Direct correlation between $1/f$ magnetonoise and magnetoresistance in $La_{0.7}Sr_{0.3}MnO_3$ and $(La_{0.5}Pr_{0.2})Ba_{0.3}MnO_3$ manganites

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Temperature- and magnetic-field-dependent electrical noise and electrical resistivity measurements were carried out on epitaxial thin films of a large bandwidth $La_{0.7}Sr_{0.3}MnO_3$ and a disordered intermediate bandwidth $(La_{0.5}Pr_{0.2})Ba_{0.3}MnO_3$ manganite system. The power spectral density was dominated by $1/f$ noise. This $1/f$ noise was observed to follow the overall temperature dependence of the resistivity. Moreover, in these compounds the magnetonoise effect was found to be of nearly the same magnitude as the magnetoresistance. This direct correlation between magnetonoise and magnetoresistance suggests that the enhanced $1/f$ noise has its origin in intrinsic charge-carrier density fluctuations.

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Noise measurements are an important tool to understand various underlying transport mechanisms in different classes of metallic and semiconducting materials. Fluctuations in the charge transport of condensed-matter systems arise from various relaxation processes of the charge carriers, defects, groups of defects, etc.; see Refs. 1 and 2 for an overview. The noise power spectral density S_V in general shows a $1/f^\alpha$ dependence, where f is the frequency of the fluctuation and α is an exponent close to unity, and is referred to as $1/f$ noise.[1,2] Among various oxides, mixed-valent manganites are a class of modern era technological materials that are rich in exhibiting exotic electronic and magnetic phases in the temperature–magnetic-field phase diagram.[3,4] Previous work has established that $1/f$ electrical noise in mixed-valent manganites showing colossal magnetoresistance is maximal in the vicinity of the metal-insulator transition temperature.[5–10] In addition, phase separation in manganites was indicated by measurements of random telegraph noise, where a continuous switching between insulating and metallic phases near the metal-insulator transition temperature shows the dynamic nature of phase separation processes in these compounds.[11,12]

Near the Curie temperature, phases with different conductivities and/or carrier concentrations exist in manganites. Recently, vibronic phase segregation into hole-rich, itinerant and hole-poor, polaronic regions was reported in orthorhombic $La_{2/3}(Ca_{1-x}Sr_x)_{1/3}MnO_3$.[13] This type of phase separation is driven by the coupling of the charge carriers to Jahn-Teller distortions. Another kind of charge-carrier fluctuations was discussed by Alexandrov and Bratkovsky.[14,15] In this case, the charge carriers are supposed to be polarons of oxygen p-character that are bound into heavy immobile bipolarons near the transition temperature. This mechanism leads to a strong temperature- and field-dependent polaron density that determines the behavior of the resistivity.[14,16] However, optical spectroscopy studies failed to find evidence of bipolaron formation, whereas small polaron formation in $La_{0.7}Ca_{0.3}MnO_3$ and large polaron formation in $La_{0.7}Sr_{0.3}MnO_3$ was observed.[17] Furthermore, the significance of quenched disorder has been pointed out[18] and the existence of a Griffith's phase has been reported.[19,20] Quenched disorder can also be incorporated in the bipolaron model leading to phase segregation into itinerant polaron and immobile bipolaron phases near the Curie temperature.[15] At constant temperature, the application of a magnetic field drives the electronic system toward an itinerant state with an increasing mobile carrier density. The susceptibility of the carrier concentration to magnetic fields is highest near the Curie temperature. Experimentally, a carrier density change as a function of magnetic field near the transition temperature is difficult to detect with conventional Hall-effect measurements, since high fields are necessary to saturate the anomalous Hall effect.[21] Here an alternative method for the study of charge-density variations is proposed.

In spite of the fact that several studies on electrical noise in manganites found evidence of a correlation between the $1/f$ noise and the magnitude of the resistivity as a function of temperature, the influence of the magnetic field on the $1/f$ noise and its correlation with the magnetoresistance has not been clearly demonstrated experimentally.[5–10] Magnetic-field-dependent studies, however, would be suitable to further elucidate the origin of the $1/f$ noise in the manganites, since it would be possible to further characterize the crossover from localized to itinerant charge carriers. In this paper, we report magnetic field- and temperature-dependent $1/f$ electrical noise and resistivity studies on two distinct manganites, namely an intermediate e_g-electron bandwidth A-site disordered manganite of composition $(La_{0.5}Pr_{0.2})Ba_{0.3}MnO_3$ (LPBMO) and a standard large e_g-electron bandwidth manganite, $La_{0.7}Sr_{0.3}MnO_3$ (LSMO). These samples have peak resistivity temperatures of $T_p \simeq 200$ K in the case of LPBMO

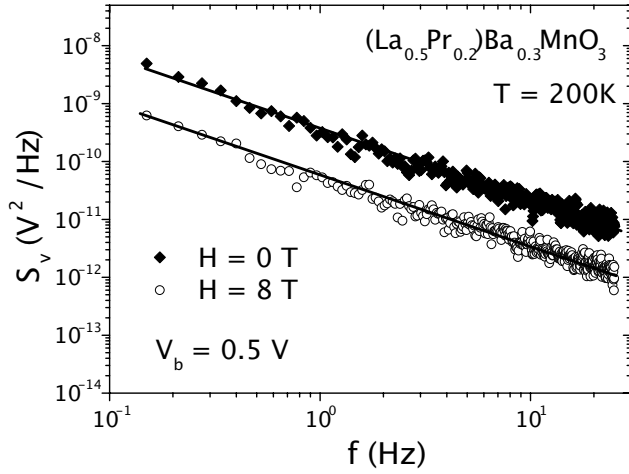FIG. 1. Frequency f dependence of the noise power spectral density S_V for the $(La_{0.5}Pr_{0.2})Ba_{0.3}MnO_3$ film in magnetic fields $\mu_0 H$ of 0 and 8 T at a temperature of 200 K. The bias voltage was 0.5 V.

and $T_p \simeq 375$ K in the case of LSMO. In spite of the large difference in the peak temperatures of the two systems, we show that the magnetonoise and magnetoresistance are of nearly the same magnitude and that the $1/f$ noise follows nearly the same trend as the electrical resistance. This indicates that the $1/f$ noise and the electrical resistance may have the same physical origin.

Epitaxial thin films of $(La_{0.5}Pr_{0.2})Ba_{0.3}MnO_3$ (LPBMO) and $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) were deposited on (001) oriented $LaAlO_3$ single-crystal substrates using pulsed laser deposition (PLD) with a KrF excimer laser. The details of the synthesis and other transport properties of the LPBMO film (thickness $\simeq 200$ nm) were reported elsewhere.[22,23] The film investigated here was grown at optimized deposition parameters as described in Ref. [23], i.e., a fluence of ~ 3.1 J/cm^2, a growth rate of 0.33 nm/s, a substrate temperature of 830 °C, an O_2 partial pressure of 400 mTorr, and a substrate to rotating target distance of 4.2 cm. The LSMO film (thickness $\simeq 150$ nm) was deposited at a substrate temperature of 700 °C in an oxygen partial pressure of 0.13 mbar.[24] The LPBMO and LSMO thin films were found to be (101) and (001) oriented, respectively. Electrical resistivity, as a function of temperature and magnetic field, was measured in a dc four-probe configuration using a Physical Property Measurement System (PPMS, Quantum Design). An ac five probe method[25] was used for the noise measurement. A voltage signal with a frequency of 679 Hz was fed to the sample from the internal oscillator of a lock-in amplifier (Stanford Research Systems, SRS, Model SR830). The compensated signal from the sample was demodulated and amplified by the same lock-in amplifier using a time constant of 1 ms and a band-pass filter setting of 24 db. The analog signals of both the in-phase and out-of-phase component were fed into a dual channel spectrum analyzer (SRS, Model SR785) and the power spectral density was measured in the frequency range between 100 mHz and 25 Hz. The in-phase noise was always considerably larger than the out-of-phase (background) noise. The measurements were performed in a He flow cry-
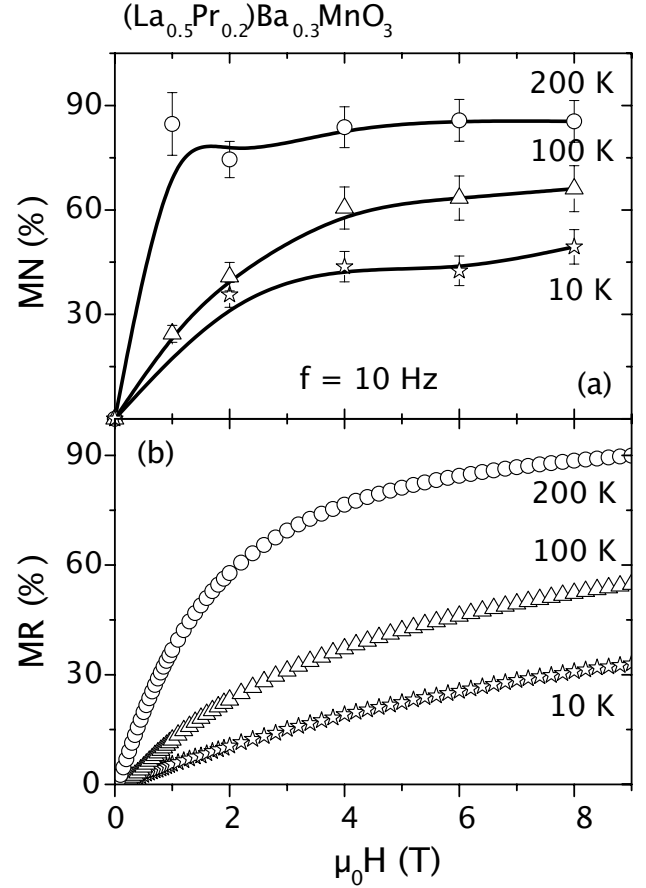

FIG. 2. (a) Magnetonoise (MN) at a frequency of 10 Hz and (b) magnetoresistance (MR) as a function of magnetic field for the $(La_{0.5}Pr_{0.2})Ba_{0.3}MnO_3$ thin film at 200, 100, and 10 K. The solid lines in (a) are guides to the eye.

ostat equipped with a superconducting solenoid up to 9 T. Measurements of the voltage dependence of the power spectral density were performed and a quadratic dependence of the $1/f$ noise on bias voltage was found. This shows that the recorded noise power spectral density is intrinsic to the samples.[9]

Figure [1] shows a typical frequency dependence of the power spectral density S_V in zero magnetic field and in a field of 8 T for the LPBMO film at a temperature of 200 K, i.e., in close vicinity of its peak resistivity temperature. Fits of a power law to the power spectral density show an $S_V \propto 1/f^\alpha$ dependence with an exponent $\alpha \simeq 1$. Additionally, it is noteworthy that there is a large suppression of S_V under the application of a magnetic field. This reveals a large sensitivity of the $1/f$ noise to the magnetic field near T_p. In order to correlate the magnetic-field dependence of the $1/f$ noise and the electrical resistance, the magnetonoise

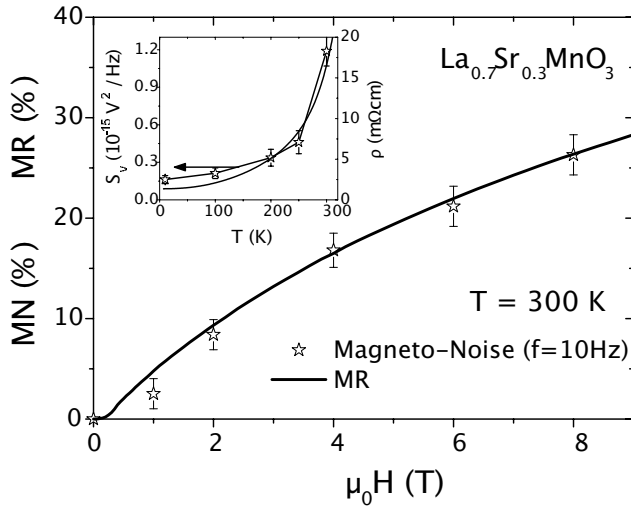$$\frac{S_V(0) - S_V(H)}{S_V(0)} \tag{1}$$

and the magnetoresistance

FIG. 3. Magnetonoise (MN) at a frequency of 10 Hz and magnetoresistance (MR) as a function of magnetic field for the La$_{0.7}$Sr$_{0.3}$MnO$_3$ thin film at 300 K. The left and the right scale of the inset show the temperature dependence of the noise power spectral density S_V and the electrical resistivity ρ, respectively.

$$\frac{R(0) - R(H)}{R(0)} \tag{2}$$

are shown in Fig. 2 as a function of magnetic field at various temperatures for the intermediate bandwidth LPBMO film. It is seen that the magnetonoise is nearly of the same magnitude as the magnetoresistance at all the temperatures investigated and the overall behavior of the magnetonoise (shown at 10 Hz) and the magnetoresistance is similar. Magnetonoise curves at other frequencies show the same field dependence, since the exponent α is independent of field. The magnetoresistance and magnetonoise for the large bandwidth LSMO film are shown in Fig. 3 at a temperature of 300 K. The agreement between the field-dependent MN and MR curves for the two effects is striking. These observations point toward a common origin of these two properties. This is a novel feature of the transport properties of the manganites that has not yet been elucidated in all the previous magnetic-field-dependent noise studies.[7–10] For instance, the magnetic-field-dependent 1/f-noise studies on La$_{0.7}$Sr$_{0.3}$MnO$_3$ by Raquet *et al.*[9] show a magnetonoise effect of ~60% versus a magnetoresistance of ~15% in a field of 5 T at 300 K; in La$_{0.7}$Ca$_{0.3}$MnO$_3$ films a suppression of the noise peak near the metal-insulator transition followed by a saturation of the magnetonoise in fields as low as 2 T was observed.[10]

In order to obtain more information on the origin of the direct correlation of the field-dependent sensitivity of the 1/f noise and the field dependence of the electrical resistance, a detailed correlation of the temperature-dependent 1/f electrical noise in zero field with the zero-field resistance of the LPBMO sample has been experimentally established from the data presented here. The noise is lowest in the semiconducting and paramagnetic region above T_C, whereas it peaks in the vicinity of the metal-insulator transition. More pre-
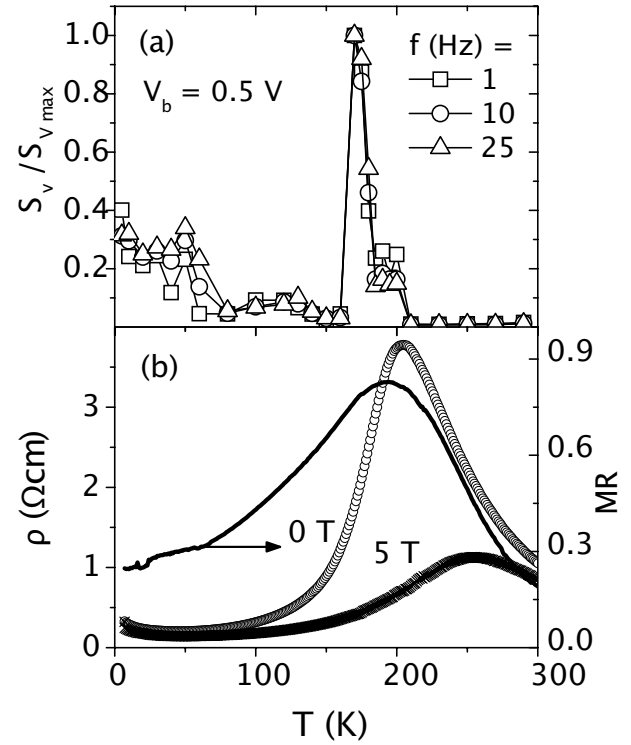

FIG. 4. (a) Noise power spectral density S_V normalized to its maximum values vs temperature at frequencies of 1, 10, and 25 Hz for the (La$_{0.5}$Pr$_{0.2}$)Ba$_{0.3}$MnO$_3$ film. (b) Temperature dependence of the electrical resistance R and magnetoresistance (MR) in a field of 5 T.

cisely, the noise is found to be highest at a temperature where the magnetoresistance and the first derivative of the zero-field resistivity are maximum, just below the peak resistivity temperature. The noise is higher in the metallic region as compared to that in the region above T_p. Further, in the metallic region, the noise continues to rise as the temperature decreases, which is similar to the low-temperature rise in resistivity, thus indicating a simultaneous low-temperature evolution of additional sources of noise and resistivity. The noise power spectral density S_V at 1, 10, and 25 Hz, the resistance, and MR are plotted as a function of temperature in Fig. 4. The scaled power spectral densities $S_V/S_{V_{max}}$ agree rather well and indicate the scatter in the data. Similar behavior is observed in the LSMO film; see the inset to Fig. 3. Throughout the temperature range investigated, for both LSMO and LPBMO samples, the noise power spectral density as a function of frequency obeys a power law $S_V \propto 1/f^{\alpha}$ with $0.9 < \alpha < 1.25$ both in the absence and presence of an applied magnetic field.

Despite the fact that the magnitude of noise in LPBMO is rather large as compared to that in LSMO, both of these samples exhibit qualitatively similar relations between the 1/f noise and the electrical resistivity. In the absence of any detailed theory of 1/f noise in manganites, here very general expressions to quantify the parameter dependence of the noise and the resistivity will be used. The power spectral density is given by Hooge's law,[26]

$$S_V = \gamma \frac{V^2}{n\Omega} \frac{1}{f^\alpha}, \tag{3}$$

as a function of the voltage V applied across the sample, the number n of carriers, the sample volume Ω, and the Hooge parameter γ that characterizes the strength of noise sources. For the resistivity, the Drude expression is used, since this represents a general principle, namely the factorization into a carrier density and a mobility,

$$\rho = \frac{m^*}{ne^2\tau} \tag{4}$$

with the effective mass m^*, electronic charge e, and relaxation time τ.

At first the zero-field values are discussed and estimates for the Hooge parameter γ are derived to facilitate comparison to previous work on $1/f$ noise in manganites. Within a factor of 3–5, Hall effect measurements[21,27,28] indicate a carrier concentration $n \sim 5 \times 10^{21}$ cm^{-3} for an optimally hole-doped system. Since this value is inferred from Hall measurements in a finite field, the zero-field carrier concentration might actually be smaller than this value. However, in order to obtain an order of magnitude estimate, this carrier concentration is used in the following. A value of $\gamma \simeq (1-2) \times 10^6$ is estimated at 200 K for the LPBMO film and $\gamma \simeq 700-1000$ at 300 K for the LSMO film. Both values are much larger than typical values[1,2] for metallic systems of $\gamma \simeq 10^{-3}$. Although γ for LPBMO is very large compared to that observed in metals and semiconductors, it is of the same order as that reported for $Pr_{0.67}Sr_{0.33}MnO_3$ and $La_{0.67}Ca_{0.33}MnO_3$ films.[6] On the other hand, γ for the present LSMO film grown on LaAlO$_3$ is almost an order of magnitude larger than that observed in LSMO grown on MgO.[9] This discrepancy might indicate a higher defect and dislocation density in the LSMO film on LaAlO$_3$, although the lattice mismatch between LSMO and MgO is larger than that between LSMO and LaAlO$_3$. This better film quality would, however, be consistent with other reports on the high quality of manganite films grown on MgO.[29] The large difference in γ of the two samples in the present study indicates additional noise sources in the A-site disordered LPBMO film which might be related to inherent local structural distortions, defects, and

dislocations. In addition to this, its low T_p of 200 K as compared to 375 K for LSMO suggests that there exists large spin disorder at T_p and below in LPBMO, which might additionally contribute to the $1/f$ noise.

The magnetic-field dependence of the magnetonoise might according to Eq. (3) arise from the field dependence of Hooge's constant γ or the charge-carrier concentration n, and in the case of the magnetoresistance, see Eq. (4), from the field dependence of n or the relaxation time τ. The agreement between magnetoresistance and magnetonoise strongly indicates that the carrier density is actually the crucial parameter. This means that the magnetonoise and magnetoresistance curves should be interpreted as curves showing the change of the inverse carrier concentration with field. This conclusion is partially supported by evidence from Hall-effect measurements reporting a field-independent Hall mobility[30] and a charge-carrier density minimum in the vicinity of the Curie temperature.[21] It is likely that these thin films contain significant levels of quenched disorder such that two competing phases are present in the samples. The application of a magnetic field leads to the growth of the itinerant phase having a higher carrier density.

In conclusion, we have observed a direct correspondence between magnetonoise and magnetoresistance effects in two largely distinct manganite systems, namely $(La_{0.5}Pr_{0.2})Ba_{0.3}MnO_3$ and $La_{0.7}Sr_{0.3}MnO_3$. Using general arguments on the dependence of the noise and resistivity on the carrier concentration, these observations establish that the $1/f$ electrical noise and the electrical resistivity originate from charge-carrier density fluctuations. This result is in agreement with both main scenarios discussed in the introduction, i.e., charge-carrier segregation into polaronic and itinerant phases or the bipolaron model. We hope that this method might be further refined for detailed studies of the charge-carrier density evolution with magnetic field, temperature, and doping. An analysis of noise processes within the aforementioned theoretical models would be desirable for further understanding.

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