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Current–Voltage Characteristics of Polycrystalline $(La_{0.5}Eu_{0.5})_{0.7}Pb_{0.3}MnO_3$ at Low Temperatures

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Abstract—The current–voltage characteristics of the polycrystalline substituted lanthanum manganite $(La_{0.5}Eu_{0.5})_{0.7}Pb_{0.3}MnO_3$ have been measured at temperatures close to the metal–insulator transition temperature and at low temperatures. In both cases, the current–voltage characteristics exhibit nonlinear properties that are strongly dependent on the strength of an applied magnetic field. The mechanisms responsible for the nonlinear properties at these temperatures are found to be different: near the metal–insulator transition, the current–voltage characteristics are determined by the phase layering inside granules, while at low temperatures, they are determined by tunneling of carriers through insulating interlayers of the granules.

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The current–voltage characteristics of substituted lanthanum manganites $R_{1-x}A_xMnO_3$ (R stands for trivalent rare-earth ions La^{3+}, Nd^{3+}, Pr^{3+}, Sm^{3+}, etc.; A is a divalent ion Ca^{2+}, Sr^{2+}, Ba^{2+}, or Pb^{2+}) have been studied in detail by many authors [1–9]. A main specific feature of the current–voltage characteristics of $R_{1-x}A_xMnO_3$ compounds can be suggested to be their nonlinear properties strongly that are dependent on the strength of an external magnetic field. For example, the nonlinearities of the current–voltage characteristics of single-crystal manganites are usually explained by assuming that a crystal is in a heterogeneous state of magnetic phase layering when the phases with different conductivities coexist in the sample and an external action, such as the magnetic field, changes the volume ratio between the phases [2, 3, 10]. In this case, main specific features of the current–voltage characteristics manifest themselves at temperatures close to the metal–insulator transition temperature that almost coincides with the Curie temperature of the single crystals. This is due to the fact that the phase layering into ferromagnetic conducting regions and antiferromagnetic insulating regions is manifested to a highest degree in this temperature range, and, as a result, the magnetoresistance of the sample is maximal.

At the temperatures close to the metal–insulator transition temperature, the current–voltage characteristics of $R_{1-x}A_xMnO_3$ polycrystals are also nonlinear and sensitive to the strength of an external magnetic field [3], which is due to the phase layering. However, in the case of polycrystalline substituted lanthanum manganites, in which the main state is the ferromagnetic metal state, the current–voltage characteristics demonstrate a significant magnetoresistive effect and a nonlinearity at temperatures that are substantially lower than the metal–insulator transition temperature. In this case, the observed effect can be due to spin-dependent tunneling of charge carriers between ferromagnetic granules through insulating intercrystallite boundaries. The low-temperature magnetoresistance is first dependent on the extent of the intercrystallite boundaries and their origin [11, 12]. For example, as shown in [13], in polycrystalline $(La_{0.5}Eu_{0.5})_{0.7}Pb_{0.3}MnO_3$, the intercrystallite boundaries are antiferromagnetic insulators, and it make additional contribution to the low-temperature magnetoresistance because of the exchange interaction of the antiferromagnetic interlayer with nearest ferromagnetic domains of granules. Undoubtedly, the high low-temperature magnetoresistance comparable with that near the metal–insulator transition must influence the current–voltage characteristics at low temperatures. Thus, the goal of this work is to study the current–voltage characteristics of polycrystalline $(La_{0.5}Eu_{0.5})_{0.7}Pb_{0.3}MnO_3$ in the temperature ranges where the magnetoresistive effect is maximal: at low temperatures and near the metal–insulator transition. The measuring current was chosen in such a way as to prevent internal heating of the carriers and appearance of segments with negative differential resistance in the current–voltage characteristics [1].

We chose this compound, since single-crystal samples of $(La_{1-x}Eu_x)_{0.7}Pb_{0.3}MnO_3$ ($x = 0$, 0.2, 0.4, 0.6) were already characterized and studied, and the sample with $x = 0.5$ was found to be on the verge of the

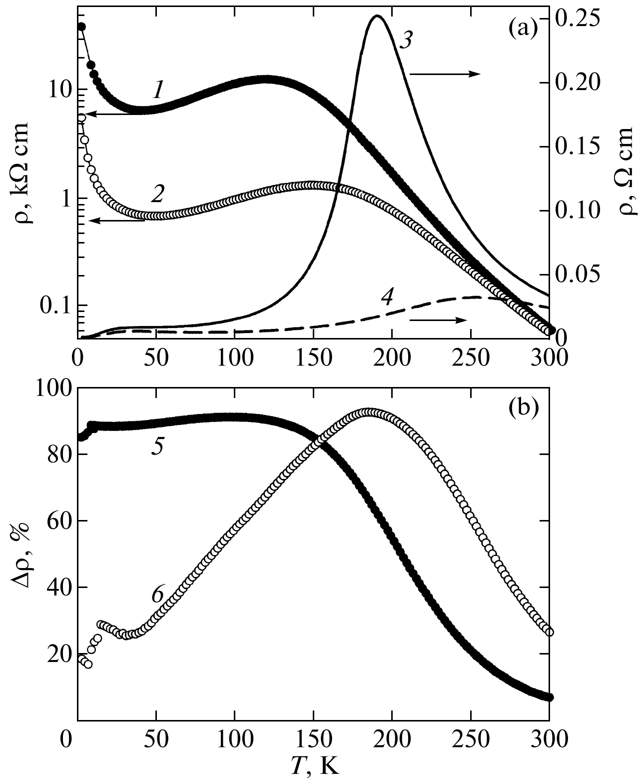

Fig. 1. Temperature dependences of (a) the resistivity ρ measured at H = (*1*, *3*) 0 and (*2*, *4*) 90 kOe and (b) (*5*, *6*) the relative magnetoresistance Δρ = {ρ(H = 0) − ρ(H = 90 kOe)}/ρ(H = 0) for (*1*, *2*, *5*) polycrystalline and (*3*, *4*, *6*) single-crystal $(La_{0.5}Eu_{0.5})_{0.7}Pb_{0.3}MnO_3$.

substitution series where the metal–insulator transition still occurs; in this case, the magnetoresistive effect was maximal [2, 13–15]. It is this sample that we chose to measure its current–voltage characteristics.

In order to prepare polycrystalline $(La_{0.5}Eu_{0.5})_{0.7}Pb_{0.3}MnO_3$, single-crystal samples synthesized earlier were grinded in an agate mortar and pressed into pellets that were annealed in a furnace at a temperature of 873 K for 6 h. The pellets have a sufficient strength to perform magnetoresistive measurements. Scanning electron microscopy shows that the granule mean size is 1–2 μm. High-resolution transmission electron microscopy reveals that the inner part of the granule is the $(La_{0.5}Eu_{0.5})_{0.7}Pb_{0.3}MnO_3$ single crystal, and the granule outer shell has a thickness of 5 nm. The magnetotransport properties of single- and polycrystalline $(La_{0.5}Eu_{0.5})_{0.7}Pb_{0.3}MnO_3$ were studied in [1, 13, 5].

The transport measurements were performed by the standard four-probe method on a PPMS Quantum Design setup. The contacts were deposited by using a silver-epoxy paste. The sample sizes were ~0.5 × 0.5 mm, and the distance between potential contacts was ~0.1 mm. The temperature dependence of the resistivity ρ(T) was measured at a stable current. The current–voltage characteristics were measured at a given current. We used the apparatus current (to ~10^{-5} A) at which there was no heating and the forward and backward runs of the current–voltage characteristics coincided. The maximum specific power was lower than 0.5 W/cm^3.

Figure 1a depicts the temperature dependences of the resistivity ρ(T) of the polycrystalline (the left axis, logarithmic scale) and single-crystal (the right axis) $(La_{0.5}Eu_{0.5})_{0.7}Pb_{0.3}MnO_3$ sample in a zero external field and at H = 90 kOe. In the single crystal, the metal–insulator transition occurs in the vicinity of T ≈ 185 K. It is known that, in polycrystalline lanthanum manganites, the metal–insulator transition temperature decreases because of existence of intergranule boundaries [11, 12]. The decrease is manifested also in the samples under study. From Fig.1a we notice that the metal–insulator transition temperature of the polycrystal is shifted, and it is ~120 K. In this case, the Curie temperature obtained from the magnetic measurements is the same for the single-crystal and polycrystalline samples, and it is ≈225 K [15].

We also see that ρ of the polycrystal is several orders higher than those for the single crystals. Moreover, the ρ(T) dependence has a minimum in the vicinity of T ≈ 40 K, and, at low temperatures, the resistance significantly increases as temperature decreases. The significant increase of the resistivity of the polycrystal as compared to the single-crystal samples even at room temperature is likely due to influence of the intergranule boundaries. This fact is very clearly pronounced at low temperatures where the ρ(T) dependence is determined by tunneling through intergranular insulating interlayers with antiferromagnetic ordering [13]. It is apparent that the minimum of ρ(T) reflects the competition of two mechanisms, namely: a decrease in ρ of the granules themselves of the manganite (through the intergranular interlayers). As seen, the application of the external field decreases the resistance near the metal–insulator transition and at low temperatures.

Note that the ρ(T) dependence of the polycrystal does not follow the activation and hopping dependence ρ(T) ~ exp(T^{-n}) (n = 0.25–1.0) [16], which can be a result of the nonlinear shape of the current–voltage characteristics (see in what follows).

The temperature dependences of the relative magnetoresistance Δρ = {ρ(H = 0) − ρ(H)}/ρ(H = 0) obtained from the data of Fig. 1a are presented in Fig. 1b. Δρ of the single crystal exhibits the maximum near the metal–insulator transition (Δρ(185 K) ≈ 92%) and becomes insignificant at low temperatures, while Δρ of the polycrystal has almost the same value (≈92%) over entire region below the metal–insulator transition temperature.

The tunneling character of the conduction in the polycrystal is confirmed by the shape of the current–voltage characteristics. Figure 2 shows the current–voltage characteristics of the polycrystalline sample

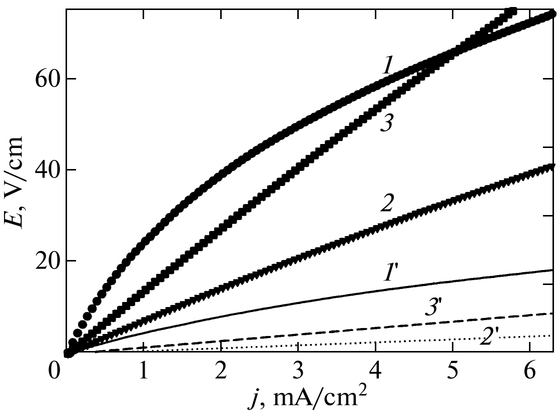

Fig. 2. Current–voltage characteristics of the polycrystalline sample measured at $H = (1–3)$ 0 and $(1'–3')$ 90 kOe at $T = (1, 1')$ 2, $(2, 2')$ 20, and $(3, 3')$ 120 K.

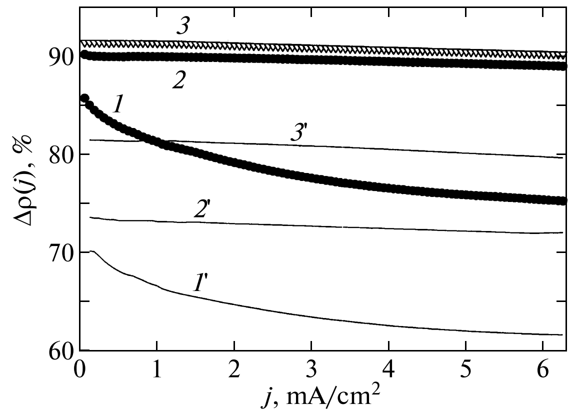

Fig. 3. Dependences $\Delta\rho(j) = \{\rho(H = 0, j) - E(H, j)\}/E(H = 0, j)$ of the polycrystalline sample obtained at $H = (1–3)$ 90 and $(1'–3')$ 50 kOe at $T = (1, 1')$ 2, $(2, 2')$ 20, and $(3, 3')$ 120 K.

measured at $T = 2$, 20, and 120 K in a zero field and at $H = 90$ kOe. A nonlinear current–voltage characteristics are clearly observed for all data presented in this figure at $H = 0$. In external field $H = 90$ kOe at the current densities to ~6.5 mA/cm^2, the current–voltage characteristics become linear for $T = 20$ and 120 K, and it takes place over entire temperature range above 20 K. It is natural that the largest nonlinearity of the current–voltage characteristics (deviation from the Ohm law) takes place at $T = 2$ K, where the tunneling make a dominant contribution. As seen from Fig. 2, the nonlinearity of the current–voltage characteristics is retained at given temperature (2 K) in the field $H =$ 90 kOe.

If the “effective resistivity” ρ_j is defined as $\rho_j = E(j)/j$, then, it follows from Fig. 2 that ρ_j at the metal–insulator transition temperature (120 K) is comparable with that at 2 K at the current density $j >$ 5 mA/cm^2. At small j, the $\rho_j(2\text{ K})/\rho_j(120\text{ K}) \approx 3$; this result can be obtained by analyzing the current–voltage characteristics and also from the $\rho(T)$ dependence for the polycrystal (Fig. 1) that was measured at $j \approx$ 0.06 mA/cm^2 (apparatus current is ~0.1 μA/cm^2).

When the current–voltage characteristic is nonlinear, the relative magnetoresistance will also be dependent on the current. Figure 3 shows the magnetoresistance $\Delta\rho(j) = \{E(H = 0, j) - E(H, j)\}/E(H = 0, j)$ as a function of j obtained from the current–voltage characteristics at $H = 50$ and 90 kOe. It is seen that, at $T =$ 2 K, $\Delta\rho(j)$ decreases markedly as the current increases to the 6.5 mA/cm^2. At temperatures of 20 and 120 K, the change in $\Delta\rho(j)$ is insignificant, since the nonlinearity of the current–voltage characteristics is smaller at these temperatures.

Thus, the magnitude of the magnetoresistive effect in polycrystalline $(La_{0.5}Eu_{0.5})_{0.7}Pb_{0.3}MnO_3$ studied in this work is almost constant (90%) at temperatures below the metal–insulator transition temperature. However, the mechanisms responsible for the observed magnetoresistance are different. This fact influences the shape of the current–voltage characteristics at various temperatures. For example, near the metal–insulator transition temperature, the magnetoresistance is determined by the influence of magnetic field on the ratio of the volumes of the conducting and insulating phases (mechanism of phase layering) inside the granules. In this case, the current–voltage characteristics remain linear in a quite wide range of the current values. At low temperatures, when the granules are even in a ground state of the ferromagnetic metal, the dominant contribution to the magnetoresistance is due to the intergranular tunneling of carriers through insulating interlayers of the granules. In this case, the current–voltage characteristics are typical of tunneling structures, i.e., they are strongly nonlinear, and the magnetoresistance depends on the current. Obviously, at intermediate temperatures, there is the superposition of above noted mechanisms that determines large magnetoresistance of the polycrystalline substituted lanthanum manganite.

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