1/*f* electrical noise in epitaxial thin films of the manganite oxides $La_{0.67}Ca_{0.33}MnO_3$ and $Pr_{0.67}Sr_{0.33}MnO_3$

M. Rajeswari,[a] A. Goyal,[b] A. K. Raychaudhuri,[a] M. C. Robson, G. C. Xiong, C. Kwon, R. Ramesh, R. L. Greene, and T. Venkatesan[b]
Department of Physics, Center for Superconductivity Research, University of Maryland, College Park, Maryland 20742

S. Lakeou
Department of Engineering and Technology, University of the District of Columbia, Washington, DC 20008

We report measurements of *1/f* electrical noise in two hole doped manganite perovskite oxides, $La_{0.67}Ca_{0.33}MnO_3$ and $Pr_{0.67}Sr_{0.33}MnO_3$, which exhibit colossal magnetoresistance. The noise magnitude represented by the Hooge parameter is nearly 8 orders of magnitude larger than that observed in ordinary metals (and semiconductors) and nearly 5–6 orders of magnitude larger than that observed in epitaxial films of the perovskite oxide $YBa_2Cu_3O_7$ in the normal state. The normalized noise spectral density increases with decreasing temperature below the resistivity peak, suggestive of the presence of additional low energy noise processes in the ferromagnetic metallic state.

The colossal magnetoresistance in the perovskite oxides of the type $A_{(1-x)}A'_{(x)}MnO_3$ (A=La, Pr, Nd) (A'=Ca, Ba, Sr) is currently a topic of active research.[1–5] The CMR oxides are characterized by a peak in the temperature dependence of resistivity resembling a metal-insulator transition, the peak occurring at a temperature T_p close to the ferromagnetic transition temperature T_c. The magnitude of the magnetoresistance (MR) shows a maximum near the resistivity peak. The CMR oxides are being actively pursued for the development of magnetoresistive read heads and other magnetic field sensors. Recently, these materials have also attracted interest for infrared detector applications based on the large temperature coefficient of resistance (TCR) in the vicinity of the resistivity peak.[6]

For both bolometric and magnetoresistive applications, an important factor to be considered is the nature of resistance fluctuations (electrical noise) which could limit the signal to noise ratio. There are several noise sources that need to be considered such as Johnson noise, *1/f* excess noise and Barkhausen noise. While the Johnson noise depends only on the device resistance, temperature and bandwidth, the excess *1/f* noise has an intrinsic material dependence via the nature of the defect fluctuations or other physical processes contributing to the resistance fluctuations.[7,8] We have studied the excess noise in two CMR oxides. The results reveal a large noise magnitude as well as an anomalous temperature dependence of the noise power spectral density.

Here we report our studies on two materials: $La_{0.67}Ca_{0.33}MnO_3$ (LCMO) and $Pr_{0.67}Sr_{0.33}MnO_3$ (PSMO). The samples were (001) oriented thin films deposited on (001) $LaAlO_3$ by pulsed laser deposition. The substrate temperature was maintained at 625 °C and the oxygen pressure was 400 mTorr during the deposition. After deposition the samples were cooled down to room temperature in 300 Torr oxygen.

The samples were characterized for structural quality by 4-circle x-ray diffraction studies. θ-2θ measurements indicate (100) oriented single phase material. Rocking angle analysis shows a full width at half-maximum (FWHM) of 0.2°–0.3° indicating a high degree of *c*-axis orientation. Φ scan analysis of the (101) peak indicates epitaxial in-plane alignment with the substrate. It may be mentioned here that the high crystalline quality of the films revealed by the above studies is of relevance to the noise studies since structural defects and inhomogeneities are known to lead to enhanced noise levels in other metallic oxides.[9]

For noise measurements, 200 nm thick films were patterned into 125 μm wide and 2 mm long bridges in the 4-probe configuration with evaporated silver as contact pads and soldered gold wires as leads. The noise in the frequency range 1–100 Hz was measured with a dc bias current (1–5 mA) through the sample. Details of the measurement system are described in Ref. 9.

In Fig. 1 we present the bias current dependence of the noise in LCMO at 100 and 295 K. The frequency dependence is shown in the inset for PSMO at 150 and 270 K. The noise spectral density S_v is quadratic in the bias current and shows a $1/f^{\alpha}$ dependence on frequency (with $\alpha \sim 1$) both in the ferromagnetic metallic state and the paramagnetic insulating state. These dependencies are characteristic of conductance fluctuations.[4] To parametrize the observed noise, we have used Hooge's empirical relation for conductance fluctuations, which has been employed in the analysis of *1/f* noise in a wide range of materials.[6,7,10] We would like to emphasize at the outset that Hooge's equation is empirical and we use it only for normalizing the noise data in a common framework in order to compare the noise magnitudes in different material systems. Hooge's relation is given by

$$S_v(f)/V_{dc}^2 = \gamma/(n_c^* v^* f^{\alpha}), \tag{1}$$

where V_{dc} is the dc voltage across the sample, n_c is the

[a]Also at the Engineering and Technology Department, University of District of Columbia, Washington, DC 20008.
[b]Department of Physics, Indian Institute of Science, Bangalore, India.
[c]Also at the Electrical Engineering Department, University of Maryland, College Park, MD 20742.

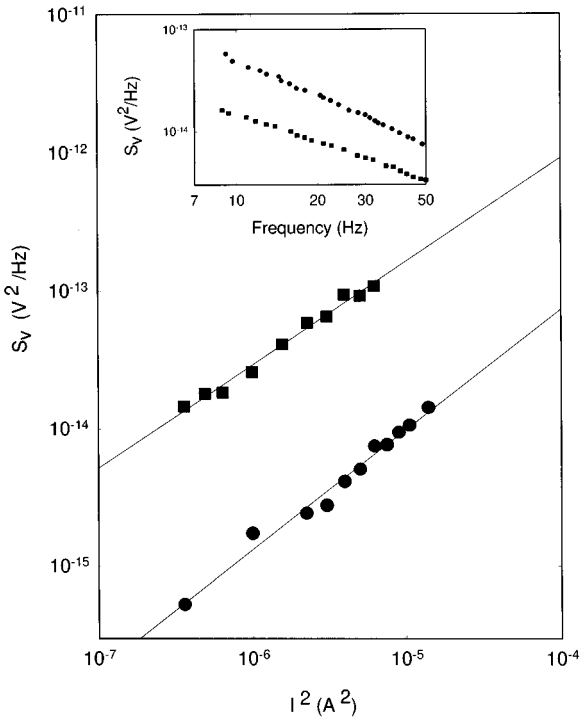

FIG. 1. Bias current dependence of the noise power spectral density S_v for $La_{0.67}Ca_{0.33}MnO_3$ at $T=295$ K (●) and $T=100$ K (■) at $f=14$ Hz. Inset: Frequency dependence of the noise power spectral density S_v for $Pr_{0.67}Sr_{0.33}MnO_3$ at 270 K (●) and 150 K (■).

volume density of charge carriers in the sample, v is the sample volume, f is the frequency, and the Hooge parameter γ represents the strength of the noise sources. From Eq. (1), the normalized noise spectral density given by $S_v^* v^* f/V_{\rm dc}^2$ represents γ/n_c.

In Figs. 2(a) and 2(b) we plot the normalized noise power spectral density S_v at $f=11$ Hz as a function of temperature for LCMO and PSMO, respectively. The temperature dependence of resistance in the same temperature range is shown in the insets. In Fig. 3 we show the temperature dependence of the unnormalized S_v for LCMO. Resistance peaks occur at $T_p \sim 200$ and 160 K for LCMO and PSMO, respectively. The noise data are interesting both in terms of the magnitude as well as the temperature dependence. First we will discuss the magnitudes. From the normalized noise power spectral density γ/n_c, we obtain Hooge parameter $\gamma \sim 10^5$ and 10^6 at 295 K for LCMO and PSMO, respectively. (We use $n_c = 5\times 10^{21}/\text{cm}^3$ which is estimated for the present composition assuming 1/3 holes per formula unit.) These γ values are anomalously large when compared to $\gamma \sim 10^{-3}$ observed in conventional metals and semiconductors.[7,8] It may be worth noting here that γ values significantly larger than that of conventional metals have been previously observed in the normal state of perovskite copper oxide systems.[10] In Fig. 4 we compare the normalized noise magnitudes in LCMO and PSMO with that of high quality epitaxial films of the copper oxide superconductor $YBa_2Cu_3O_7$. The noise level in the CMR oxides is 5–6 orders of magnitude larger than that of $YBa_2Cu_3O_7$ in the entire temperature range. γ values in these CMR oxides are also nearly 5 orders larger than the (zero field) γ of metallic

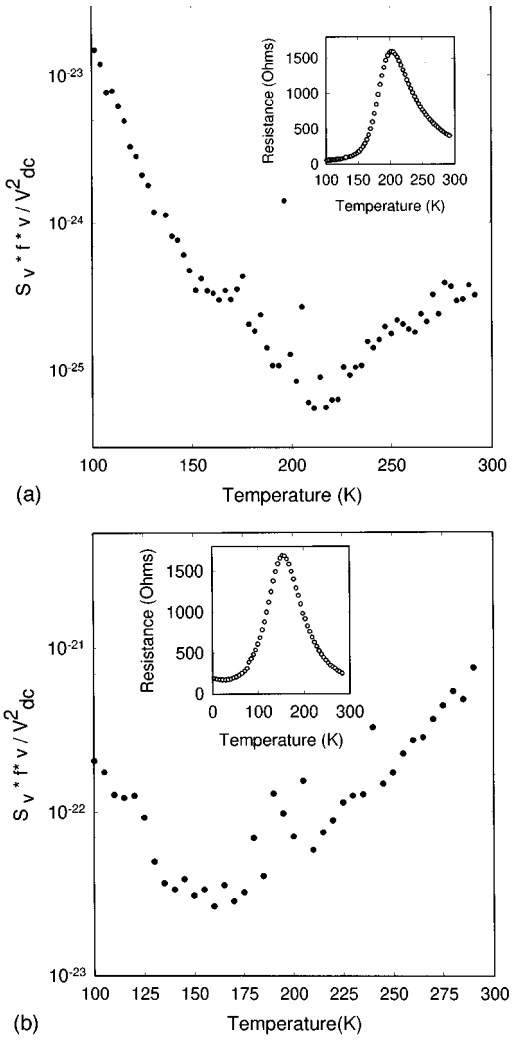

FIG. 2. Temperature dependence of the normalized noise power spectral density and resistance (shown in the inset) for: (a) $La_{0.67}Ca_{0.33}MnO_3$, (b) $Pr_{0.67}Sr_{0.33}MnO_3$.

multilayers exhibiting giant magnetoresistance (GMR).[12]

It is important to rule out extrinsic contributions to the enhanced noise, such as the effects of grain boundaries and other microstructural defects. In the present case, the high

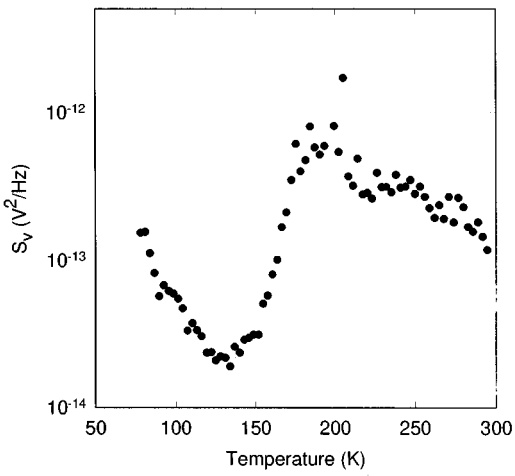

FIG. 3. Temperature dependence of the unnormalized noise power spectral density for $La_{0.67}Ca_{0.33}MnO_3$.

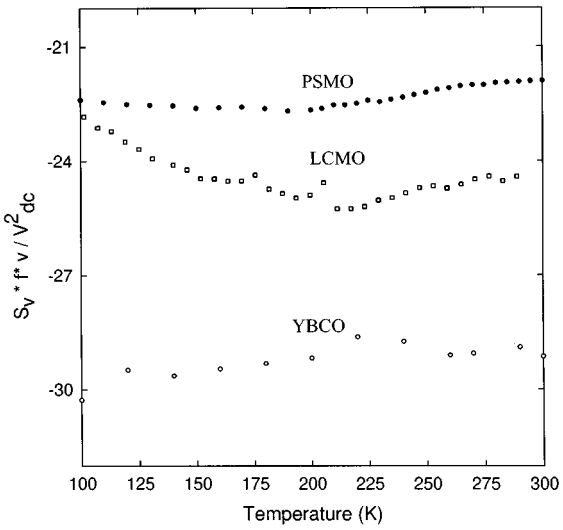

FIG. 4. Comparison of the normalized noise power spectral density of epitaxial thin films of $La_{0.67}Ca_{0.33}MnO_3$ and $Pr_{0.67}Sr_{0.33}MnO_3$ with that of epitaxial thin films of $YBa_2Cu_3O_7$ as a function of temperature.

crystalline quality of the CMR films suggest that the enhanced noise levels may be intrinsic to the manganites. At present we do not have a clear picture of the origin of this large noise. Based on the current understanding of transport, there may be several possible origins: The electronic properties in these materials have been shown to have a strong correlation with the oxygen content.[12] The charge transport has been suggested to be governed by the Zener double exchange mechanism associated with the mixed valence of the Mn ion.[5] In this model, the conductivity is proportional to the hopping rate of the electron between the Mn ions in the Mn^{3+}–O–Mn^{4+} configuration. This hopping rate is very sensitive to the Mn–O bond length and the Mn–O–Mn bond angles which in turn are influenced by oxygen vacancies. Thus, as in $YBa_2Cu_3O_7$,[10] it is likely that the enhanced noise in the CMR oxides is also associated with the oxygen vacancy migration. Another possible source of noise in the ferromagnetic state could be fluctuations in spin alignments since these couple to conductivity.

The temperature dependence of the normalized noise spectral density also presents interesting anomalies. On the high temperature ($T > T_p$) insulating side, the normalized noise spectral density S_v / V^2_{dc} decreases with decreasing temperature, while below the peak ($T < T_p$) in the ferromagnetic state this quantity increases with decreasing temperature. It is important to mention here that the rise at low temperatures is not a consequence of dividing by V^2_{dc} since we see this behavior in the unnormalized data as well (shown in Fig. 4). The behavior on the insulating side conforms to what is expected from the Dutta–Horn model of defect fluctuations[6] where the temperature dependence of the noise power spectral density is related to the energy distribution *D(E)* of the density of the fluctuating states contributing to the noise. However, the noise data on the metallic side is strikingly different and if interpreted in the framework of the Dutta–Horn model, implies a rise in *D(E)* with decreasing temperature, suggestive of the presence of additional modes of fluctuations with lower characteristic energies.

For the purpose of applications, the quantity of relevance is the actual value of the integrated rms noise voltage V_n in the bandwidth of operation. For a given material, V_n depends on the normalized noise power, device geometry, the bias current density, and the bandwidth. To present some typical estimates, we consider a device of size 10 μm$\times$20 μm in lateral dimensions and 200 nm thick, with a bias current density of 10^4A/cm^2. The *1/f* noise integrated over a frequency interval of 1 Hz–1 kHz for this device gives V_n $\sim$50 μV for LCMO and $\sim$100 μV for PSMO. The corresponding resistance noise (i.e., R_n/R) is $\sim$.01% and $\sim$.03%, respectively. For a bolometer with a TCR of 10%, this translates to a noise equivalent temperature difference (NETD) of 1 mK associated with this noise source. For magnetoresistive applications, it is important to keep in mind that the above resistance noise estimates are based on the zero field noise characteristics.

This work was partially supported by the following grants: NAG5-2348 at the University of the District of Columbia and ONR-N000149510547 at the University of Maryland.

[1] K. Chahara, T. Ohno, M. Kasai, and Y. Kozono, Appl. Phys. Lett. **63**, 1990 (1993).
[2] R. Von Helmlot, J. Weckerg, B. Holzapfel, L. Schultz, and K. Samwer, Phys. Rev. Lett. **71**, 2331 (1993).
[3] S. Jin, T. H. Tiefel, M. McCormack, R. A. Fastnacht, R. Ramesh, and L. H. Chen, Science **264**, 413 (1994).
[4] G. C. Xiong, Q. Li, H. L. Ju, S. N. Mao, L. Senapati, X. X. Xi, R. L. Greene, and T. Venkatesan, Appl. Phys. Lett. **66**, 1427 (1995).
[5] R. Mahendiran, R. Mahesh, N. Rangavittal, S. K. Tiwary, A. K. Raychaudhuri, T. V. Ramakrishnan, and C. N. R. Rao, Phys. Rev. B **53**, 3348 (1996).
[6] M. Rajeswari, C. H. Chen, A. Goyal, C. Kwon, M. C. Robson, R. Ramesh, T. Venkatesan, and S. Lakeou, Appl. Phys. Lett. **68**, 3555 (1996).
[7] P. Dutta and M. Horn, Rev. Mod. Phys. **53**, 497 (1981).
[8] M. B. Weissman, Rev. Mod. Phys. **60**, 537 (1988).
[9] M. Rajeswari, S. Lakeou, E. A. Wood, D. D. Choughule, and T. Venkatesan, J. Superconductivity (to be published).
[10] S. Scouten, Y. Xu, B. H. Mockley, and R. A. Buhrman, Phys. Rev. B **50**, 16121 (1994).
[11] H. T. Hardner, S. S. P. Parkin, M. B. Weissman, M. B. Salamon, and E. Kita, J. Appl. Phys. **75**, 6533 (1994).
[12] H. L. Ju, J. Gopalakrishnan, J. L. Peng, Q. Li, G. C. Xiong, T. Venkatesan, and R. L. Greene, Phys. Rev. B **51**, 6143 (1995).