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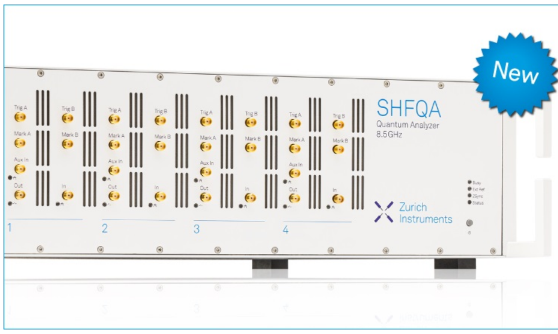

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Magnetoresistance of substituted lanthanum manganites $La_{0.7}Ca_{0.3}MnO_3$ upon nonequilibrium overheating of carriers

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Current–voltage characteristics of the polycrystalline substituted lanthanum manganite $La_{0.7}Ca_{0.3}MnO_3$ were experimentally studied at T = 77.4 K in magnetic fields up to 13 kOe. In these characteristics, a portion of negative differential resistivity was observed above a certain threshold value of critical current density j caused, in our opinion, by nonequilibrium heating of the electron gas due to low thermal conductivity of the manganite material. Because of the nonlinearity of the current–voltage characteristics, the field dependences of resistivity $\rho(H)$ appear extremely sensitive to the value of a transport current. In this case, the $\rho(H)$ dependences reveal both ordinary negative and positive magnetoresistance. © *2011 American Institute of Physics*. [doi:10.1063/1.3573666]

I. INTRODUCTION

Current–voltage characteristics (CVCs) of single-crystal, polycrystalline, and thin-film substituted lanthanum manganites $R_{1-x}A_xMnO_3$, where R is a trivalent rare-earth ion (La^{3+}, Nd^{3+}, Pr^{3+}, Sm^{3+}, etc.) and A is a divalent ion (Ca^{2+}, Sr^{2+}, Ba^{2+}, or Pb^{2+}), have been thoroughly investigated by many researchers.[1–9] The main feature of the $R_{1-x}A_xMnO_3$ CVCs is their nonlinearity strongly dependent on the values of a transport current and an external magnetic field. In particular, nonlinearity of the CVCs of single-crystal manganite samples are usually explained on the assumption that a crystal is in the inhomogeneous state of structural phase separation, when the phases of different conductivity coexist in a sample volume and the volumetric ratio between these phases is varied by external factors, such as a transport current and a magnetic field.[1,2] In the case of polycrystalline samples, at temperatures far below the temperature of the metal–dielectric transition, the CVCs are also nonlinear. Here this is related to spin-dependent tunneling of carriers between ferromagnetic grains via dielectric intercrystallite boundaries. However, at sufficiently wide ranges of the measuring current and voltage, the CVCs of manganites demonstrate one more intriguing phenomenon, namely, the so-called current switching effect, when, starting from a certain critical current or voltage value, portions of negative differential resistance are observed in the CVCs.[3–9] The origin of this phenomenon is still unclear; it may be related to breaking the regions of charge ordering in the manganites that leads to a sharp increase in conductivity,[2,5–8] sample overheating,[3] or nonequilibrium heating of carriers.[4,9] This effect observed in the CVCs is obviously of great interest, as the materials characterized by such CVCs are promising for microelectronic applications.

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II. EXPERIMENT

In this study, a sample of the classical composition $La_{0.7}Ca_{0.3}MnO_3$ prepared using a standard ceramic technique was used for studying the CVCs in wide ranges of the measuring currents and voltages. The sample, according to x-ray diffraction data, has a perovskitelike structure without foreign impurities. The data of microstructural studies by scanning electron microscopy (SEM) were reported in detail in Ref. 10. It appeared that the shape of the $La_{0.7}Ca_{0.3}MnO_3$ grains is close to spherical, with an average size of ∼200 nm. High-resolution transmission electron microscopy of an individual grain showed that the grain interior is a single crystal, while the grain shell is amorphous dielectric that forms potential barriers between the grains. Thus, the polycrystalline $La_{0.7}Ca_{0.3}MnO_3$ sample represents a network of tunnel contacts that consists of the conducting grains and dielectric shells. The magnetic and magnetotransport properties of these samples were reported previously.[10,11]

The temperature and field dependences of resistivity ρ(T) and $\rho(H)$ and the CVCs were taken using a standard four-probe method. The current–voltage characteristics were obtained in the mode of current scan up to high instrumental values of a transport current (1 A); the current was controlled with a Keithley 2430 source and the voltage drop was measured with a Keithley 2000 Voltmeter. During the CVC measurements, the sample was placed in liquid nitrogen to avoid possible self-heating and contact heating. To obtain the CVCs at different values of the external magnetic field and the $\rho(H)$ and $\rho(H, T)$ dependences, a cryostat with the sample was placed between the poles of an electromagnet; the magnetic field was applied perpendicular to the transport current direction.

Note that during the CVC measurements in a pumped Dewar flask where the temperature was stabilized by helium gas circulation, typical self-heating of the sample was observed, at which fixation of the transport current led to either the growth of the temperature of the metal–dielectric transition (below T_{max}) or the drop of resistivity (at $T \geq T_{MAX}$).

In addition, we observed a wide thermal hysteresis and simultaneous growth of the temperature of a heat sensor located near the sample. The current–voltage characteristics were measured at constant current scan and in the pulse mode with the length of a rectangular pulse from 200 ms to 1 s. No effect of the current scan modes on the CVCs was found.

III. RESULTS AND DISCUSSION

Figure 1 presents the ρ(T) dependences of polycrystalline $La_{0.7}Ca_{0.3}MnO_3$ at the external magnetic fields $H=0$ and 13 kOe that are typical for the substituted lanthanum manganites. The measuring current density was less than 0.01 A/cm^2. The temperature of the metal–dielectric transition at $H=0$ is $T_{max} \approx 115$ K. The experimental CVCs taken at liquid nitrogen temperatures in different external magnetic fields H are shown in Fig. 2. One can see the following main features. First, at small current densities (up to $\approx$2.2 A/cm^2), the characteristics are linear over the entire range of the magnetic fields. Then, the CVCs become nonlinear and after reaching a certain critical value of the current density (j_{cr}) the differential resistivity drops and becomes negative. At high current densities, the CVCs measured at different values of the external magnetic field tend to the universal curve. In addition, a small hysteresis is observed. With an increase in the external magnetic field, the j_{cr} value grows and critical strength E_{max} decreases. Thus, the CVC of polycrystalline $La_{0.7}Ca_{0.3}MnO_3$ also has a portion of the negative differential resisitivity, which was observed on both single-crystal and thin-film substituted lanthanum manganites.[9]

Qualitatively, such a behavior resembles that observed by us previously on the CVCs of the $(La_{0.5}Eu_{0.5})_{0.7}Pb_{0.3}MnO_3$ single crystals.[9] We explained this feature by internal nonequilibrium heating of carriers, when the temperature of the electron gas substantially (by hundreds of degrees) differs from the thermodynamic temperature of a sample. Such nonequilibrium overheating of the electron gas causes the formation of the CVC with the portion of the negative differential resistivity. Due to the fluctuating concentration of electrons and their temperature in the sample with a semiconductor-type temperature-dependent resistivity, the current flows inhomogeneously. The sample divides into areas of strong and weak current. The areas of strong current form cylindrical current streams inside the sample volume and the final temperature of the electron gas is determined by heat exchange with the rest of the sample volume and the refrigerant.[12] A necessary condition for implementation of nonequilibrium heating is low thermal conductivity of the sample that weakens energy exchange between the electron and phonon subsystems.

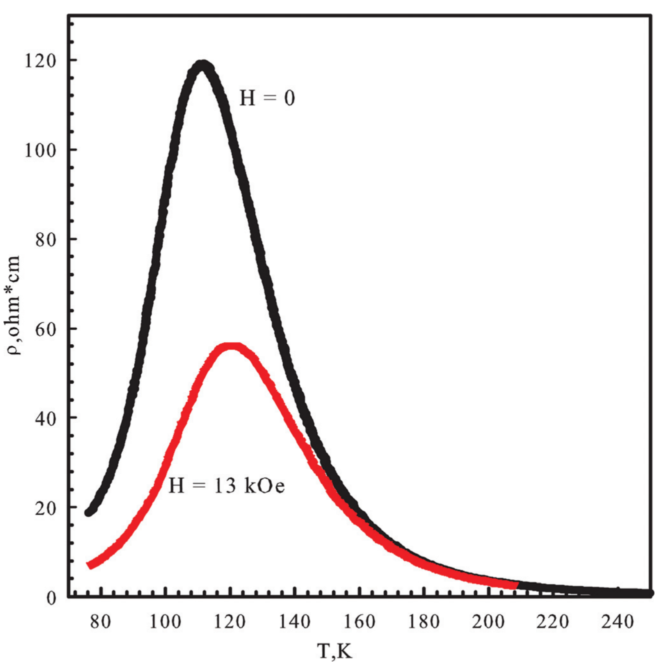


FIG. 1. (Color online) Experimental ρ(T) dependences of polycrystalline $La_{0.7}Ca_{0.3}MnO_3$ at $H=0$ and $H=13$ kOe.

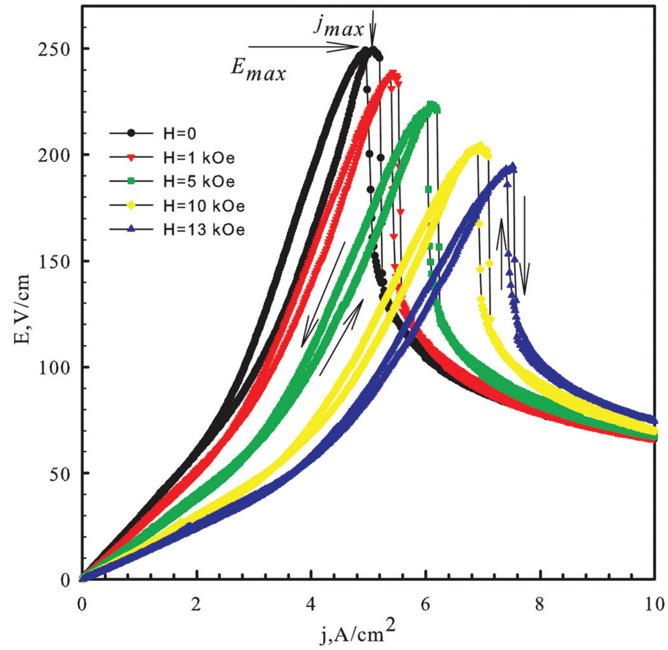


FIG. 2. (Color online) Experimental current–voltage characteristics (CVC's) of $La_{0.7}Ca_{0.3}MnO_3$ measured at T = 77.4 K at different applied magnetic fields.

The same approach can be used for explanation of the CVC behavior of the polycrystalline $La_{0.7}Ca_{0.3}MnO_3$ samples. The mechanism of carrier overheating proposed earlier[4,9] is proved also by the fact that the specific power values at E_{max} and j_{max} [see Fig. (2)] at which the current switching effect is observed are the same for all the values of external magnetic field H. The specific power released in the sample during the measurements is $P = E \times j = E^2/\rho$. Consequently, if the dependence of E^2 on ρ is linear, its slope determines the P value. Figure 3 demonstrates this dependence, linear within the measurement error; the specific power is $P = 1300 \pm 100$ W/cm^3. Thus, the position of the E_{max} is independent of the parameters of the sample itself and determined only by the power released during the measurements.

Now, estimate quantitatively the internal overheating of the sample, using simple relations similar to those reported in Ref. 9. The Fourier equation of thermal conductivity for simple heating by means of the Joule heat allows estimating the carrier overheating

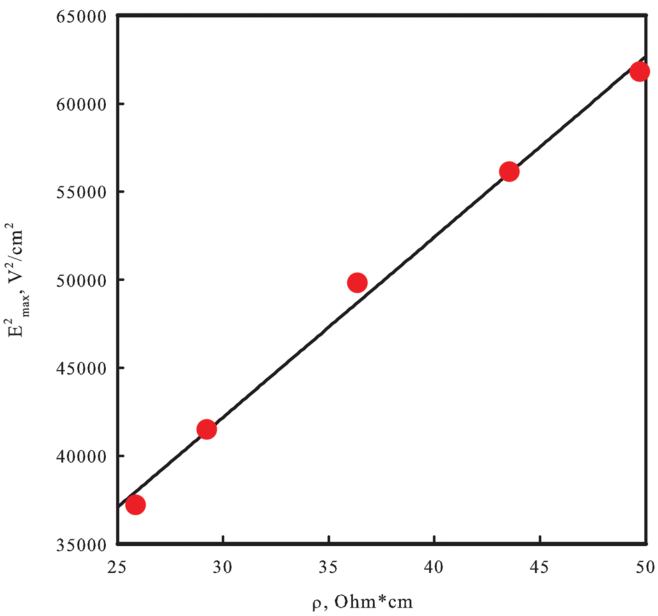


FIG. 3. (Color online) E^2 versus ρ plot of $E_{max}(H)$ at j_{max} (Fig. 2) at which the current switching effect is observed (circles). The slope of linear interpolation gives the value of specific power $P = 1300 \pm 100$ W/cm^3 same for all applied magnetic fields.

$$\Delta T = P \times l/(k \times S), \tag{1}$$

where P is the power released in the sample during the current flow, ℓ is the length of the current channel, κ is the thermal conductivity of the sample, and S is the total cross-section area of all the current channels. All the variables in this expression are, generally, functions of temperature; therefore, accurate calculation requires using its temperature dependences. The final temperature of the current channel is

$$T_{fin} = T_0 + \Delta T,$$

where T_0 is the initial temperature, i. e., the temperature of the CVC measurement. Thus, the overheating of the current channel in the sample can be estimated as

$$T_{fin}(E) = T_0 + E^2 \times l^3(T) \times [R(T,H) \times k(T) \times S(T)]^{-1} \tag{2}$$

where E is the electric field strength, R(T,H) are the experimental temperature dependences of resistivity at different values of the external magnetic field (in this case, $H = 0$ and 13 kOe). Since the contribution of electron thermal conductivity is minor and all the transfer will be concentrated inside the current channels, just the sample volume and not the refrigerant serves as a heat sink. The thermal conductivity of the substituted lanthanum manganites is $\kappa(300\ \text{K}) = 1.5$ W/m $\times$ K.[13] In this study, we did not determine quantities ℓ and S, unlike, for instance, the work in Ref. 14, when they were obtained by visualization of the current channels. Estimation can be made using parameter ℓ equal to the distance between the potential contacts and considering S as a simulation fitting parameter that is not higher than the cross-section area of the sample. The results of simulation of the current stream heating using expression (2) for different magnetic fields are given in Fig. 4. In the simulation, the fitting parameter $S/\ell = 0.11$ cm was used close to the sample geometry. This parameter determined E_{max} in the figure. At this E value, the CVC passed to the portion of the negative differential resistivity. One can see that the calculated E_{max} values are in good agreement with the experimental data for zero and maximum magnetic fields H [see Fig. (2)]. The simulation results allow as to conclude that at the field strengths of hundreds of W/cm, considerable sample overheating occurs that differs from the initial temperatures by hundreds of

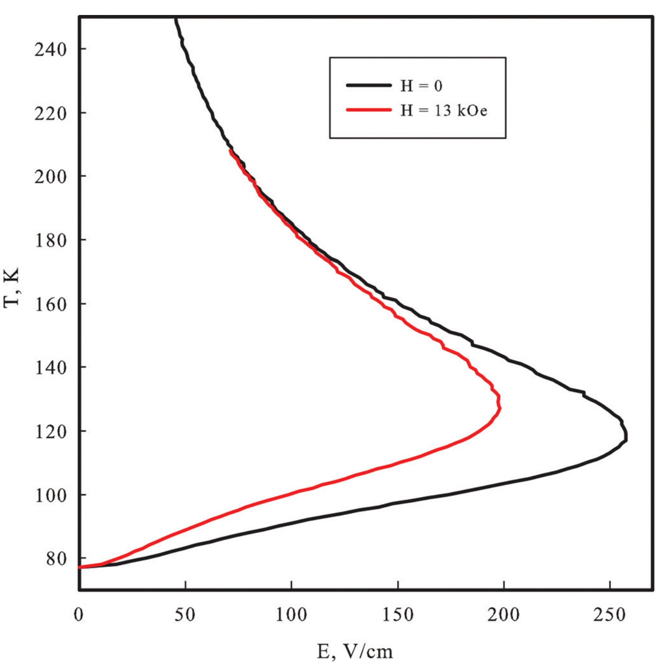


FIG. 4. (Color online) Simulation results of overheating of current streams at different magnetic fields.

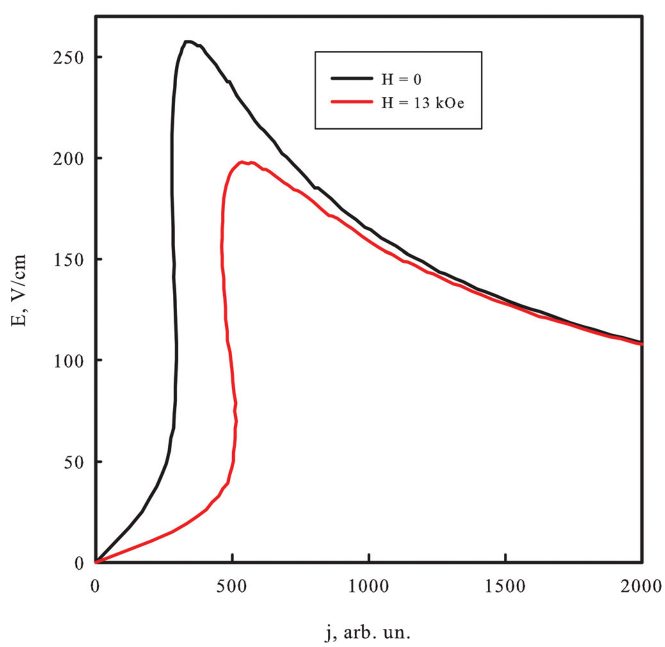
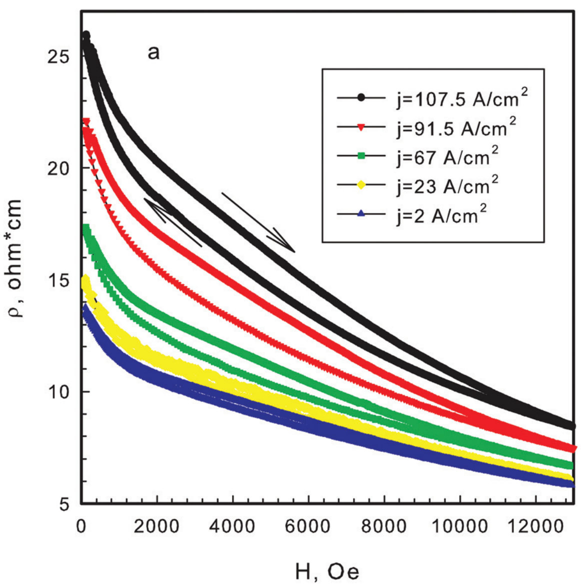


FIG. 5. (Color online) Obtained $E(j)$ dependences using the simulation results at different magnetic fields.



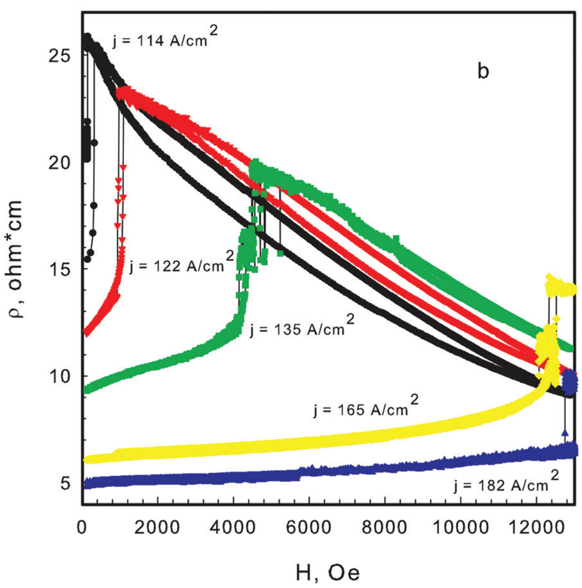


FIG. 6. (Color online) Experimental $\rho(H)$ dependences of polycrystalline $La_{0.7}Ca_{0.3}MnO_3$ (a) at $j < j_{max}$ ($H = 0$) and (b) at $j > j_{max}$ ($H = 0$).

degrees. The overheating is the most rapid upon approaching the temperature of the metal–dielectric transition from the low-temperature area. It is seen from Fig. 4 that within this consideration the CVCs measured at temperatures below the maximum of the metal-dielectric transition will reveal the hysteresis feature, as the T(E) function at $T_0 < T_{max}$ is double-valued. This is confirmed by the CVC measurements at the boiling point of liquid nitrogen [see Fig. (2)]. The obtained $T(E)$ functions make it possible to get the approximate $E(j)$ dependences by associating an R value to each E value at a given temperature [see Fig. (1)] and, then, obtaining a current value (j) following the Ohm's law. It should be noted that such association will yield only a qualitative form of the CVCs, as the temperature dependences of resistivity were measured at $j = 1$ mA, i.e., in the linear portion of the CVCs, where the overheating process is not decisive yet. In our case, determination of the real current density requires consideration of its variation with increasing temperature of the current channel. Nevertheless, this procedure was made; the resulting $E(j)$ dependences at T = 77 K are presented in Fig. 5. Qualitative agreement between the experimental [see Fig. (2)] and calculated [Fig. (5)] data is observed.

Stability of the thermodynamic temperature of the sample together with the nonequilibrium heating of the electron gas (during the measurements in both $j < j_{max}$ and $j > j_{max}$ modes) and the strong effect of the external field on the CVCs and j_{max} value cause the anomalous field dependence of magnetoresistance at the high measuring transport current densities ($j > j_{max}$). Figure 6 shows the $\rho(H)$ dependences measured in the mentioned modes [$j < j_{max}$ in Fig. 6(a) and $j > j_{max}$ in Fig. 6(b)]. The dependences taken in the $j < j_{max}$ mode are typical of the polycrystalline substituted lanthanum manganites and explained by spin-dependent tunneling of carriers via intergrain dielectric boundaries.[10,15] A different picture, atypical of the substituted lanthanum manganites, is observed in the $j \geq j_{max}$ mode. There are portions of both negative and positive magnetoresistance. The portions of the sharp variation in magnetoresistance and the transition from positive to ordinary negative magnetoresistance are related to the field effect on j_{max}, i.e., on the temperature of the transition from equilibrium to nonequilibrium current flow (Fig. 4) corresponding to the maximum of the R(T) dependences (Fig. 1).

IV. SUMMARY AND CONCLUSIONS

Thus, if the CVCs of the substituted lanthanum manganites are determined by the nonequilibrium overheating of carriers, the $\rho(H)$ dependences become extremely sensitive to the value of measuring current j and sometimes have portions of positive magnetoresistance, which is observed for the first time. The analysis of the results obtained also allows one to conclude that the internal overheating of carriers during the transport current flow plays an important, often decisive, role in the formation of the CVCs and, as a consequence, the field dependences of resistivity of the materials with low thermal conductivity and specific heat, such as manganites and other oxides.

ACKNOWLEDGMENTS

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