Nonlinear electrical conduction and broad band noise in the charge-ordered rare earth manganate $Nd_{0.5}Ca_{0.5}MnO_3$

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(Received 21 July 1999; accepted for publication 29 September 1999)

Measurements of the dc transport properties and the low-frequency conductivity noise in films of charge-ordered $Nd_{0.5}Ca_{0.5}MnO_3$ grown on Si substrate reveal the existence of a threshold field in the charge-ordered regime beyond which strong nonlinear conduction sets in along with a large broad band conductivity noise. Threshold-dependent conduction disappears as $T \rightarrow T_{CO}$, the charge-ordering temperature. This observation suggests that the charge-ordered state gets depinned at the onset of the nonlinear conduction. © *1999 American Institute of Physics.* [S0003-6951(99)05247-X]

Rare-earth manganites with a general chemical formula $Re_{1-x}Ae_xMnO_3$ (where Re is a trivalent rare earth and Ae is a divalent alkaline earth cation) show a number of interesting phenomena like colossal magnetoresistance (CMR) and charge ordering (CO).[1] These compounds belong to the ABO_3 type perovskite oxides where Re and Ae ions occupy the *A* site and Mn occupies the *B* site. It has been known for some time that these manganites (depending on the size of the average *A*-site cationic radius $\langle r_A \rangle$) can charge order, for certain values of *x*. The nature of the CO state depends on the value of $\langle r_A \rangle$ and it is stabilized if the value of $\langle r_A \rangle$ is smaller. The CO transition is associated with a lattice distortion as well as orbital and spin ordering.

Recent experiments have established that the CO state is strongly destabilized by a number of different types of perturbations. An applied magnetic field of sufficient magnitude can lead to a collapse of the CO gap, Δ_{CO}, and melting of the CO state.[2,3] The CO phenomenon is stabilized by lattice distortion. A perturbation to the distortion can also destabilize the CO state.[4] Recently, it has been reported that application of an electric field,[5,6] optical radiation,[7] or x-ray radiation[8] melts the CO state in $Pr_{0.7}Ca_{0.3}MnO_3$. It is not clear, however, what causes destabilization of the CO state in these cases and whether the underlying mechanism is the same for all perturbations.

Electric field induced melting of the CO state leads to a strong nonlinear conduction as seen in the bulk[5] as well as in films.[6] This raises a very important question whether there is a threshold field associated with the nonlinear conduction. In a driven system pinned by a periodic potential there exists a threshold force beyond which the system is depinned.[9] If the system is charged and the driving force comes from an electric field then this shows as a threshold field or bias for the onset of a nonlinear conduction. Existence of a threshold field would imply that the melting of the CO state by an applied electric field can actually be a depinning phenomena. We investigated this in films of the CO system $Nd_{0.5}Ca_{0.5}MnO_3$ by careful measurement of field dependent dc transport at various temperatures and also followed it up with a measurement of electrical noise (voltage fluctuation) as a function of applied dc bias. We made the following important observations: (1) There indeed exists a threshold field (E_{th}) below the CO temperature T_{CO} and for $E > E_{th}$ a strong nonlinear conduction sets in; (2) E_{th} strongly depends on T and $E_{th} \rightarrow 0$ as $T \rightarrow T_{CO}$; (3) For $T < T_{CO}$, a large voltage fluctuation ($\langle \delta V^2 \rangle / V^2$) appears at the threshold field. Both E_{th} and $\langle \delta V^2 \rangle / V^2$ reaches a maximum at $T \approx 90$ K ($\approx 0.4 T_{CO}$). (4) The spectral power distribution of the voltage fluctuation is broad band and has nearly $1/f$ character.

In $Nd_{0.5}Ca_{0.5}MnO_3$, a system with relatively small $\langle r_A \rangle$, the CO transition takes place from a high temperature charge-disordered insulating phase to a charge-ordered insulating phase (COI). Charge ordering in this system has been studied by us in details previously.[10] Polycrystalline films of $Nd_{0.5}Ca_{0.5}MnO_3$ (average thickness $\approx$1000 nm) were deposited on Si(100) single crystal substrates by nebulized spray pyrolysis of organometallic compounds. The details of sample preparation and characterization (including x ray) have been given elsewhere.[6] Contacts were made by sputtering gold on the films and connecting the current and voltage leads on the gold contacts by silver paint. The $I-V$ characteristics were measured by dc current biasing and the voltage between the voltage leads was measured by a nanovoltmeter. For measuring the electrical noise, the fluctuating component of the voltage δV was amplified by 5×10^3 times by a low noise preamplifier. The output of the preamplifier was sampled by an ADC card and the data were directly transferred to the computer. The temperature was controlled to within 10 mK for both measurements.

The films have a $T_{CO} \approx 250$ K as seen from the resistivity data. The resistivity was measured at a measuring current of 3 nA, which is much lower than the current where nonlinear conductivity sets in. The experiment was conducted down to 80 K where the sample resistance becomes more than 100 MΩ, the limit of our detection electronics.

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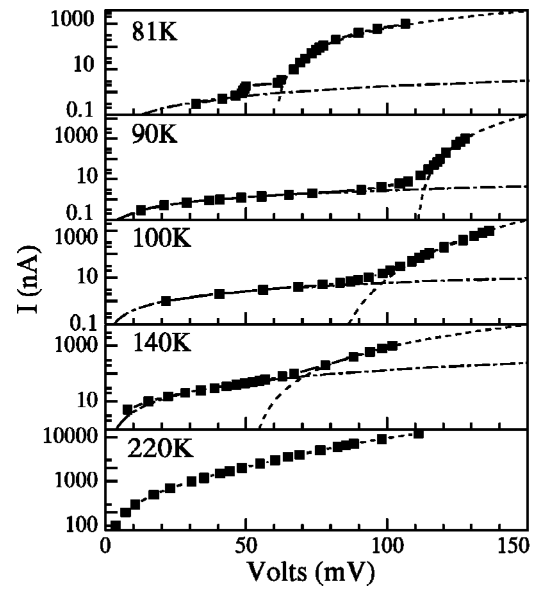

FIG. 1. $I-V$ curves at different temperatures, solid line shows the total I, dashed and dashed–dotted lines show the components I_1 and I_2 as obtained from fit to data using Eq. (1).

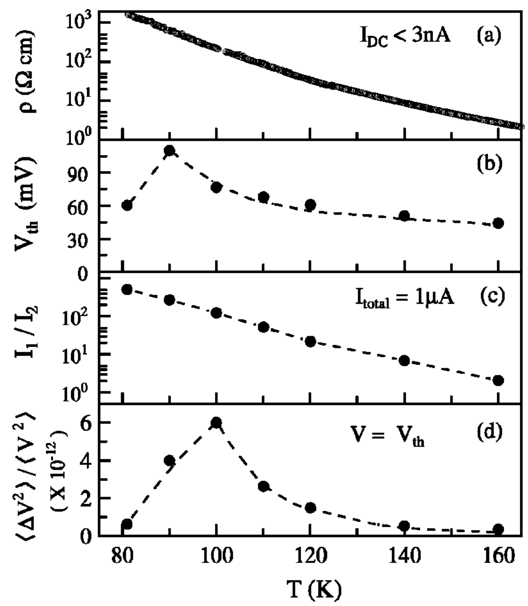

FIG. 2. Temperature variation of (a) resistivity; (b) magnitude of threshold voltage; (c) relative contributions of I_1 and I_2; and (d) noise magnitude at the threshold voltage. The values of I_1 and I_2 are obtained from Eq. (1).

In Fig. 1, we show the typical $I-V$ curves at few characteristic temperatures. At all the temperatures (except at 220 K) there is a clear signature of a threshold voltage V_{th} beyond which the current rises significantly signaling the onset of strong nonlinear conduction. (The separation of electrodes in our experiment is 2 mm, so that $E_{th}=5V_{th}$ V/cm.) $I-V$ curves show two components of conduction: a normal component which exists at all V and a strongly nonlinear component starting at $V>V_{th}$. We fit our $I-V$ data using the following empirical expression which allows us to separate out the two components I_1 and I_2:

$$I=I_1+I_2=g_1(V-V_{th})+g_2(V)=C_1(V-V_{th})^{n_1}+C_2V^{n_2}, \tag{1}$$

where g_1, a function of $(V-V_{th})$, is the component of current (I_1) that has a threshold associated with it and $C_1=0$ for $V<V_{th}$. The component g_2 is the normal conduction component of current (I_2). C_1, C_2, n_1, and n_2 are constants for a given temperature. The data at all temperatures can be well fitted to Eq. (1) for $T>90$ K as shown by the solid lines in Fig. 1. The dashed and dashed–dotted lines give the contributions of each of the terms. For $T<90$ K certain additional features show up (see data at 81 K) in the $I-V$ data which give the impression that there may be multiple thresholds. In Fig. 2(a) we have plotted the threshold voltage V_{th} as a function of T as obtained from Eq. (1). It can be seen that $V_{th}\rightarrow 0$ as $T\rightarrow T_{CO}$. Within the limitations of our detectability, we could see a finite nonzero V_{th} up to $T\approx 170$ K $\approx 0.7T_{CO}$. Beyond this temperature it is difficult to distinguish between the two conduction components. At voltages much below the threshold voltage the $I-V$ can be fitted to a linear equation. However, for $V<V_{th}$ (but comparable to V_{th}) the best fit obtained needs a nonlinear dependence on V. Equation (1) therefore is valid for a range $V>0.1\,V_{th}$. It must be pointed out that the normal component I_2 also has a small nonlinearity since $n_2\approx 1.1-1.4$. This is much less nonlinear than the I_1 component seen above V_{th}.

The relative contributions of I_1 and I_2 to the total current (expressed as the ratio I_1/I_2 evaluated at $I=1\ \mu$A) has been obtained from Eq. (1) and has been plotted as a function of T in Fig. 2(b). At $T\ll T_{CO}$, the nonlinear component is orders of magnitude larger than the normal conduction component and they are comparable as $T\rightarrow T_{CO}$. The exponent n_1 is strongly temperature dependent and from a value ≈ 2 at 160 K it reaches a value more than 5 at $T\approx 100$ K. The exponent n_2 does not have much of a temperature dependence and is $\approx 1.1-1.4$ for $T\leqslant 180$ K.

In a pinned driven system one often sees the onset of broad band noise as the system is depinned at the threshold voltage.[9] We find that such is indeed the case in this system. In Fig. 3 we show the magnitude of the voltage fluctuation $\langle\Delta V^2\rangle/V^2$ as a function of the applied bias V at $T=100$ K along with the $I-V$ curve. The arrow indicates V_{th}. It is clear that the voltage fluctuation has a nonmonotonous de-

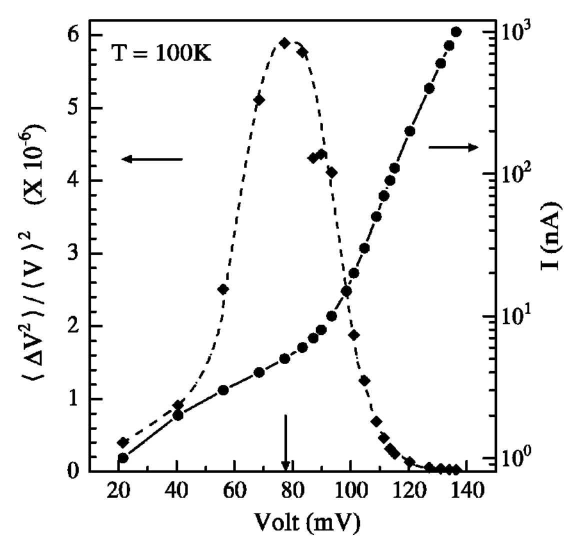

FIG. 3. The noise magnitude and $I-V$ characteristics at 100 K. The arrow indicates the threshold voltage.

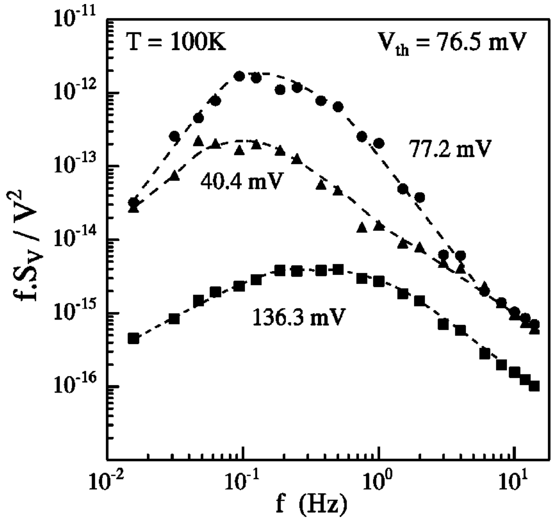

FIG. 4. Frequency spectrum of the noise at 100 K for different bias values.

pendence on V and reaches a peak at $V \approx V_{th}$. This fluctuation has been seen at all $T < 0.7\,T_{CO}$ where we can detect measurable V_{th}. The peak values of the fluctuation measured at different T are shown in Fig. 2(c). The fluctuation$\rightarrow 0$ as $T \rightarrow T_{CO}$ and has a peak at 90 K where T_{th} also shows a peak.

Frequency dependencies of the spectral power $S_V(f)$ measured at 100 K with biases $V < V_{th}$, $V \approx V_{th}$, and $V > V_{th}$ are shown in Fig. 4. We have plotted the data as $f.S_V/V^2$ vs f. For a pure $1/f$ noise ($S_V \propto 1/f$), this should be a straight line parallel to the f axis. It can be seen that the predominant contribution to noise has $1/f$ character and the noise becomes more $1/f$ at higher voltages.

The onset of strong nonlinear conduction at a threshold voltage and the accompanied broad band noise has been seen in solids like $NbSe_3$, TaS_3 which show depinning of charge density waves (CDW) by a threshold field.[9] Though the physics of CDW and CO states are entirely different, the underlying phenomenological description of depinning can be similar. Electron diffraction (ED) and electron microscopy studies on a CO system ($La_{0.5}Ca_{0.5}MnO_3$) have shown that the CO is associated with formation of stable pairs of $Mn^{3+}O_6$ stripes. The $Mn^{3+}O_6$ octahedra in the stripes are strongly distorted by the Jahn–Teller (JT) distortion.[11] It is possible that the stability of the CO system depends on the stability of the stripes which can be pinned. The strong JT distorted pairs of the $Mn^{3+}O_6$ octahedra can act as periodic pinning sites due to local strain field. From our data for $T < 90$ K, it seems there are changes occurring below 90 K. We are not clear about the changes. We only note that in magnetic studies we found that strong irreversibility sets in below 80 K.[10]

To conclude, the present study demonstrates that there is a threshold field associated with the onset of nonlinear conduction in the CO system along with the existence of a broad band noise. The observation is taken as evidence of depinning of the CO state as the origin of nonlinear conduction in these solids.

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