. In this paper we propose a simple phenomenological theory based on the assumption that the metal–insulator transition in the heavily doped manganites is caused by motion of the mobility edge. The theory involves a small set of parameters which have clear physical meaning and on the other hand can be easily found experimentally.

2. Mobility edge

The lanthanum manganites are known to be ferromagnets if $0.1 \leqslant x \leqslant 0.6$ [1]. We shall deal with $x < 0.5$ and consider $\mathrm{La}_{1-x}\mathrm{A}_x\mathrm{MnO}_3$, as a p-type ferromagnetic semiconductor. The band calculations show that the valence band in the compounds under study is formed mainly by $\mathrm{e}_g^{(1)}$ states of Mn^{3+}. The band is narrow: the band width W is about 1–1.5 eV which value is markedly less than Hund's rule coupling [7]. The level of doping is high; interaction of charge carriers with magnetic fluctuations as well as with phonons is strong. All this implies that disorder plays an essential role in the formation of transport properties of the manganites, so that to use the concepts of the physics of disorder [8–10] is quite natural.

The disorder created by impurity atoms and vacancies can be considered as not depending on temperature whereas the disorder induced by lattice vibrations and the magnetic fluctuations is temperature dependent. Hence, the position of the mobility edge of holes, i.e. the energy ε_c separating localized states from extended ones, changes as temperature T is changed. We will suppose that the temperature dependence of ε_c is controlled mainly by the magnetic fluctuations (this supposition will be testified below by comparison with experiment). For a narrow band ferromagnetic semiconductor the temperature dependence of ε_c was calculated in [11] within the framework of the s–d model with special application to n-$\mathrm{CdCr_2Se_4}$, the compound that seems to be most similar to the lanthanum manganites among the family of ferromagnetic semiconductors. The ordinary s–d model is inapplicable to $\mathrm{LaMaO_3}$-based materials, so that the formulae obtained in [11] cannot be applied directly. Nevertheless the results of [11] together with the well known general concepts of the physics of disorder enable us to understand how the properties of $\mathrm{LaMnO_3}$-based compounds depend on temperature and doping.

At $T = 0$ the disorder enters through impurity atoms and lattice defects only. As temperature is increased, the disorder is enhanced and thus the mobility edge is shifted to higher energies. The shift is likely to be sharpest near the phase transition point because of the sharp increase of magnetic fluctuations. We may suppose that in the close vicinity of T_C the characteristic energy ε_c is scaled by the magnetic part of the energy, so that

$$\frac{\mathrm{d}\varepsilon_c}{\mathrm{d}T} \propto C \tag{1}$$

where C is the magnetic part of the specific heat. In the paramagnetic region, ε_c is determined by the short range order and therefore the ordinary high temperature expansion of the form $\varepsilon_c(T) = \Delta_0 + \mathrm{constant}/T + \cdots$ has to be valid.

As is shown in [11], in a narrow band ferromagnet $\varepsilon_c(T \gg T_C) - \varepsilon_c(T = 0) = \gamma W$ with $\gamma = 1/4$ if the nonmagnetic disorder is absent. This result is quite clear: in a narrow band material W is the smallest parameter of the dimension of energy, therefore an expansion of ε_c in powers of W must contain γW as the first term. The large value of γ points out that the shift of the mobility edge can be significant.

Magnetic field lowers the mobility edge due to reduction of magnetic disorder. We may write for $T \gg T_C$

$$\varepsilon_c = \Delta_0 - \Delta_1 m^2 \tag{2}$$

where m is relative (with respect to saturation value) magnetization, Δ_1 is a positive constant just like Δ_0. Our basic assumption is that the relation (2) is fulfilled at any temperatures; in other words, the temperature and magnetic field dependence of ε_c enters only through $m(T, H)$, H being applied magnetic field strength. This simple approximation is consistent with the qualitative description of the temperature dependence of ε_c given above except that in zero magnetic field the mobility edge reaches its maximum already at the Curie temperature. Moreover, if magnetic properties are described in the framework of Landau theory, equation (1) is also valid.

3. Resistivity

In this section we apply the relation (2) to the calculation of resistivity. We assume that the change of resistivity is determined mainly by the concentration of holes in the extended states:

$$n_{ext} = \int_{\varepsilon_c}^{\infty} g(\varepsilon) F(\varepsilon)\, \mathrm{d}\varepsilon \tag{3}$$

where $g(\varepsilon)$ is the density of states of holes, $F(\varepsilon)$ is the Fermi distribution function. As is well known, the type of conduction is controlled by the relation between ε_c and hole Fermi energy ε_F: if $\varepsilon_c < \varepsilon_F$, conduction is metallic, otherwise conduction is due to hops between localized states and activation to the mobility edge. The hopping conductivity has been reported in many papers, see e.g. [12, 13] and references therein, but to consider this type of conduction one needs information about the nature of localized states which is absent so far; thus in the present work we assume that at high temperatures the activation to the mobility edge dominates. Then the resistivity can be expressed as

$$\rho = \rho_0\, \mathrm{e}^{\varepsilon_c - \varepsilon_F / T} \tag{4}$$

where ρ_0 weakly depends on temperature and magnetic field.

For the metal–insulator transition to take place, the Fermi energy must exceed ε_c at low temperatures and be less than ε_c in the paramagnetic region, as shown in figure 1. In the following, unless stated explicitly otherwise, we shall deal with this situation. It follows from (2) that the temperature T_{MI} of the metal–insulator transition, i.e. the temperature at which $\varepsilon_c = \varepsilon_F$, obeys the equation

$$m^2(T_{MI}, H) = m_{MI}^2 = \frac{E_{a0}}{\Delta_1} \tag{5}$$

where $E_{a0} = \Delta_0 - \Delta_F$ is the activation energy at $T > T_C$ in zero magnetic field. Obviously, the temperature of the metal–insulator transition is less than T_C and $E_{a0} < \Delta_1$. In the frame of the Landau theory, near T_C the magnetization obeys the equation

$$H = \alpha_0 \frac{T - T_C}{T_C} m + \beta m^3 \tag{6}$$

where α_0 and β are constants. It follows from (5) and (6)

$$T_{MI} = T_C \left(1 + \frac{H - \beta m_{MI}^3}{a_0 m_{MI}} \right). \tag{7}$$

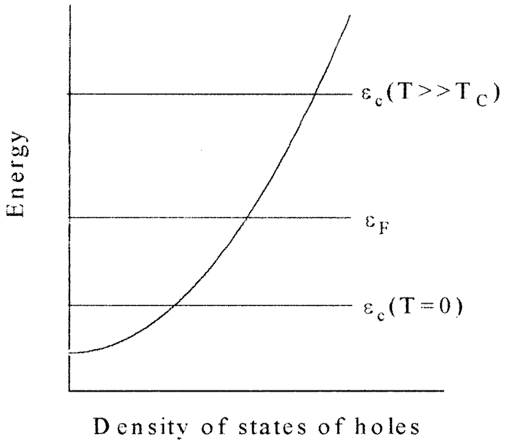

Figure 1. Fermi level and mobility edge at $T = 0$ and $T \gg T_C$ in the samples with largest magnetoresistance.

The second term in the parenthesis is assumed to be much less than unity. Since m_{MI} is fixed, a magnetic field shifts the metal–insulator transition point to higher temperatures, T_{MI} being a linear function of H.

Of particular interest is the dependence of resistivity on magnetic field and temperature. Let us first consider the H-dependence of ρ at a given temperature. As the resistivity depends on H only through m^2, in weak magnetic fields the magnetoresistance $r = [\rho(H) - \rho(0)]/\rho(0)$ is proportional to H in the ferromagnetic and $r \propto H^2$ in the paramagnetic state. At the phase transition point $r \propto H^{2/3}$. If the conductivity is determined by activation to the mobility edge, we obtain that in the weak field

$$r = \frac{\Delta_1}{T}(m^2(0) - m^2(H)). \tag{8}$$

Thus one can use magnetoresistance measurements to evaluate Δ_1.

The largest change of resistivity can be achieved at $T = T_C$ if application of a (sufficiently strong) magnetic field transfers a sample from the insulating into the metallic state. It is easy to show that ρ_0 in (4) is of the order of the resistivity in the metallic state (in the next section this will be also confirmed by comparison with experiment). Using this fact, we obtain a simple estimation for the maximum value of ratio $\rho(0)/\rho(H)$:

$$\left(\frac{\rho(0)}{\rho(H)}\right)_{max} \approx \exp(E_{a0}/T_C) \tag{9}$$

so that the maximum attainable value of magnetoresistance can be estimated as

$$|r|_{max} \approx 1 - \exp(-E_{a0}/T_C). \tag{10}$$

Let us now discuss the temperature dependence of ρ at a fixed H. Remembering equation (1), one may think that in the zero field the derivative $\partial\rho/\partial T$ has a maximum at $T = T_C$, as in an ordinary ferromagnetic metal [14–16]. However, from equations (4) and (6) we find that at $H = 0$ the resistivity has a maximum at the Curie point if $\Delta_1/E_{a0} > \beta/\alpha_0$, $\partial\rho/\partial T$ being discontinuous at $T = T_C$. The peak of the resistivity and the discontinuity of $\partial\rho/\partial T$ at $T = T_C$ are a consequence of the roughness of our approximation (2).

Magnetic field eliminates the discontinuity of $\partial\rho/\partial T$ and shifts the peak to higher temperatures. To find the temperature T_R, at which ρ is maximum, let us use equation (6) and rewrite the argument of the exponential function in (3) as

$$\frac{\varepsilon_c - \varepsilon_F}{T} = \frac{E_{a0} - \Delta_1 m^2}{T_C(1 + (H - \beta m^3)/\alpha_0 m)}. \tag{11}$$

If H is small, the function in right-hand side of equation (11) has a maximum when

$$m^3 = \frac{H}{2\alpha_0(\Delta_1/E_{a0} - \beta/\alpha_0)}. \tag{12}$$

Here and below we assume the inequality $\Delta_1/E_{a0} > \beta/\alpha_0$ to be fulfilled. Expressing T as a function of m in accordance with (6), we easily obtain

$$T_R(H) = T_C + \text{constant } H^{2/3}. \tag{13}$$

4. Comparison with experiment

Some results have been published recently which confirm that there is strong disorder in the CMR compounds and these manganites in the paramagnetic state are similar to amorphous rather than crystalline materials. Thus the large disorder effect due to size differences between A-site R^{3+} (R = La, Pr, Nd, Sm) and M^{2+} (M = Ca, Sr, Ba) cations have been found in magnetoresistive $(R_{0.7}M_{0.3})MnO_3$ perovskites [17]. The results of x-ray-absorption fine-structure measurements on $La_{1-x}Ca_xMnO_3$ and $La_{0.67}Pb_{0.33}MnO_3$ reported in [18] strongly suggest that charge carriers delocalize as the magnetization increases in materials which exhibit a metal–insulator transition. The thermal conductivity κ of manganite perovskites has been measured in [19]. In the metallic state, κ has been found to behave as expected for a crystalline solid whereas in the high temperature insulating phase $d\kappa/dT > 0$, the behaviour of an amorphous solid. Thus we may state that our assumption about the principal role of disorder in the CMR effect agrees with the experiments.

Let us turn to magnetoresistance. That r is directly proportional to H in the ferromagnetic state has been reported in [20] for an $La_{2/3}Sr_{1/3}MnO_3$ single crystal and in [21] for an epitaxial thin film $La_{2/3}Ca_{1/3}MnO_3$. The H^2-dependence of magnetoresistance in the CMR materials in the paramagnetic region can be easily derived from the results published, for example, for the $La_{1-x}Sr_xMnO_3$ single crystals in [22], for $La_{0.67}Ca_{0.33}MnO_3$ epitaxial film in [21], for epitaxial thin films of $LaMnO_3$ doped with vacancies in [23] and other works.

Although ρ versus T for some values of H are reported in almost every article on the CMR, only a few papers, the works [21–23] being among them, give information which is sufficiently complete to test the relation (13). Figure 2 shows T_R, evaluated from the data reported in [21–23], as a function of $H^{2/3}$ for some samples. One can see that experimental points really do obey the $H^{2/3}$-law.

In the work by Urushibara *et al* [22], the measurements of resistivity were carried out on single crystals of $La_{1-x}Sr_xMnO_3$ along with the measurements of magnetic properties. The information given in that paper enables us to perform the detailed comparison between the theory and experiment.

The main features of ρ–T curves found in [22] are the following. A conspicuous change in ρ is observed at around T_C ($x \geqslant 0.1$). In the paramagnetic region, the temperature dependence of the resistivity is approximately exponential if $0.1 \leqslant x \leqslant 0.2$, the activation energy decreasing with increasing x. In the low-temperature ferromagnetic state, metallic conduction is observed above $x = 0.175$; by contrast, the resistivity at $x = 0.1$ and $x = 0.15$

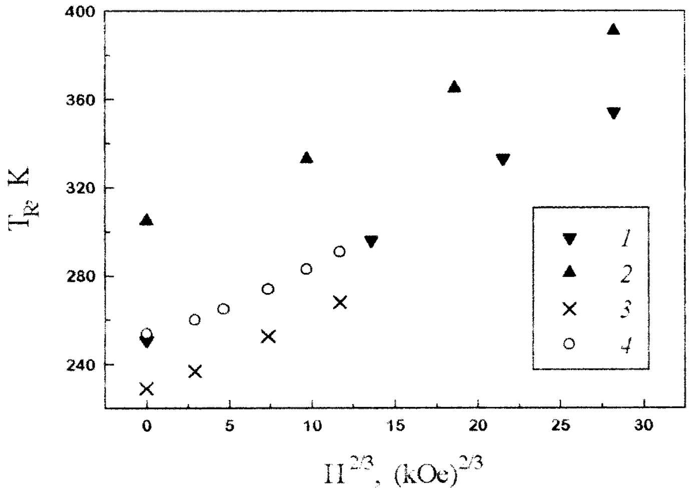

Figure 2. Position of the resistivity peak versus $H^{2/3}$ for: 1, single crystal of $La_{0.85}Sr_{0.15}MnO_3$ [22]; 2, single crystal of $La_{0.825}Sr_{0.175}MnO_3$ [22]; 3, epitaxial thin film of $La_{2/3}Ca_{1/3}MnO_3$ [21]; 4, $La_{0.75}\square_{0.25}MnO_3$ where $\square$ is vacancy [23].

first decreases with decreasing temperature immediately below T_C but then increases. The slope of the curves sharply changes somewhat below T_C approximately at 210 K, 260 K and 275 K for the $x = 0.15$, $x = 0.175$ and $x = 0.2$ samples, respectively. The distinct change of slope is seen also on the ρ–T curves for the $x = 0.3$ and $x = 0.4$ samples although these samples are in the metallic state both above and below T_C.

Large negative magnetoresistance is observed in the vicinity of the phase transition point at $x = 0.15$, $x = 0.175$ and $x = 0.2$. All three samples demonstrate similar temperature and magnetic field dependences of ρ and r but the detailed data are reported for the $x = 0.15$ and $x = 0.175$ samples only.

Let us compare the estimation of the maximum magnetoresistance value given by formula (9) with the experiment. We have to know T_C and E_{a0}. In [22] the Curie temperature was determined by ac measurements; in what follows, the value of T_C obtained by this method will be denoted by T_C^{ac}. It was found that $T_C^{ac} = 238$ K at $x = 0.15$ and $T_C^{ac} = 283$ K at $x = 0.175$. For our purpose, however, T_C must be evaluated though Arrott–Belov curves because the magnetization curves are taken to obey the equation (6); the value found by this method will be referred to as T_C^{AB}. Using the magnetization curves given in [22] we have calculated T_C^{AB} and found $T_C^{AB} \approx 256$ K and $T_C^{AB} \approx 292$ K for the $x = 0.15$ and $x = 0.175$ samples, respectively. Notice that the values of T_C^{AB} are higher than T_C^{ac} especially for the $x = 0.15$ sample in which case the difference $T_C^{AB} - T_C^{ac}$ is as large as 18 K.

The temperature dependence of ρ of the $x = 0.15$ sample in the paramagnetic region can be described well by the relation $\rho = \rho_0 \exp(E_{a0}/T)$ with $\rho_0 = 0.07\ \Omega$ cm and $E_{a0} = 720\text{ K} = 0.062$ eV. The value of ρ_0 is practically equal to the resistivity of the sample at 210 K where the slope of the ρ–T curve sharply changes. Inserting T_C^{AB} and E_{a0} into (10), we obtain the estimation: $|r|_{max} = 0.94$ which value is very close to the maximum experimental one: $|r| = 0.95$ at $H = 150$ kOe.

Let us find the parameter Δ_1 which plays the essential role in the present theory. In our case, equation (8), which in principle could be exploited, is useless because the magnetoresistance data of [22] refer to $H = 150$ kOe only. One can use, however, $\rho(H)$ and $m(H)$ curves measured at one and the same temperature if T belongs to the region where conductivity is due to activation to the mobility edge. It follows from (2) and (4) that in such a case

$$\ln \rho = \ln \rho_0 + \frac{E_{a0}}{T} - \frac{\Delta_1}{T} m^2. \tag{14}$$

The $\ln \rho$ versus m^2 curve at $T = 254$ K is plotted in figure 3 for the $x = 0.15$ sample. The curve is indeed a straight line and can be described by the equation: $\ln \rho = 4.38 - 5.06\ \mathrm{m}^2$, so that we obtain $\Delta_1 = 5.06$, $T \approx 1300$ K. The data for $T = 274$ K and $T = 303$ K give practically the same value.

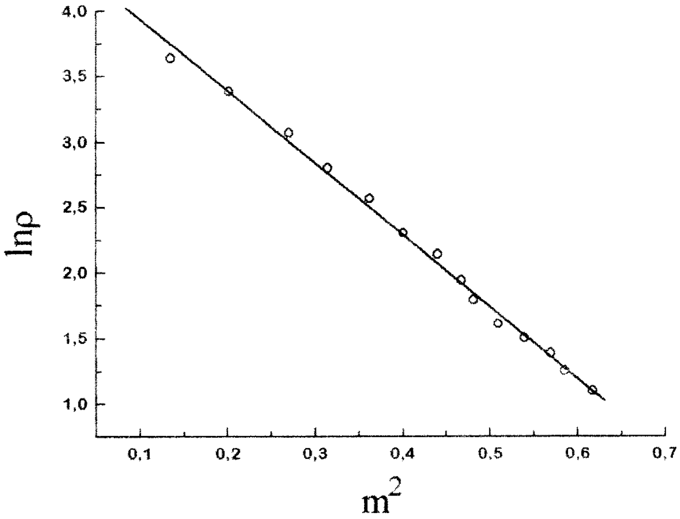

Figure 3. $\ln \rho$ versus m^2 for the $x = 0.15$ sample at $T = 254$ K. The points have been calculated from the ρ–H and m–H curves presented in [22].

Now we are able to calculate the temperature dependence of the resistivity of the $x = 0.15$ sample at a fixed magnetic field near T_C and in the paramagnetic state where the change of ρ is assumed to be caused by the change of concentration of holes in extended states. Equation (3) shows that we have to know the density of states $g(\varepsilon)$, the Fermi energy of holes, ε_F, Δ_0 (or $E_{a0} = \Delta_0 - \varepsilon_F$), and Δ_1; also we need the temperature dependence of magnetization at a given H. The true form of $g(\varepsilon)$ is unknown. The level of doping of the sample suggests, however, that the Fermi level lies inside the valence band far enough from the band edge and the density-of-states tail in the energy gap and, on the other hand, far from the middle of the band; this allows us to choose $g(\varepsilon)$ in the simplest form, namely, $g(\varepsilon) \propto \varepsilon^{1/2}$. Taking into account that the bandwidth is about 1–1.5 eV, we set $\varepsilon_F = 3000$ K; it is worth noting that the results of calculations weakly depend on the value of ε_F. The values of E_{a0} and Δ_1 are given above.

The temperature dependence of m was calculated according to (6) with $\alpha_0 = 1130$ kOe, $\beta = 295$ kOe and $T_C = T_C^{AB} = 255$ K, ρ_0 and β being determined on base of the magnetization curves given in [22].

Figure 4 shows the results of calculations (dashed lines) and the experimental curves from [22] (solid lines). The theoretical curves are close to the experimental ones, but if $H = 0$ the agreement is not so good as at $H \neq 0$. This is likely to indicate that the temperature dependence of Δ_0, which is completely ignored in the calculations, in fact exists and results, for example, in the smearing of the resistivity peak at $H = 0$ while in the presence of sufficiently strong magnetic field the temperature dependence of the mobility edge is determined mainly by the m^2-term in (2).

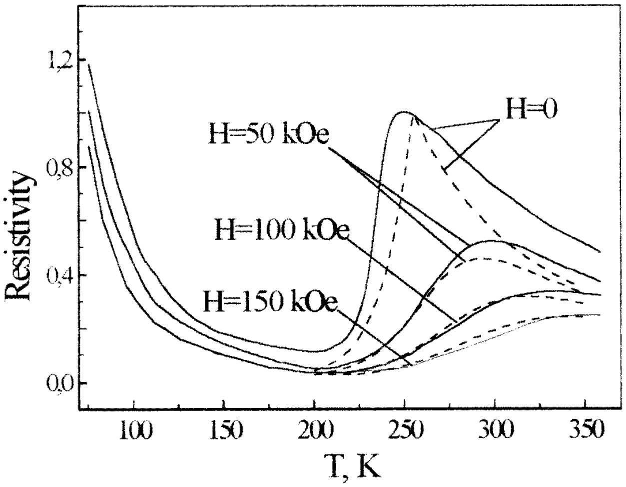

Figure 4. Experimental (solid lines) and theoretical (dashed lines) ρ–T curves normalized to the peak value of resistivity for the $x = 0.15$ sample. The experimental data are from [22].

5. Concluding remarks

In our theory we presume a sample to be homogeneous. This means in particular that the theory is inapplicable to polycrystals. It is important however that even a single crystal can be in an inhomogeneous state. Indeed, the temperature dependence of magnetization of the $Ln_{1-x}Sr_xMnO_3$ single crystals taken at $H = 1$ kOe in [24] is not typical for a homogeneous ferromagnet if $x = 0.175$ or lower; moreover, two $x = 0.1$ samples extracted from the same single crystal turn out to exhibit very different temperature dependences of magnetization. In contrast, the curve for $x = 0.25$ is quite smooth. The inhomogeneity of $Ln_{1-x}Sr_xMnO_3$ is likely to be essential if x is less than a percolation threshold x_{th} whose value is about 0.15 [8]. Perhaps, the large difference between T_C^{ac} and T_C^{AB} for the $x = 0.15$ sample also indicates that the sample is magnetically inhomogeneous. We may suppose that in the ferromagnetic state far enough the Curie point the $x = 0.15$ sample decomposes into domains of high (metallic) conductivity connected by the small 'bridges' of semiconductor phase, and it is these 'bridges' that determine the increase of resistivity with decreasing temperature at $T < T_C$. The same must be true also at $x = 0.1$. For the $x = 0.175$ sample reported in [22] the difference $T_C^{AB} - T_C^{ac}$, found above, is less than 10 K; in addition the temperature dependence of magnetization at $H = 1$ kOe, given in [24] for a similar single crystal, is not so distorted as at $x = 0.15$, therefore we conclude that at $x = 0.175$ the material is magnetically more homogeneous than at $x = 0.1$. This correlates with $x > x_{th}$ and the metallic conductivity in the ferromagnetic state. The single crystal samples at $x > 0.175$ are likely to be more or less homogeneous at any temperature.

To summarize, many experimental facts concerning the $LaMnO_3$-based materials with a high level of doping ($x = 0.15$ or more) can be successfully explained in the frame of our approach. It is unlikely to be a simple coincidence, therefore we may state that the colossal magnetoresistance in heavily doped samples of $La_{1-x}A_xMnO_3$, results from the shift of the mobility edge induced by change of temperature or application of a magnetic field.

Acknowledgments

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