Current-induced giant electroresistance in $La_{0.7}Sr_{0.3}MnO_3$ thin films

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The electroresistance (ER) and magnetoresistance (MR) of $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) thin films with different thicknesses (t) are investigated. We found a metallic-to-insulating (*M-I*) transition in films with $t=80$ nm. For the 80-nm film, a giant room-temperature ER ratio of 11.3% is achieved with an electric current of 0.9 mA. This value of the ER ratio is four times larger resistive response than that of the MR ratio under 1 T. The enhancement of the ER value in these disordered metallic LSMO thin films is correlated with the coexistence of metallic and insulating phases and attributed to the mechanism of phonon-assisted delocalization.

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INTRODUCTION

The phenomenon of colossal magnetoresistance (CMR) in perovskite compounds $La_{1-x}A_xMnO_3$ (Refs. 1 and 2) (A $=$Ca, Sr, Ba, Pb) has attracted considerable attention in recent years due to the related physics and potential applications. A large number of experiments on polycrystals, single crystals, and thin films have been carried out to explore the dependence of the magnetoresistance (MR) on the temperature (T), magnetic field (H), and compositions as well as on synthesis process. Indisputably, shaping them into the form of a thin film is mostly required for the application of magnetronic devices.[3–5] The basic behavior of CMR's generally the same in both bulk and thin-film samples, except for some properties associated with the strain effect induced by the lattice mismatch between film and substrates.[6,7] One of the serious problems in a practical application of CMR materials is the insufficient magnetoresistive response at room temperature (RT) under a low field ($H<1\ \mathrm{KO}_e$). Fortunately, some studies showed that the manipulation of resistive states in CMR manganites can be achieved not only by a magnetic field but also by an electric (E) field. It has been reported that an electric current (I) could trigger the transformation of the electrically insulating charge-ordered state to a ferromagnetic metallic state.[8,9] Furthermore, a correlation[10] between electroresistance (ER) and MR has been established in $La_{0.82}Ca_{0.18}MnO_3$ (LCMO) single crystal, and the function of an electric current of 0.3 mA was shown to be equivalent to 1.5 T at a temperature below Curie temperature (T_c), and 0.4 T at RT. A rough estimation[10] shows that a 1-nm-wide filamentary path biased with 1 mA could produce a magnetic field of 1 T. Since the ER effect is strongly correlated with the MR effect and the MR ratio is very much dependent on the film thickness (t),[11,12] the simultaneous investigation of ER and MR effects with different thicknesses is essential for not only the basic research but also for technological applications. In this paper, we report our ER and MR studies on $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) thin films. We choose this composition because it possesses a $T_c>300$ K, which is a great advantage for practical applications at room temperature. Our finding is the giant ER ratio of 11.3% at RT for a t $=80$ nm film, which is four times larger than the MR ratio produced by an applied field of 1 T. This large current-induced resistive change cannot be explained simply by the percolation model, and is associated with the quantum effect. A detailed analysis will be discussed in the following.

EXPERIMENTS

A series of films with various thicknesses were fabricated by rf magnetron sputtering[13] under identical deposition conditions using a sintered stoichiometric LSMO target in an $Ar+O_2$ atmosphere, with a pressure and a rf power of 20 mTorr and 3.56 W/cm^2, respectively. Films were synthesized on a $LaAlO_3$ (LAO) (100) substrate. After deposition the samples were post-annealed at 920 °C under flowing oxygen. The processing parameters were first optimized to obtain $T_c>300$ K for 150–300-nm films; then the same parameters were adopted for other thicknesses. The four-point contact preparation and the experimental setup for the electrical resistivity (ρ) and current-voltage (I-V) measurements was already described in Refs. 14 and 15. For a resistance-field (R-H) measurement a sweep field from -1 to 1 T is applied. The MR ratio is defined as $[\{(\rho_{H=1T}-\rho_{H=0})/\rho_{H=0}\}\times 100\%]$, and the ER ratio is defined as $[\{(dV/dI(I)-dV/dI(0))/dV/dI(0)\}\times 100\%]$. The film thickness was measured by the stylus method on a Dektek-3030 ST profilometer. The surface morphology and composition of films were determined using a high resolution (JEOL-JSM 6700F) scanning electron microscope (SEM) and by energy-dispersive x-ray EDX analysis (Hitachi-S570), respectively. The phase purity and structure of the film were identified by the x-ray-diffraction method.

RESULTS AND DISCUSSION

EDX results indicate that our films were slightly Mn deficient, and the La/Sr ratio was found to be 2.29 after post-annealing. Figure 1 displays $\rho(T)$ plots at zero and 1 T for films with different thicknesses. For $t>100$ nm, the $\rho(T)$ behavior is very similar to that of $t=100$ nm; we therefore do not show the data here. As seen in Figs. 1(a) and 1(c), the 60-nm film behaves as insulating with an upturn at 150 K, while the 100-nm film shows a metallic feature with ρ decreasing with lowering temperature. The result in these two panels clearly demonstrates that there exists a critical thickness for driving the insulating phase to the metallic phase. For the intermediate thickness $t=80$ nm [see Fig. 1(b)], ρ

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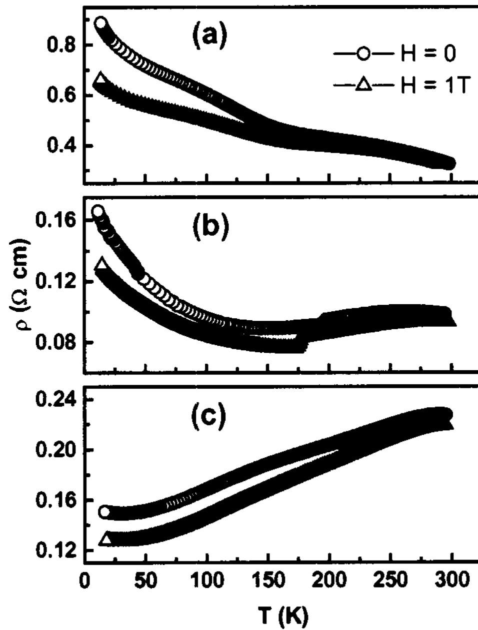

FIG. 1. ρ vs T under zero and 1-T fields for LSMO/LAO films with different thicknesses: (a) 60 nm, (b) 80 nm, and (c) 100 nm.

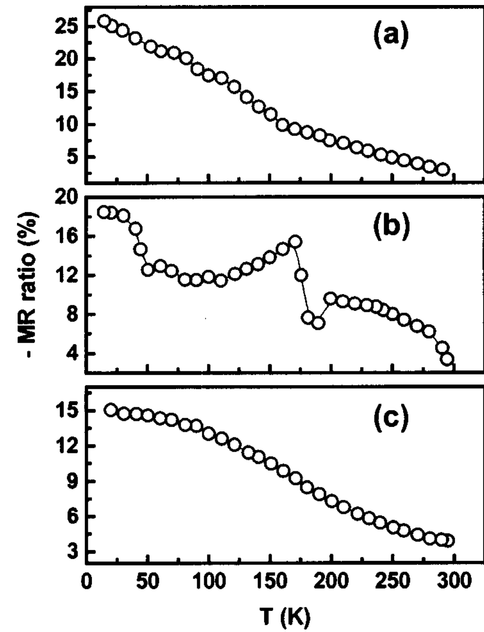

FIG. 2. MR(T) plots of LSMO/LAO films with different thicknesses: (a) 60 nm, (b) 80 nm, and (c) 100 nm.

first decreases with a sharp drop at 200 K, then behaves as an insulator when T decreases from 200 to 10 K. This type of low-temperature ρ upturn has been observed in a polycrystalline sample[16] as well as in thin films.[17] The former was attributed to an intergranular Coulomb gap between grains (20–25 nm), and the latter to the coexistance of high-strain/low-strain mixed phases. Our sample condition is very similar to the latter case. First, the average grain size of our polycrystalline films is more than 50 nm (based on SEM data), which should not yield much difference in their electrostatic energy. Second, the high-strain/low-strain mixed phases were observed in a LCMO/LAO film, in which the lattice mismatch between the film (0.386 nm) and substrate (0.379 nm) is -1.85%. Therefore, the possible origin for the ρ upturn behavior in our film of 80-nm thickness is the structural disorder by strain in association with the large lattice mismatch (-2.37%) between LSMO (0.388 nm) and LAO. Similarly, the strain-induced lattice distortion still remains for 60-nm LSMO films,[18,19] and the strained lattice deformation is shown to control the direction of easy magnetization of LSMO films whatever the thickness of the film may be. Moreover, disorder has been observed even in epitaxial thin $(La,Ca)MnO_3$ films[20] deposited by the sputtering technique. The structural disorder can result in spin disorder, and enhance the electron localization,[21] or may lead to the absence of the characteristic insulator-metal transition in ferromagnetic manganites.[22] This behavior very likely appears in our 60/80-nm film, since the structural disorder and surface spin disorder effects become more significant as the film thickness is reduced. The disorder yields a reduction in the mobility of the conduction electrons and enhances the resistivity. Thus the upturn of $\rho(T)$ in our films seems to be the localization effect due to structural disorder. Recently, a similar ρ upturn was also seen in 80-nm La-Sn-Mn-O epitaxial films and was described as the localization effect.[12,23]

Figure 2 displays the $-\mathrm{MR}(T)$ plots for $t=60$, 80, and 100 nm films. It is observed that the MR ratio for all films increases with decreasing temperature, but its value is thickness dependent. At 15 K, it increases from 15.2% to 25.8% as the film thickness decreases from 100 to 60 nm. In a simple negative MR picture, the application of a magnetic field aligns the spins of ferromagnetic metallic regions; as a result ρ of the sample decreases. Our 100-nm film shows metallic conduction [Fig. 1(c)] and the 60-nm film shows insulating behavior [Fig. 1(a)]. However, the MR of a 100-nm film is observed to be slightly higher than that of 60 nm at RT, but at low temperature the MR behavior is opposite to that at RT. We interpret the results as follows. Structural disorder can enhance the electron localization, and the conduction electrons in FM mixed valence manganites are declocalized on an atomic scale; however, they may be weakly localized in large wave packets as described in an extended localization model.[24] It is obvious that at low temperature the spin disorder reduces, and the application of H may easily induce a net spin alignment of the localized states; as a result ρ reduces substantially. Hence the low-temperature MR is higher for a high-strained 60-nm film. Indeed, several groups have reported[25–27] an enhanced MR effect in manganite films and bulks by introducing structural and spin disorder. The ratio (%) of low (15-K) and high-temperature (300-K) MR are found to be 8.7, 6.5, and 3.9 for $t=60$, 80, and 100 nm, respectively. This increase in the MR ratio at low temperature with decreasing thickness is consistent with the reported results.[11]

Figure 3 shows the MR(H) graphs derived from R-H plots at RT. We observed a linear decrease of R with increasing H, which indicates spin-related electron scattering at grain boundaries.[28] In this case the MR increases from 2.96% to 3.85% as the film thickness increases from 60 to 100 nm. This marginal increase of the MR with thickness is

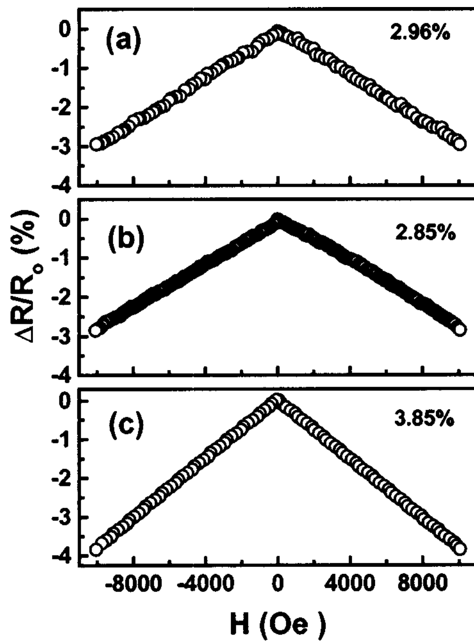

FIG. 3. MR ratio (%) vs H at RT for LSMO/LAO films with different thicknesses: (a) 60 nm, (b) 80 nm, and (c) 100 nm. MR values at 1-T are displayed on right top corner of each layer.

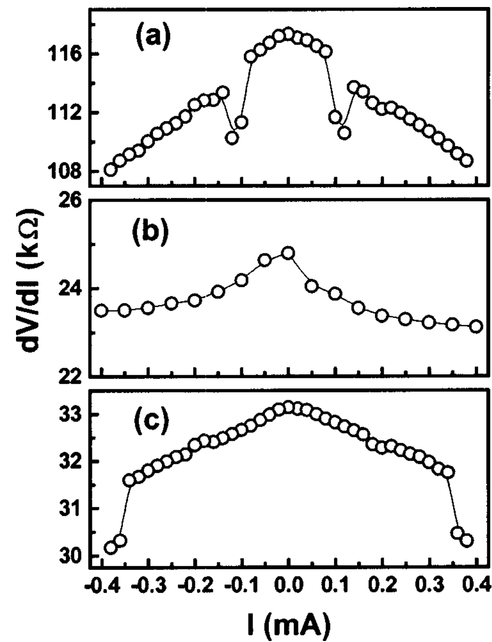

FIG. 4. Differential resistance (dV/dI) vs current (I) at RT for LSMO/LAO films with different thicknesses: (a) 60 nm, (b) 80 nm, and (c) 100 nm.

consistent with the report[21] that high-field MR remains almost constant for a film with $t>20$ nm. This low MR at RT is ascribed to the spin disordering of LSMO at high T.[29] Our result from MR (3.85%) for $t=100$ nm is very close to the reported value of 3.81% for a $t=200$ nm LSMO film under a 1-T field.[30]

The ER effect was studied by measuring the differential resistance[10] (dV/dI) as a function of I. The obtained results are shown in Fig. 4. The main features of ER curves are almost same as the MR graphs, as shown in Fig. 3. The effect of the Joule heating due to the current flow can be examined in the higher current range. The differential resistance decreases with increasing current, which is opposite to the Joule heating effect. It is therefore reasonable to assume that Joule heating is irrelevant to our transport results, qualitatively at least. Figure 5 shows the ER ratio (%) vs current (I) derived from $dV/dI-I$ plots. With $I=0.3$ mA, we obtained 5.6%, 6.4%, and 3.5% of ER ratio at RT for $t=60$, 80, and 100 nm, respectively. With $I=0.9$ mA the ER ratio increases to 11.3% and 4.6% for $t=80$ and 100 nm, respectively. This indicates that the room-temperature ER value decreases 1.6 times when t increases from 60 to 100 nm (see Fig. 5), whereas the MR increases to 1.3 times (see Fig. 3) for the same thickness variation. Although there is similarity between MR(H) and $dV/dI(I)$ curves (Figs. 3 and 4), the increasing (decreasing) tendency of the MR (dV/dI) with increasing thickness suggests that both effects may not have exactly the same origin as normally argued for the magnetic and electric field effects. According to the percolation model, an electric field perturbs the coexistence of metallic and insulating regions by creating metallic inclusions[31] within the insulating regions. This metallic inclusion may in turn produce a filamentary path where the outer layer is insulating and the inner one is metallic. Besides CMR materials, this type of filamentary pattern has also been observed by applying current in amorphous hydrogenated silicon devices.[32] Hence current flow through space, limited within the filamentary regions, induces an intense local magnetic field which polarizes the FM regions and induces the CMR effect. In this case the resistive changes due to a magnetic field and applied current are expected to be equivalent, as reported for a LCMO crystal.[10] However, our studies show that the thickness dependence of the MR and ER effects are in the oppo-

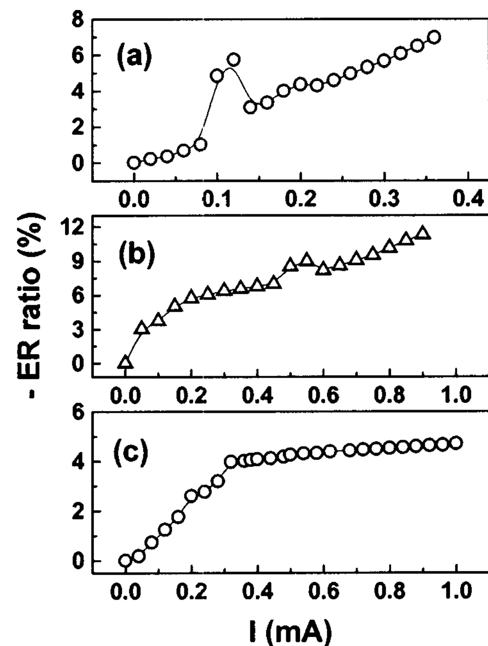

FIG. 5. ER ratio (%) vs current (I) for LSMO/LAO films with different thicknesses: (a) 60 nm, (b) 80 nm, and (c) 100 nm.

TABLE I. A comparison of room-temperature MR_H and MR_I. MR_H: MR ratios (%) obtained at 1 T. The MR_I: ER ratio (%) at different applied current (I) in mA as shown in the bracket. r: the ratio of MR_I and MR_H.

Film thickness (nm)	MR_H	MR_I	r
60	2.96	5.6 ($I=0.3$)	1.9
80	2.85	6.4 ($I=0.3$)	2.3
		11.3 ($I=0.9$)	4.0
100	3.85	3.5 ($I=0.3$)	0.9
		4.6 ($I=0.9$)	1.2

site direction. Furthermore, based on the ER and MR studies in a single crystal (Ref. 10), the value of the ER ratio should be nearly equivalent to that of the MR ratio at various temperatures from 295 and 65 K. In the percolation picture, a local electric field perturbs the coexistence of phases of different electronic densities, and sets up filamentary currents across nonconductive regions. This filamentary current, in turn, produces a magnetic field to induce MR drop. Keeping in mind that the local magnetic field generated by the same current could be different at different temperatures depending on the resistance change, the increase or decrease rate of the local field should be the same as the increase or decrease rate of the MR ratio. As described in Ref. 10, for the single crystal, a current of 0.3 mA is equivalent to the effect of applying a magnetic field of 1.5–2 T at low temperature, and only 0.4 T at room temperature. Therefore, both the ER and MR increase five times at low temperature. Nevertheless, this model could not explain the results of our films with high strain (60 and 80-nm films), because the ER value is much higher than the MR ratio at room temperature, while they are

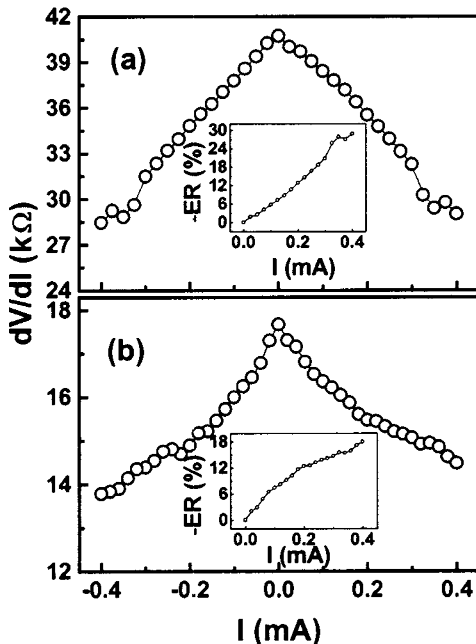

FIG. 6. Differential resistance (dV/dI) vs current (I) at low temperature (15 K) for LSMO/LAO films with different thicknesses: (a) 80 nm and (b) 100 nm. Insets display the ER ratio (%) vs current (I) for corresponding thicknesses.

TABLE II. A comparison of MR_H, MR_I, and r values obtained at 15 K.

Film thickness (nm)	MR_H	MR_I	r
80	18.5	20.8 ($I=0.3$)	1.1
100	15.0	14.7 ($I=0.3$)	0.98

almost equivalent at 15 K. Some simple calculations further demonstrate why the argument of filamentary current could not work in our films. The room-temperature ρ values (from the raw data of Fig. 1) are 0.098 and 0.228 Ω cm for 80 and 100-nm films, respectively. Based on the percolation scenerio, the local electric field ($E=J\rho$; J is the current density) produced by 0.3-mA current in an 80-nm film should be lower than that produced in a 100-nm film, and should result in less MR ratio for an 80-nm film. But our data for the MR_I ratio (see Table I) show an opposite result. Another example is as follows: ρ of an 80-nm film at 15 K is 1.6 higher than that at the room-temperature value. Hence the 0.3-mA current would produce a stronger local electric field and, accordingly, the MR_I ratio should increase 1.6 times. However, our data show that its MR_I ratio at 15 K is 3.2 times larger than that at room temperature.

A comparison of the resistive changes due to magnetic field and current effects is given in Table I. The ER ratio (%) at different applied currents (in mA, as shown in brackets), the MR ratio (%) at 1-T magnetic field, and their ratio are designated as MR_I, MR_H, and r, respectively. It is seen that MR_H and MR_I are almost equal for a 100-nm film. But the higher values of MR_I, as compared to MR_H for 60- and 80-nm films, indicate that there is some other mechanism involved in addition to the percolation scenario. According to Fig. 1(b), the $\rho(T)$ of the 80-nm film has a M-I transition around 200 K, but the ρ value at the low temperature is much lower than the regular insulator. It is also well known that any kind of disorder in the conduction channel can lead to the localization of electrons and to a possible M-I transition and also to an instability against the formation of an insulator.[33,34] Disorder has been observed in epitaxial thin (La,Ca)MnO_3 films[20] deposited by the sputtering technique. It is, therefore, reasonable to consider our sample to be a strongly disordered metallic system, and the occurence of the M-I transition may be caused by a quantum effect—the Anderson transition.[35] In the model of the Anderson transition, an electron moving in a random potential may have either a localized or extended eigenstate depending on the energy of the electron. Extended states can carry a direct current whereas localized states are bound to a certain region and can move only with the assistance of another mechanism (e.g., phonon-assisted hopping).[36] When one passes a current to the sample, localized electrons gain energy and become conductive via phonon-assisted hopping. Accordingly, the electrical resistance decreases. This explains why the MR_I of an 80-nm film for a current of 0.3 mA (equivalent to a 1.5-T field; see Ref. 10) is 2.3 times larger than that of MR_H at a 1-T field. Hence, to get a clear picture on the phonon-assisted delocalization effect, we studied the ER effect for $t=80$ and 100 nm films at low temperature to minimize the phonon contribution.

The results are displayed in Fig. 6 in which insets depict

the corresponding ER ratio (%) vs the I plot. It is seen that ER ratio increases with increasing current. The ER value is found to be 1.4 times higher in an 80-nm film than that of 100-nm film. Table II gives comparison between MR_H and MR_I values. It is seen that MR_I values at 15 K for both 80- and 100-nm films are close to the corresponding MR_H values (the r values are all close to 1). The evidence, that the current effect is almost the same as the magnetic-field effect at low temperature, while it is more influential than the magnetic-field effect at room temperature, is consistent with the model of phonon-assisted delocalization.

In summary, we have investigated simultaneously the ER and MR effects in LSMO films with different thicknesses and we have observed a room-temperature ER ratio of 11.3% in an 80-nm film which is about a four times larger resistive response than that observed in a MR ratio under 1 T. Based on our analysis, we suggest that phonon-assisted delocalization may be the origin of the giant ER effect in the disordered metallic LMSO thin films. It is also revealed that a mixture of localized and FM metallic states are advantageous for the enhancement of room temperature ER in LSMO thin films.

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[1] Y. Shimakawa, Y. Kubo, and T. Manaka, Nature (London) **379**, 53 (1996).

[2] P. Schiffer, A. P. Ramirez, W. Bao, and S. W. Cheong, Phys. Rev. Lett. **75**, 3336 (1995).

[3] J. H. Park, E. Vescovo, H. J. Kin, C. Kwon, R. Ramesh, and T. Venkatesan, Nature (London) **392**, 794 (1998).

[4] T. Venkatesan, M. Rajeswary, Z. W. Dong, S. B. Ogale, and R. Ramesh, Philos. Trans. R. Soc. London, Ser. A **356**, 1661 (1998).

[5] J. Daughton and J. Granley, Ind. Phys. **5**, 22 (1999).

[6] H. L. Ju, C. Kwon, Q. Li, R. L. Greene, and T. Venkatesan, Appl. Phys. Lett. **65**, 2108 (1994).

[7] W. Prellier, A. Biswas, M. Rajeswari, T. Venkatesan, and R. L. Greene, Appl. Phys. Lett. **75**, 397 (1999).

[8] A. Asamitsu, Y. Tomioka, H. Kuwahara, and Y. Tokura, Nature (London) **388**, 50 (1997).

[9] C. N. R. Rao, A. R. Raju, V. Ponnambalam, Sachin Parashar, and N. Kumar, Phys. Rev. B **61**, 594 (2000).

[10] V. Markovich, E. Rozenberg, Y. Yuzhelevski, G. Jung, G. Gorodetsky, D. A. Shulyatev, and Ta. M. Mukovskii, Appl. Phys. Lett. **78**, 3499 (2001).

[11] J. G. Lin, C. Y. Huang, S. Y. Lin, and P. C. Kuo, Physica C **364**, 668 (2001).

[12] X. Guo, S. Dai, Y. Zhou, G. Yang, and Z. Chen, Appl. Phys. Lett. **75**, 3378 (1999).

[13] A. K. Debnath, J. G. Lin, S. L. Cheng, P. C. Kao, and K. C. Lin (unpublished).

[14] C. W. Chang, A. K. Debnath, and J. G. Lin, Phys. Rev. B **65**, 024422 (2001).

[15] C. W. Chang, A. K. Debnath, and J. G. Lin, J. Appl. Phys. **91**, 2216 (2002).

[16] Ll. Balcells, J. Fontcuberta, B. Martinez, and X. Obradors, Phys. Rev. B **58**, R14697 (1998).

[17] A. Biswas, M. Rajeswari, R. C. Srivastava, Y. H. Li, T. Venkatesan, and R. L. Greene, Phys. Rev. B **61**, 9665 (2000).

[18] A. M. Haghiri-Gosnet, J. Wolfman, B. Mercey, Ch. Simin, P. Lecoeur, M. Korzenski, M. Hervieu, R. Desfeux, and G. Baldinozzi, J. Appl. Phys. **88**, 4257 (2000).

[19] R. A. Rao, D. Lavric, T. K. Nath, C. B. Eom, L. Wu, and F. Tsui, Appl. Phys. Lett. **73**, 3294 (1998).

[20] J. Aarts, S. Freisem, R. Hendrikx, and H. W. Zandbergen, Appl. Phys. Lett. **72**, 2975 (1998).

[21] H. S. Wang, E. Wertz, Y. F. Hu, Qi Li, and D. G. Schlom, J. Appl. Phys. **87**, 7409 (2000).

[22] A. P. Ramirez, J. Phys.: Condens. Matter **9**, 8171 (1997).

[23] A. Urushibara, Y. Moritomo, T. Arima, A. Asamitsu, G. Kido, and Y. Tokura, Phys. Rev. B **51**, 14103 (1995).

[24] J. M. D. Coey, M. Viret, L. Ranno, and K. Quandjela, Phys. Rev. Lett. **75**, 3910 (1995).

[25] J. Fontcuberta, L. Balcells, B. Martinez, and X. Obaradors, in *Nano-Crystalline and Thin Film Magnetic Oxides*, edited by I. Nedkov and M. Ausloos (Kluwer, Dordrecht, 1999), p. 105.

[26] S. B. Ogale, K. Ghosh, J. Y. Gu, R. Shreekala, S. R. Shinde, M. Downes, M. Rajeswari, R. P. Sharma, R. L. Greene, T. Venkatesan, R. Ramesh, R. Bathe, S. I. Patil, R. Ravikumar, S. K. Arora, and K. Mehta, J. Appl. Phys. **84**, 6255 (1998).

[27] E. S. Gillman, M. Li, and K.-H. Dahmen, J. Appl. Phys. **84**, 6217 (1998).

[28] J.-M. Liu, Q. Huang, J. Li, C. K. Ong, X. Y. Chen, Z. G. Liu, and Y. W. Du, Mater. Lett. **50**, 97 (2001).

[29] J.-M. Liu, Q. Huang, J. Li, L. P. You, S. Y. Xu, C. K. Ong, Z. G. Liu, and Y. W. Du, J. Appl. Phys. **88**, 2791 (2000).

[30] S. Y. Yang, W. L. Kuang, C. H. Ho, W. S. Tse, M. T. Lin, S. F. Lee, Y. Liou, and Y. D. Yao, J. Magn. Magn. Mater. **226–230**, 690 (2001).

[31] N. A. Babushkina, L. M. Belova, D. I. Khomskii, K. I. Kugel, O. Yu. Gorbenko, and A. R. Kaul, Phys. Rev. B **59**, 6994 (1999).

[32] A. Avila and R. Asomoza, Solid State Electron. **44**, 17 (2000).

[33] For review, see, e.g., J. M. Ziman, *Models of Disorder* (Cambridge University Press, Cambridge, UK, 1971).

[34] T. R. Kirkpatrick and D. Belitz, in *Electron Correlation in the Solid State*, edited by N. H. March (Imperial College Press, London, 1999), p. 297.

[35] P. W. Anderson, Phys. Rev. **109**, 1492 (1958).

[36] For a review, see *Localization, Interaction and Transport Phenomena*, Springer Series Solid-State Sciences Vol. 61, edited by B. Kramer, G. Bergmann, and Y. Bruynseraede (Springer-Verlag, Berlin, 1985).