Non-Gaussian resistance noise in the ferromagnetic insulating state of a hole-doped manganite

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We report the observation of a large $1/f$ noise in the ferromagnetic insulating state (FMI) of a hole doped manganite single crystal of $La_{0.80}Ca_{0.20}MnO_3$, which manifests hopping conductivity in the presence of a Coulomb gap. The temperature-dependent noise magnitude decreases in the FMI state indicating a sharp freeze-out of the noise magnitude with temperature on cooling. As the material is cooled down below the ferromagnetic transition T_C, the noise becomes non-Gaussian, as seen through the probability density function and second spectra. On further cooling in the FMI state, the noise becomes more non-Gaussian. The non-Gaussian noise is proposed to arise from charge fluctuations in a correlated glassy phase of the polaronic carriers which develop in these systems, as reported in recent simulation studies.

I. INTRODUCTION

Electronic transport through localized states in disordered and correlated electronic systems has been a topic of considerable interest.[1–3] A fascinating state appears at a low hole doping level, where the ground state of the system has become ferromagnetic but not yet metallic. The unexpected coexistence of ferromagnetism and insulating behavior seems to contradict the conventional double exchange[4] model. The origin of this coexistence is not clear but might stem from a delicate balance of charge localization by orbital ordering (OO),[5] due to the Jahn Teller effect, and ferromagnetic interactions between Mn^{3+} and Mn^{4+} ions.[6] Some theoretical[7] and experimental[8] results suggest that the OO is an important factor for controlling the electron-hole mobility. In such systems, long-range Coulomb interaction can lead to opening up of a soft gap in the density of states (referred to as the Coulomb gap, Δ_{CG}) and hopping conduction in the presence of such a gap.[9] Another consequence is the emergence of "glassy" slow relaxations of charge carriers arising from a large number of low-lying states separated by barriers.[10] Such a glassy phase can lead to enhanced low-frequency (f) non-Gaussian resistance noise (typically with a power spectrum varying as $1/f$)[11,12] arising from charge fluctuations.[13,14] These issues in a Coulomb glass have been reviewed recently.[2] The experimental investigations on these questions are few and were carried out only in the doped semiconductors with electron density close to the critical concentration (n_C) for the metal-insulator (MI) transition[15,16] or in two-dimensional electron glass in MOSFET's.[17] Here, we focus on the issue of non-Gaussian low-frequency noise in the Coulomb glass phase of a very different material, namely, the low hole doped rare-earth manganites which can have a ferromagnetic insulating (FMI) state below a certain temperature (T). In these systems, the Coulomb glass phase occurs for the localized polaronic carriers (in sharp contrast to doped semiconductors) which arise due to strong electron-phonon coupling from Jahn-Teller distortion around the Mn^{3+} ions.

The FMI state of hole doped manganites ($La_{1-x}Ca_xMnO_3$) arises when hole concentration x in $LaMnO_3$ exceeds the critical concentration for ferromagnetism $x \approx 0.125$ yet it is smaller than the concentration needed for the formation of the metallic ground state $x \geqslant x_C = 0.225$. This region of the manganite phase diagram is a topic of current investigations[18] and is marked by the presence of a mixed phase of different orbital orders/disorders that can have different conductivities.[5,8,19] In this range of hole doping in manganites at low temperatures, there is a majority insulating ferromagnetic phase that coexists with a conducting phase. The extent of the phase separation of the two phases, the nature of orbital order, will determine the exact physical property, among other things, which can lead to a spin-cluster glass-type behavior arising from competing spin interactions in the presence of disorder. The existence of the glassy behavior with a long-time tail has been seen in time-dependent resistance relaxation[20,21] and also in NMR relaxation studies at low temperatures deep into the FMI state.[6]

In this paper we address the issue of Coulomb interaction and investigate experimentally whether there is a signature of entry into a Coulomb glass phase which can occur along with the spin-cluster glass phase. Existence of a Coulomb glass behavior in the FMI state has been inferred from certain transport experiments[22,23] and also predicted theoretically recently.[24] We observe large low-frequency resistance fluctuations (noise) with nontrivial temperature dependence in the FMI state. As the material is cooled down below the ferromagnetic transition temperature T_C, the noise becomes non-Gaussian. On further cooling, the noise becomes strongly non-Gaussian. At low temperatures, well below the transition to the insulating state, the noise shows a sharp fall on cooling. We propose below that the large noise as well as its temperature dependence arise from charge fluctuations due to the special nature of carriers in it.

II. EXPERIMENTAL DETAILS

We have investigated the low-frequency resistance noise (50 mHz $< f <$ 10 Hz) in a single crystal of the manganite,

$La_{0.80}Ca_{0.20}MnO_3$ (LCMO20). The noise experiments on single crystals allow us to avoid extraneous influences coming from structural defects[25] which may mask the noise arising from some of the intrinsic effects like the one being investigated here.

The crystals used in this work were grown by the floating-zone technique.[26] We note that the exact composition of the crystal was checked by a number of procedures, including a quantitative chemical method like inductively coupled plasma (ICP): atomic emission spectroscopy. The sample chemical inhomogeneity was tested by a microprobe, and the sample used in the experiment was cut from a portion which shows a homogeneous chemical composition over the length and cross section. In addition, we did titration to check the oxygen stoichiometry.

The resistivity (ρ) versus T was measured by four probes (with evaporated contact pads) using the ac biasing technique with phase-sensitive detection procedure. Typical current (I) bias was $\leqslant 1\ \mu$A. At low T, the measured resistance is a strong function of I when the bias crosses a threshold. As a result, a low-current bias is used.

The noise measurements were carried out with a five-probe[27] ac detection scheme with low ac current, $I = 1\ \mu$A. The same contact pads were used for both the resistivity and noise measurements. We use an ac Wheatstone bridge configuration to measure resistance fluctuations, as depicted schematically in Fig. 1(a). Two parts of the sample (with five probes) show that resistances (r_1 and r_2) with respect to the ground lead (center port) and serve as two lower arms of the bridge circuit. The resistance of the balancing arms (R_1 and R_2) of the bridge are kept much greater than r_1 and r_2 to keep the biasing I constant. The five-probe ac technique has distinct advantages compared to dc biasing techniques.[28] First, the background noise can be measured simultaneously along with the sample noise which allows the background subtraction more reliable one. Also the noise can be measured with much less measuring power, allowing the measurements to stay close to the equilibrium noise. The background noise was measured at each T simultaneously and was found to be "white." The measured noise power for the background noise was found to be very close to the Nyquist value $4k_BTR$. This ensures that the there is no spurious noise in the measurement system. This also removes the possibility of a changing noise contribution due to a current shunting effect, if any. We have also changed the contact size and relative spacing to ensure that such effects like contacts or current shunting do not affect the measurements.

The details of the method and its practical realization have been discussed previously.[28,29] The output from the amplifier [see Fig. 1(a)] was digitized as a time series. A typical example is shown in the inset of Figs. 1(b) and 1(c). The technique uses a sample bias with carrier frequency f_C (we used typically $f_C = 228$ Hz) which is chosen to be the frequency region where detection of electronics noise is low. This minimizes considerably the $1/f$ noise of the detection electronics, which is a problem for dc noise measurements. The voltage fluctuation arises as a side band of f_C after demodulation of the f_C by the lock-in amplifier (LIA). The demodulated signal is fed to the differential input of the analog-to-digital converter (ADC) card. The demodulated signal makes the time series and constitutes the voltage fluctuations ΔV(t). These voltage fluctuations arise from the resistance fluctuation of the current biased sample. The time series was digitized with a 32-bit 200-Ks/s ADC card. A complete set of voltage time series [ΔV(t)] consists of 5×10^6 data points or even more at each T (stabilized to within ± 1 mK). ΔV(t) was processed through a number of digital signal processing (DSP) techniques that include an antialiasing filter, decimation, and digital low-pass filtering.[29] The power spectral density of voltage fluctuations $S_V(f)$ was obtained numerically by using fast Fourier-transformation (FFT) techniques from the stored and digitally processed time series.[29] The apparatus was calibrated down to a spectral power $S_V(f) = 10^{-20}\ V^2$/Hz by measuring the Nyquist noise $4k_BTR$ for a calibrated standard resistor at each T. The noise data for the sample are taken down to $40 \leqslant T \leqslant 300$ K. Below 40 K the resistivity of the material becomes very large for a reliable noise measurement. No heating effects are seen during noise or resistivity measurements.

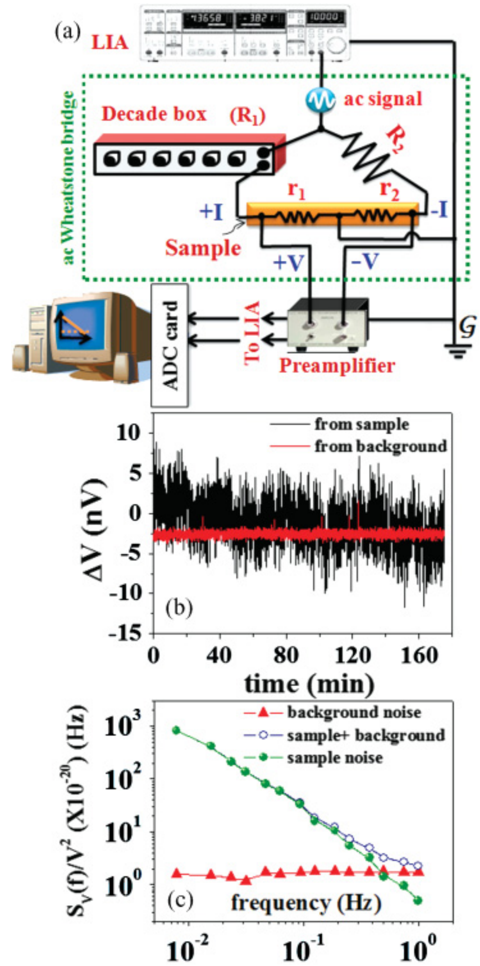

FIG. 1. (Color online) (a) Five-probe noise measurements setup with ac detection scheme. (b) Representative time series data collected simultaneously from the sample (1-Ω carbon resistor) and the background at $T = 300$ K. (c) Noise spectral power $S_V(f)$ along with "white" background noise calculated from the same time series.

The investigation of the non-Gaussian component (NGC) was done by two methods that give complimentary