

## Local magnetic order in manganite thin films studied by $1/f$ noise measurements

P. Reutler,\* A. Bensaid, F. Herbstritt, and C. Höfener

*II. Physikalisches Institut, Universität zu Köln, Zùlpicherstraße 77, D-50937 Köln, Germany*

A. Marx<sup>†</sup> and R. Gross

*II. Physikalisches Institut, Universität zu Köln, Zùlpicherstraße 77, D-50937 Köln, Germany*

*and Walther-Meißner-Institut für Tieftemperaturforschung, Walther-Meißner Straße 8, D-85748 Garching, Germany*

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The low-frequency  $1/f$  resistance fluctuations of epitaxial  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  thin films on different substrates have been studied as a function of temperature and applied magnetic field. A film with a high Curie temperature  $T_C=250$  K grown on a substrate with perfect lattice match ( $\text{NdGaO}_3$ ) shows a low overall noise amplitude, comparable to that of a conventional metal. In contrast to previous work, no increase of the  $1/f$  resistance noise in the ferromagnetic regime of the  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film could be observed. The magnitude of the normalized resistance fluctuations decreases with decreasing temperature except for a sharp peak at  $T_C$ ; this peak is completely suppressed by an applied magnetic field of about 2 T. In contrast, the noise level in a film with low Curie temperature ( $T_C=110$  K) grown on a substrate ( $\text{SrTiO}_3$ ) with large lattice mismatch (1.5%) is several orders of magnitude larger and shows a qualitatively different temperature dependence, with a huge noise increase below  $T_C$ . These results are discussed in terms of a strong coupling of electric resistance and magnetic ordering, as well as within a percolative picture of the ferromagnetic transition.

### I. INTRODUCTION

The mixed-valence manganites  $\text{Ln}_{1-x}\text{D}_x\text{MnO}_3$  (with  $\text{Ln}$  a rare earth and  $\text{D}$  a divalent alkaline earth) recently have attracted renewed interest because of their colossal magnetoresistance (CMR) (Refs. 1 and 2) and the interplay between charge, spin, orbital, and structural degrees of freedom in these materials resulting in interesting ordering phenomena and very rich phase diagrams.<sup>3-5</sup> Both the fundamental mechanism of the CMR effect and its potential for device applications have been addressed in a large number of studies.<sup>6-9</sup>

By changing the dopants at a fixed value of  $x=\frac{1}{3}$  the perovskite structure is modified by what usually is described by the introduction of the tolerance factor  $t$  and the mismatch  $\sigma^2$  of the ionic radii. For large  $\sigma^2$ , or  $t$  deviating significantly from unity, the Mn-O-Mn bonding angle deviates from  $180^\circ$  leading to a reduction of the double exchange transfer integral. This, in turn, may result in large changes of the Curie temperature<sup>10,11</sup> giving the transition temperature between a paramagnetic (PM) semiconducting state above  $T_C$  and a ferromagnetic (FM) metallic state below  $T_C$ . Additionally, the appearance of an insulating spin-glass state may be triggered by either a small tolerance factor  $t\leq 0.907$  (Ref. 12) or a larger tolerance factor  $t=0.934$  together with a large size mismatch  $\sigma^2\geq 2.20$ .<sup>13</sup> This is explained by the competition of the ferromagnetic double exchange and the antiferromagnetic super-exchange interaction<sup>12</sup> or by the random distribution of static Jahn-Teller distorted  $\text{MnO}_6$  octahedra.<sup>13</sup> Furthermore, large relaxation effects may occur close to the Curie temperature related to the percolative conduction between coexisting metallic and insulating phases.<sup>14</sup> Although in the past most attention was focused on models of homogeneous phase transitions, recent experimental data<sup>15-20</sup> have shown that in some parts of the phase diagram of the doped

manganites there may be percolationlike transitions between more and less conductive phases. In general, for such inhomogeneous transitions one expects large resistance fluctuations in the transition regime.

In addition to standard transport measurements the detailed investigation of the  $1/f$  noise is a well established, valuable tool for clarifying the charge transport mechanism in a variety of physical systems.<sup>21</sup> Low-frequency resistance fluctuations have been investigated in manganite thin films<sup>22-30</sup> and single crystals.<sup>29,31</sup> In these samples, the  $1/f$  noise was found to be orders of magnitude larger than in conventional metals or semiconductors and to display a broad spectrum of different temperature dependencies. The resistance fluctuations are intimately related to the conduction mechanism in different temperature regimes. Thus the detailed investigation of the  $1/f$  noise is expected to further clarify the interplay of polaronic charge transport with mainly localized charge carriers in the PM phase and charge transport mediated by double exchange in the FM phase.

In this article we present a detailed study of the temperature and magnetic-field dependence of the low-frequency resistance fluctuations in  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  thin films of high epitaxial quality. Film preparation on different substrates resulting in different amounts of lattice mismatch between film and substrate is found to have a tremendous effect on both the overall temperature dependence and the magnitude of the resistivity and the normalized resistance noise. The strikingly different magnitude and temperature dependence of the  $1/f$  noise most likely originates from the different amount of local strain in the epitaxial films deposited on different substrates.

### II. SAMPLE PREPARATION AND EXPERIMENTAL TECHNIQUES

The  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  (LCMO) films were prepared by laser molecular-beam epitaxy<sup>32,33</sup> on  $\text{SrTiO}_3$  (STO) and

NdGaO<sub>3</sub> (NGO) single-crystal substrates with the (001) plane parallel to the substrate surface. The film thickness was 200 nm for the films on STO and 40 nm for the films on NGO. The as-prepared films have a surface roughness below 1 nm as was confirmed by atomic force microscopy. The structure of the films was analyzed by x-ray diffraction. Rocking curve analysis of the (002) peak yields a small full width at half maximum (FWHM) of  $\leq 0.05^\circ$ . The measured values are similar to the typical FWHM of the substrate peak which ranges between  $0.03^\circ$  and  $0.05^\circ$  for commercial substrates. This shows the high degree of *c*-axis orientation of our films. At present, the mosaic spread of the films seems to be limited by the substrate quality. The microstructure of the films was also studied by high-resolution transmission electron microscopy (HRTEM).<sup>34,35</sup> The TEM analysis showed that the films are of a high epitaxial quality and that the lattice mismatch between the film and the substrate is incorporated into the film.<sup>36,37</sup> That is, the La<sub>2/3</sub>Ca<sub>1/3</sub>MnO<sub>3</sub> films grown on STO are highly strained due to the considerable lattice mismatch of 1.5%, whereas there is very little strain in the films grown on NGO due to the perfect lattice match between LCMO and NGO.

The oxygen content of the films could not be directly determined. An additional *ex-situ* annealing process of a 80-nm-thick La<sub>2/3</sub>Ca<sub>1/3</sub>MnO<sub>3</sub> film grown on a STO substrate in pure oxygen atmosphere was found to result in an increase of the Curie temperature from  $T_C = 120$  K to about  $T_C = 250$  K. This increase of  $T_C$  was accompanied by an increase of the surface roughness of the film to about 8 nm rms and an increase of the FWHM of the rocking curve of the (002) peak by a factor of about 3. This strongly suggests that the observed change in  $T_C$  induced by the annealing procedure is not predominantly related to an oxygen deficit in the as prepared film, but is partly caused by the relaxation of the internal strain. Further details of the film preparation are described elsewhere.<sup>38</sup>

Low-frequency  $1/f$  noise is found in almost all physical systems. Usually it is attributed to material specific mechanisms, therefore to date no universal description has been established.<sup>21,39,40</sup> Intrinsic  $1/f$  noise arising from resistance fluctuations can be quantified by

$$\frac{S_R(f)}{R^2} = \frac{a_R}{\Omega f^\alpha}, \quad (1)$$

where  $S_R$  is the spectral density of the resistance noise,  $\Omega$  the sample volume, and the exponent  $\alpha$  close to unity. The temperature and magnetic-field-dependent quantity  $a_R = a_R(T, H)$  is used to characterize the magnitude of the  $1/f$  resistance fluctuations as a function of temperature and applied magnetic field and is related to the Hooge parameter<sup>21</sup>  $\gamma$  through the density  $n$  of charge carriers as

$$a_R = \frac{\gamma}{n}. \quad (2)$$

In magnetic materials the  $1/f$  resistance noise may be caused by magnetization fluctuations coupled to the resistivity. For example, in metallic magnetoresistive multilayers it has been shown that the  $1/f$  noise can occur from magnetic domain

fluctuations.<sup>41</sup> If the resistivity is a unique function of the net magnetization, then the spectral density of the resistance noise can be expressed as

$$S_R(f) = S_M(f) \left( \frac{dR}{dM} \right)^2, \quad (3)$$

where the spectral density of the magnetization fluctuations  $S_M(f)$  is related to the out-of-phase magnetic susceptibility  $\chi''(f)$  via the fluctuation-dissipation relation as

$$S_M(f) = \frac{2}{\pi} \frac{1}{f} \chi''(f) \frac{k_B T}{\Omega}. \quad (4)$$

Here,  $k_B$  is Boltzmann's constant and  $T$  the sample temperature.

Our noise measurements have been performed in a five-probe configuration.<sup>42</sup> The films have been patterned into microbridges using optical lithography and Ar ion beam etching. Typically, the width and length of the microbridges were 20 and 200  $\mu\text{m}$ , respectively. The sample dimensions have been chosen to optimize the signal to noise ratio of the preamplifier. In order to reduce the contact resistance for the current and voltage leads the La<sub>2/3</sub>Ca<sub>1/3</sub>MnO<sub>3</sub> films were covered *ex situ* with a thin silver or gold layer immediately after film deposition followed by the patterning of the silver/gold layer into contact pads. The resistance fluctuations were investigated by measuring the voltage fluctuations at a constant current bias. Special care was taken for the choice of the load resistors to assure that the current source did not contribute noticeably to the measured voltage noise. The voltage difference between the two branches in the five-probe arrangement was carefully adjusted to zero to maximize the sensitivity for the noise measurements. The voltage fluctuations were amplified by a low-noise amplifier (SR 560) and processed by a digital spectrum analyzer (HP 35665A). Magnetic fields up to 5 T were applied perpendicular to the film surface. Large efforts have been taken for electromagnetic shielding of the sample. Current-voltage characteristics (*IVC*'s) and the resistance vs temperature curves of the samples were measured in the standard four-probe configuration.

The noise spectra usually were taken in a frequency range between 0.1 and 200 Hz. Only for a high sample resistance (up to 8 M $\Omega$ ) it was necessary to reduce the upper frequency limit to about 20 Hz.<sup>43</sup> For all measurements the background noise of the measurement setup was determined for zero bias current and subsequently subtracted from the spectra measured at finite bias current. The noise spectra obtained in this way showed a  $1/f^\alpha$  frequency dependence with  $0.8 < \alpha < 1.3$ . The spectral density of the voltage fluctuations  $S_V$  was found to be proportional to the square of the applied bias current  $I_b^2$  for at least two orders of magnitude. Therefore we are confident that the observed noise originates from  $1/f$  resistance fluctuations.

### III. EXPERIMENTAL RESULTS AND DISCUSSION

#### A. Noise data of a strain free LCMO film

In order to clarify the influence of strain and structural disorder on the noise properties of epitaxial La<sub>2/3</sub>Ca<sub>1/3</sub>MnO<sub>3</sub> thin films we have fabricated films on STO and NGO sub-

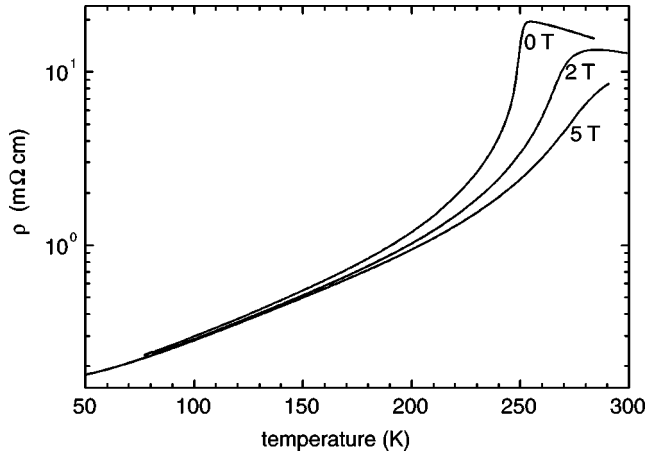


FIG. 1. Temperature dependence of the resistivity of a 40-nm-thick  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film grown on a NGO substrate showing a familiar transition from a semiconducting to a metallic behavior at  $T_C \approx 250$  K in zero applied magnetic field.

strates under identical conditions. We first discuss the noise properties of a 40-nm-thick  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film grown on a perfectly lattice matched NGO substrate.

The temperature dependence of the resistivity of the LCMO film grown on the NGO substrate is shown in Fig. 1 for different values of the applied magnetic field. The curves show the familiar transition from a semiconducting to a metallic behavior which qualitatively can be explained by a transition from a paramagnetic to ferromagnetic state in terms of the double exchange model. In the following the temperature of the resistance maximum in the  $R(T)$  curves is denoted as  $T_{\max}$ , which in zero applied magnetic field is very close to the Curie temperature  $T_C$ . We note that the Curie temperature  $T_C \approx 250$  K of the film is only slightly reduced compared to the highest values obtained for bulk materials (e.g.,  $T_C \approx 270$  K in Ref. 5). The resistivity values of the films are comparable to those of the best bulk materials. The  $IVC$ 's of the microbridges patterned into the films were linear in the whole temperature range (4–300 K) we have investigated.

The temperature dependence of the normalized noise magnitude  $a_R$  is shown in Fig. 2 for different values of the applied magnetic field. We note that the values of  $a_R$  obtained for our high quality thin films are by several orders of magnitude smaller than all data published so far for manganites, both for bulk samples<sup>31</sup> and thin films.<sup>22–30</sup> In particular, we do not observe an increase of the noise level in the ferromagnetic regime. On the other hand, the  $1/f$  noise level is comparable to that of a conventional metal<sup>21</sup> as discussed in more detail below. This result suggests that the low resistance noise level is related to the high epitaxial quality and very low internal strain in the  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film grown on the  $\text{NdGaO}_3$  substrate. This is further supported by the high value of  $T_C$  and the low resistivity of the film. Moreover, a recent study of the effect of structural disorder, oxygen deficiency, and local strain<sup>23,24,27</sup> showed that both the magnitude of the normalized noise  $a_R$  and the resistivity increase dramatically as a function of disorder.

The inset of Fig. 2 shows that in the ferromagnetic regime ( $T < T_C$ ) the ratio  $a_R(T)/R(T)$  is almost constant although both  $a_R$  and  $R$  vary by more than two orders of magnitude in

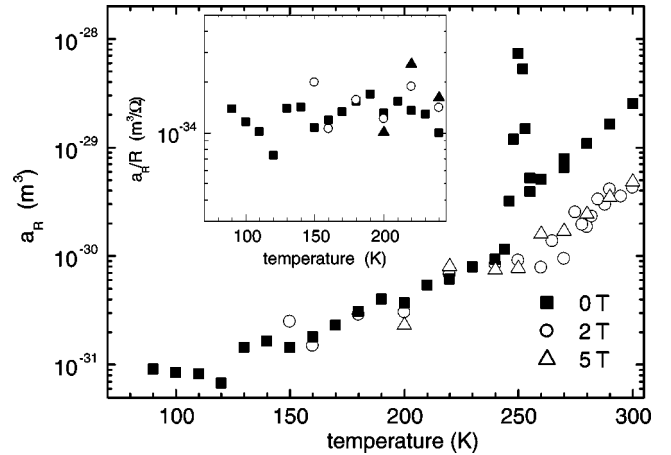


FIG. 2. Temperature dependence of the normalized resistance noise amplitude  $a_R$  of a 40-nm-thick  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film grown on a  $\text{NdGaO}_3$  substrate for different applied magnetic fields. The inset shows  $a_R/R$  as a function of temperature in the ferromagnetic phase.

this temperature range. This clearly suggests that  $a_R(T) \propto R(T)$ . At present, there is not theoretical explanation for this observation.

The zero-field measurements in Fig. 2 show a peak of the low-frequency noise just at  $T_C = 250$  K. This peak in the noise is already completely suppressed by an applied magnetic field of 2 T. We note that we could not observe any noise peak at an elevated temperature close to  $T_{\max}(2\text{ T}) \approx 284$  K. Figure 2 also shows that for zero applied magnetic field there is an increase of the normalized noise amplitude  $a_R$  by a factor of about 5 going from the ferromagnetic ( $T < T_C$ ) to the paramagnetic phase ( $T > T_C$ ). This increase is completely suppressed by an applied magnetic field as can be most clearly seen by comparing the noise amplitude obtained for zero applied magnetic field and 5 T at temperatures above 260 K.

We now discuss possible reasons for the increase of  $a_R$  going from the ferromagnetic to the paramagnetic phase. Evidently, an increase of  $a_R$  can be caused by an additional noise source in the paramagnetic regime which is not directly linked to the magnitude of the resistance, e.g., extended disordered regions adjacent to some small ferromagnetically ordered regions. There is some evidence that the complex magnetic structure in the paramagnetic state can be viewed as consisting of small ferromagnetically ordered clusters embedded into a paramagnetic environment. In this scenario, in addition to defect scattering one may have fluctuations of the domain magnetization or large fluctuations in disordered regions at domain boundaries. The significant reduction of  $a_R$  in the paramagnetic regime by an applied magnetic field, which is discussed below, suggests that the increased noise level in the paramagnetic as compared to the ferromagnetic regime is due to the existence of small ferromagnetically ordered clusters embedded into a paramagnetic environment.

Since we could not observe a peak in the  $a_R(T)$  dependence obtained at an applied magnetic field of 2 T, we directly measured the magnetic-field dependence of  $a_R$  in the paramagnetic regime at a fixed temperature of  $T = 280$  K. The data are shown in Fig. 3. Interestingly, a large decrease of the normalized noise amplitude is observed already at small applied magnetic fields of about 0.2 T. The observation

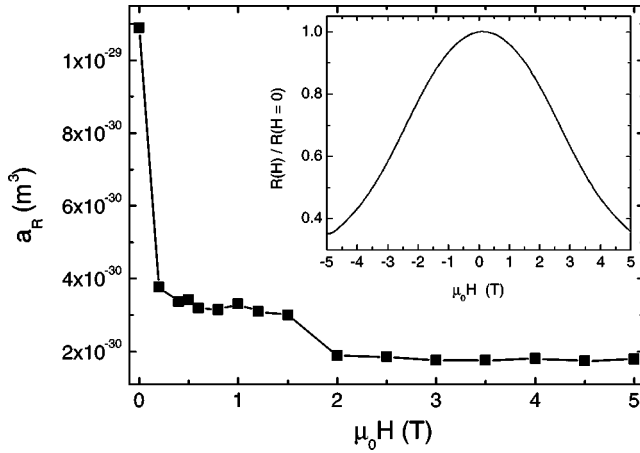


FIG. 3. Magnetic-field dependence of the normalized resistance noise  $a_R$  at  $T=280$  K. The inset shows the magnetic-field dependence of the resistance at the same temperature normalized to  $R(0)$ .

of the noise peak just at  $T_C$  and its suppression by an applied magnetic field most likely is related to an order/disorder transition at the magnetic-phase transition. As the transition temperature  $T_C$  is approached large fluctuating ferromagnetic clusters are formed, which freeze out below the transition temperature giving rise to the formation of ferromagnetic domains. By applying magnetic fields above 2 T the sample enters a homogeneous ferromagnetic metallic phase, which is accompanied by a further reduction of  $a_R(H)$ . By the strong coupling of resistance to magnetization in the doped manganites, these fluctuating spin clusters directly give rise to low-frequency resistance fluctuations. The effect of an applied magnetic field on magnetization fluctuations is to align the spin clusters and suppress spontaneous fluctuations of the relative alignment of neighboring clusters. That is, fluctuations close to  $T_C$  are suppressed significantly by an applied magnetic field as observed in our experiment.

Discussing the normalized resistance noise in the ferromagnetic regime we note that there is only a very weak magnetic-field dependence of  $a_R \propto S_R/R^2$ . The data in Fig. 2 suggest that there is a slight decrease of the  $1/f$  noise with increasing field. In contrast to our data, Raquet *et al.*<sup>28</sup> found a significant decrease of the resistance fluctuations by more than 60% increasing the applied field to 5 T. These authors suggest that by applying a magnetic field domains are aligned and spontaneous fluctuations of neighboring domains are suppressed. From the very weak magnetic-field dependence of our noise data we can conclude that such a mechanism plays a negligible role in our samples and the measured, almost field-independent noise is caused by another mechanism. We also note that the negligible magnetic-field dependence of the normalized resistance noise  $a_R \propto S_R/R^2$  in the ferromagnetic regime (despite the significant field dependence of the resistance) strongly suggests that the measured noise is arising from the same component of the resistance which accounts for the magnetoresistance. If the resistance noise would, e.g., be caused by fluctuations in the interdomain hopping rates but the resistance would arise primarily from other processes, it would be very surprising that  $a_R \propto S_R/R^2$  would stay constant while  $R(H)$  changes by more than an order of magnitude upon increasing the applied field from zero to 5 T.<sup>22</sup>

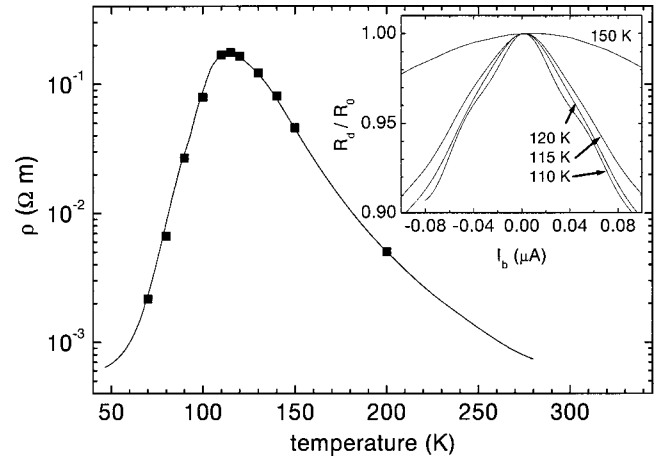


FIG. 4. Temperature dependence of the resistivity of a 200-nm-thick  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film on a  $\text{SrTiO}_3$  substrate measured with a bias current of  $I=0.1 \mu\text{A}$ . In the inset the differential resistance  $R_d = \partial V/\partial I$  normalized to  $R_0 = R_d(I_b=0)$  is shown for temperatures close to  $T_{\text{max}}$ .

It is instructive to do a quantitative comparison of the magnitude of the resistance noise measured in the ferromagnetic phase of the doped manganites to that found in conventional metals. In order to do so, a charge-carrier density of  $n = 5 \times 10^{21} \text{ cm}^{-3}$  ( $\frac{1}{3}$  hole per unit cell) is assumed.<sup>28,23,24</sup> Using this value for  $n$  we obtain a Hooge parameter of  $\gamma \approx 8 \times 10^{-2}$  for the investigated  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film at room temperature. This value is close to what is observed in conventional metals.<sup>21</sup> From this and from the negligible magnetic-field dependence at temperatures below the transition we can conclude that for the ferromagnetic/metallic phase of the investigated  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film there is no significant contribution to the low-frequency resistance noise due to fluctuations in the magnetic order. We also emphasize that the derived Hooge parameter of  $\gamma \approx 8 \times 10^{-2}$  is, to our knowledge, the lowest value published so far for doped manganites. This demonstrates that high quality thin films with very little strain and disorder can have low resistance noise comparable to those of conventional metals with no increase in the ferromagnetic regime.

### B. Noise data of a highly strained LCMO film

We now turn to the noise data obtained for a  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film grown under identical conditions on a  $\text{SrTiO}_3$  substrate. Due to the considerable lattice mismatch between the film and the substrate (1.5%) this film has significantly different transport and noise properties. As shown in Fig. 4, due to the significant strain effect,<sup>34,35</sup> the film is characterized by a much lower value of  $T_{\text{max}} \approx 115$  K and a resistivity that is about 1 order of magnitude larger than that of the film grown on the  $\text{NdGaO}_3$  substrate in the paramagnetic regime at room temperature and 3 orders of magnitude larger in the ferromagnetic regime at 50 K. Furthermore, in contrast to the film on  $\text{NdGaO}_3$  the IVC's of the microbridge patterned into the  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film grown on  $\text{SrTiO}_3$  were found to be nonlinear in a temperature range close to  $T_{\text{max}}$ . As shown in the inset of Fig. 4, the differential resis-



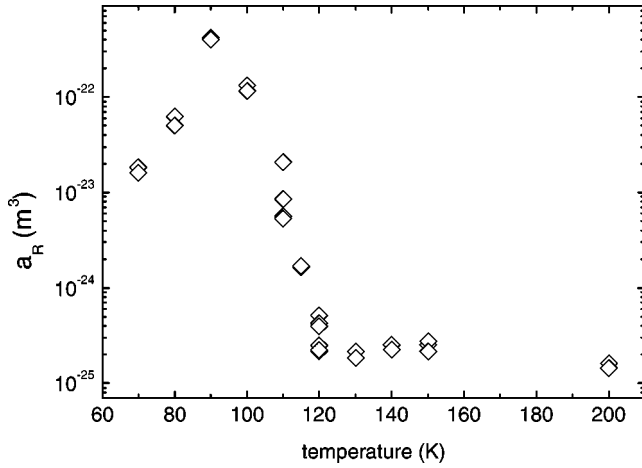


FIG. 5. Temperature dependence of the normalized noise amplitude  $a_R$  of a 200-nm-thick  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film grown on a  $\text{SrTiO}_3$  substrate showing a huge increase of the noise level in the vicinity of the transition from the paramagnetic/semiconducting to the ferromagnetic/metallic phase.

tance  $R_d = \partial V / \partial I$  slightly decreases with increasing bias current  $I_b$ . For temperatures below 100 K and above 200 K the  $IVC$ 's were linear.

There is a strong difference both in the magnitude and the temperature dependence of the normalized low-frequency resistance noise between the  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film grown on  $\text{SrTiO}_3$  and  $\text{NdGaO}_3$ . Whereas for the film on  $\text{NdGaO}_3$  there is a reduction of the resistance noise going from the paramagnetic/semiconducting to the ferromagnetic/metallic regime, the opposite is observed for the film grown on  $\text{SrTiO}_3$ . As shown in Fig. 5, on decreasing temperature, a huge increase of the magnitude of the normalized resistance noise  $a_R$  is observed just in the vicinity of the paramagnetic to ferromagnetic transition. We emphasize that this strong increase of the normalized noise by about 3 orders of magnitude takes place in a relatively wide temperature regime extending over about 40 K. The maximum value of  $a_R$  is measured at about 80 K, that is, at a temperature where the ferromagnetic/metallic phase is expected to be already well established with  $R(80 \text{ K})/R_{\text{max}}(115 \text{ K}) = 4\%$ . We also note that the noise amplitude measured at a specific temperature could not be well reproduced. Repeated measurements at a fixed temperature of  $T = 80 \text{ K}$  showed variations of the noise amplitude of about 20%. Therefore we suspect that a non-Gaussian component of the noise may be present, as reported recently by Hardner *et al.*<sup>22</sup> for a  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film ( $T_{\text{max}} = 110 \text{ K}$ ) displaying similar properties such as a broad peak in the temperature dependence of the resistance noise with a maximum at about  $T = 80 \text{ K}$ .

Discussing the origin of the noise properties of the  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film grown on  $\text{SrTiO}_3$  we have to consider different scenarios. The observed huge increase of the normalized resistance noise  $a_R$  suggests that the resistance in the temperature range near the transition from the paramagnetic to the ferromagnetic phase is mostly determined by charge hopping between domains in an inhomogeneous, disordered magnetic material.<sup>22</sup> Such a transport mechanism is a likely source for both the noise peak and the nonlinearity of the current-voltage characteristics. Low-frequency fluctuations

of domains are known to give rise to an anomalous high normalized low-frequency noise level. We note that a  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film with an intermediate transition temperature  $T_{\text{max}} = 180 \text{ K}$  studied by Alers *et al.*<sup>25</sup> showed a less pronounced increase of the noise at the phase transition by only two orders of magnitude. Furthermore, the peak was found to be restricted to a narrower temperature range of less than 40 K around the phase transition.

Beyond the explanation of the noise data in terms of a disordered magnetic material, there is another possible scenario. Podzorov *et al.*<sup>31</sup> interpret the noise properties of their polycrystalline samples and single crystals with low transition temperature as evidence for a percolative nature of the charge transport in the ferromagnetic regime (see also Refs. 29 and 30). In the noise data of their bulk samples an increase of the absolute value of noise level of up to 10 orders of magnitude over a broad temperature range has been observed. Their scaling analysis of the  $1/f$  noise is claimed to be consistent with a percolation model of conducting domains randomly distributed in an insulating matrix. In these experiments an increase of the normalized noise magnitude by more than 4 orders of magnitude was observed. Further evidence for a percolation nature of the phase transition comes from the large width of the noise peak.

### C. Comparison of the noise data

In order to further clarify the origin of the observed noise properties we compare the noise data obtained for the  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  films grown on  $\text{NdGaO}_3$  and  $\text{SrTiO}_3$  substrates. Evidently, both films have completely different noise properties both regarding the absolute magnitude of the noise and its temperature dependence. Since both films have been prepared under identical conditions, there is strong evidence that the different amount of strain in the films causes the observed differences.

The noise data of the unstrained  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film grown on  $\text{NdGaO}_3$  can be interpreted in a straightforward way. In the ferromagnetic/metallic regime the noise properties are comparable to those of a conventional metal demonstrating that in the high quality film magnetization fluctuations are negligible. In the paramagnetic/semiconducting regime the noise is increased and shows a peak at  $T_C$ , which is suppressed completely by an applied magnetic field of 2 T. This most likely is caused by the suppression of magnetization fluctuations by an applied magnetic field in the paramagnetic regime.

Obviously, the strain introduced in the  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film grown on  $\text{SrTiO}_3$  due to the large lattice mismatch of 1.5% results in a strongly increased noise level both in the paramagnetic and ferromagnetic regime with a huge enhancement in the ferromagnetic regime (almost 10 orders of magnitude as compared to the strain free film). It is known that strain due to a large lattice mismatch between film and substrate in the way as a large tolerance factor<sup>31</sup> causes significant local structural disorder in the doped manganites. Keeping in mind this strain effect in the  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film on  $\text{SrTiO}_3$ , the huge increase of  $a_R$  in the ferromagnetic regime within a broad temperature range most likely is associated to domain fluctuations well known for disordered magnetic materials. An abrupt increase of the normalized

magnitude of the  $1/f$  noise by 1–2 orders of magnitude at magnetic transitions has been observed for many macroscopically homogeneous magnetic materials<sup>44,45,41</sup> and recently has been found also for thin films and bulk samples of the doped manganites.<sup>22,25,31</sup> That is, strain in the film is responsible for defect fluctuations which couple to the local magnetization and thus cause resistance fluctuations as proposed recently by Alers *et al.*<sup>25</sup>

Using  $n = 5 \times 10^{21} \text{ cm}^{-3}$  as for the  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film on  $\text{NdGaO}_3$  we derive a Hooge parameter  $\gamma \approx 10^3$  at  $T = 200 \text{ K}$  and  $\gamma = 10^6$  at  $T = 90 \text{ K}$  for the  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  film grown on  $\text{SrTiO}_3$ . The fact that the Hooge parameter is by more than 5 orders of magnitude larger in the strained low- $T_C$  than in the strain free high- $T_C$  sample further illustrates the amount of disorder introduced by the lattice mismatch between film and substrate. So far, we have no direct proof that the observed magnetic disorder bears similarities with a spin glass. However, the interplay between the strongly disordered magnetic domains can be interpreted as an ensemble of ferromagnetic clusters in a paramagnetic matrix, a model that has been discussed recently by several authors.<sup>46,47</sup> Within this model the low-frequency dynamics show many features which are similar to a spin glass. Interestingly, there seems to be a correlation between the transition temperature  $T_{\text{max}}$ , the increase of the noise level, and the temperature range over which this increase takes place: the lower  $T_{\text{max}}$  is the stronger is the increase of the noise level and the wider is the temperature range over which the increase takes place.

#### IV. CONCLUSIONS

We have performed a comparative study of the low-frequency resistance noise in epitaxial  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  films

with different amounts of strain. In a strain free film grown on  $\text{NdGaO}_3$  with a high Curie temperature a very low normalized resistance noise  $a_R$  was found comparable to that found for conventional metals. The noise peak measured close to  $T_C$  in zero field could be completely suppressed by an applied magnetic field. This can be understood in the framework of a conventional ferromagnetic metal by taking into account the coupling of magnetic and resistance fluctuations. For a highly strained film grown on  $\text{SrTiO}_3$  with a low Curie temperature the normalized resistance noise was found to be enhanced by several orders of magnitude. In addition, a broad peak in the temperature dependence of the resistance noise in the vicinity of  $T_C$  was found. This behavior can be explained in terms of charge hopping between domains in an inhomogeneous, disordered magnetic material. Our comparative study shows that there is a strong coupling between local magnetic disorder and structural disorder introduced by strain effects due to a large lattice mismatch between film and substrate. We note that the strain due to lattice mismatch between film and substrate may have a similar effect as the internal local strain in bulk samples of the doped manganites caused by the mismatch of the ionic radii of different dopants. Judging from our noise analysis, the low- $T_C$  film shows strong magnetic disorder, whereas the high- $T_C$  film behaves more like a conventional ferromagnetic metal.

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\*Present address: Laboratoire Physico-Chimie des Solides, Université Paris-Sud XI, 91405 Orsay Cedex, France.

†Electronic address: Achim.Marx@wmi.badw.de

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