Charge density waves enhance the electronic noise of manganites

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The transport and noise properties of $Pr_{0.7}Ca_{0.3}MnO_3$ epitaxial thin films in the temperature range from room temperature to 160 K are reported. It is shown that both the broadband $1/f$ noise properties and the dependence of resistance on electric field are consistent with the idea of a collective electrical transport, as in the classical model of sliding charge density waves. On the other hand, the observations cannot be reconciled with standard models of charge ordering and charge melting. Methodologically, it is proposed to consider noise-spectra analysis as a unique tool for the identification of the transport mechanism in such highly correlated systems. On the basis of the results, the electrical transport is envisaged as one of the most effective ways to understand the nature of the insulating charge-modulated ground states in manganites.

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I. INTRODUCTION

The narrow-band, mixed-valence manganites are since over ten years under the focus of a wide scientific community, mainly because of the "colossal" responses of their electronic properties to magnetic field and to other external perturbations. It was soon recognized that many effects are related to charge-modulated phases, described in the earlier models as due to the full charge disproportionation on the Mn site, and to the consequent crystallization (i.e., "charge ordering," CO) of a lattice of ordered Mn^{3+}/Mn^{4+} ions. Such ordered systems can be abruptly driven to the conducting state by several kinds of perturbation, including electrical[1] and magnetic field,[2] optical,[3,4] x ray radiation,[5] and pressure.[6] Within the classical interpretation, such effects are ascribed to the "melting" of the CO phase[1,2,4] meaning that the perturbation destroys the ordered state and frees the charge carriers, enhancing the conductivity up to several orders of magnitude. Recently, this interpretation has been challenged by several authors, on the basis of an increasing body of evidence pointing to more complex descriptions of both the static and the dynamic properties of such compounds. As we discuss in the following, the results of our research on the transport properties of the $Pr_{0.7}Ca_{0.3}MnO_3$ manganite also support the same view.

$Pr_{1-x}Ca_xMnO_3$ (PCMO) is a very interesting system, where charge, orbital, lattice, and spin degrees of freedom strongly interact and determine the macroscopic properties. The phase diagram of bulk PCMO (Ref. 7) includes an insulating ferromagnetic phase at $x<0.3$, where the CO state is suppressed. In the higher doping range ($0.3<x<0.75$), an ordered phase is found below the transition temperature T_{CO}. T_{CO} exceeds 200 K in the range $0.3<x<0.5$, then it reaches its maximum at the "commensurate" stoichiometry $x=0.5$, and it finally drops at higher doping.[8,9] The structure of PCMO at $0.3<x<0.5$ has been recently subject of extensive investigation,[10,11] in order to settle the issue of charge localization. Both quoted papers indicate that the lattice hosts Zener polarons, i.e., dimers of Mn ions resonantly sharing an electron. The ordered state is then depicted as a Zener polaron lattice, that is well ordered at $x=0.5$ and that is stable down to $x=0.3$, where the extra Mn^{3+} ions are hosted as impurities. The conduction mechanism in PCMO above the ordering transition is well described by models based on the motion of thermally activated Jahn-Teller polarons.[12] The same authors show that, below the ordering transition temperature, the conduction is instead due to a cooperative mechanism involving the coherent motion of clusters of Zener polarons. Above a certain threshold current, a transition toward a light-polarons state sets in determining the giant electroresistive effect.

The existence of a cooperative mechanism of motion is also found in the classical model of charge density waves (CDW), introduced in his pioneering work by Peierls[13] and later by Grüner.[14] Within this model, the one-dimensional electron density is (incommensurately) modulated, due to the interaction with phonons. The CDW picture has been invoked in recent papers on manganites. Cox *et al.*[15,16] base this claim on an accurate characterization performed by resorting to transmission electron microscopy and to transport measurement on $La_{0.5}Ca_{0.5}MnO_3$ and $Pr_{0.7}Ca_{0.3}MnO_3$ films. Nucara *et al.*[17] provided support to the same ideas on the base of optical conductivity measurements on several compositions of manganites, showing spectral features that are associated with the typical excitations (phasons and amplitudons) of a CDW system. Previous data supporting this concept had also been reported, based on nonlinear transport,[18] optical spectroscopy,[19,20] and specific-heat measurements.[21]

The debate is still quite hot since it is also argued[22] that the CDW regions of the phase diagrams, if any, are limited to restricted areas at the border with a metallic phase. Different intermediate situations may probably take place in different materials, according to bandwidth, doping, quenched disorder, and strain. Under this point of view, PCMO with a doping level $x<0.5$ is an interesting subject of investigation. Furthermore, a clear-cut distinction between the CDW slippage (that is, a coherent motion) and the disordered creep of a rigid CO lattice is not straightforward. The experimental identification of a CDW state by only dc transport measure-

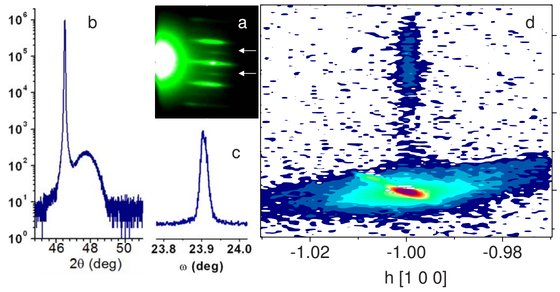

FIG. 1. (Color online) (a) RHEED patterns of a PCMO film; the arrows indicate the (110) and reflections. (b) $\theta/2\theta$ showing the (002) STO and the (004) PCMO peaks (counts are in logarithmic scale); (c) the (004) PCMO rocking curve; (d) reciprocal space map showing the (310) STO and the (622) PCMO.

ments is quite cumbersome and, probably, it cannot be unambiguous. In this paper, concerning the electric properties of high-quality $Pr_{0.7}Ca_{0.3}MnO_3$ films, we propose instead that the transport regime is better identified by investigation of the electronic noise as a function of the temperature and of the applied electric field.

II. EXPERIMENT

The samples adopted in this work are thin (10 nm) $Pr_{0.7}Ca_{0.3}MnO_3$ (PCMO) epitaxial films grown under tensile strain on (100) $SrTiO_3$ (STO) substrates. Details of the complex pulsed laser deposition chamber where the growth took place are reported in Ref. 23. Briefly, the films are grown in a 0.1 mbar oxygen atmosphere, at a rate of 2 unit cells per minute, by resorting to a Kr-F excimer laser (248 nm) that radiates the target with 50 $\text{mJ}/2.4\ \text{mm}^2$ effective fluency. The reflection high-energy electron diffraction (RHEED) patterns collected during deposition [Fig. 1(a)] show flat surfaces (two-dimensional growth) preserved until the end of the deposition. The bulk $Pr_{0.7}Ca_{0.3}MnO_3$ is orthorhombic, with lattice parameters $a=0.5426$ nm, $b=0.5478$ nm, and $c=0.7679$ nm. The substrate lattice spacing, $a_s=0.3905$ nm, is matched to $Pr_{0.7}Ca_{0.3}MnO_3$, being $c\approx 2a_s$ and $a\approx b\approx\sqrt{2}a_s$. Such a pseudocubic orthorhombic film can be accommodated on STO in several ways.[24,25] Our x-ray diffraction characterization is indicative of a c-axis-orientated single domain with vertical spacing $c=0.7622$ nm, Fig. 1(b). The in-plane $Pr_{0.7}Ca_{0.3}MnO_3$ lattice is fully strained [Fig. 1(d)], with the a and b axes aligned to the (110) STO so that the unit cell is tetragonal. Such epitaxial accommodation results in a relatively small strain ($\epsilon_a=0.018;\epsilon_b=0.008;\epsilon_c=-0.007$) and a high crystal quality [Fig. 1(c)].

The stress applied by the substrate, in principle, may set a difference between film and bulk properties. The bulk samples, with $x=0.3$, are just on the borderline, in the bulk phase diagram, between the insulating ferromagnetic and the insulating antiferromagnetic, charge-ordered phase. The films have an intermediate behavior, that will be thoroughly discussed elsewhere and that we briefly anticipate here. The samples are ferromagnetic below 120 K, even though the large difference between zero-field-cooled and field-cooled magnetization vs temperature, the high coercivity and other experimental signatures suggest a glassy ferromagnetic state. On the other hand, as we show and discuss in the following, the samples also undergo a transition that we interpret as due to charge modulation at about 230 K. In our understanding, this charge-modulated state is stable in the range 120–230 K while it is melted at about 230 K; below 120 K there is indication of coexistence of charge and ferromagnetic ordering.

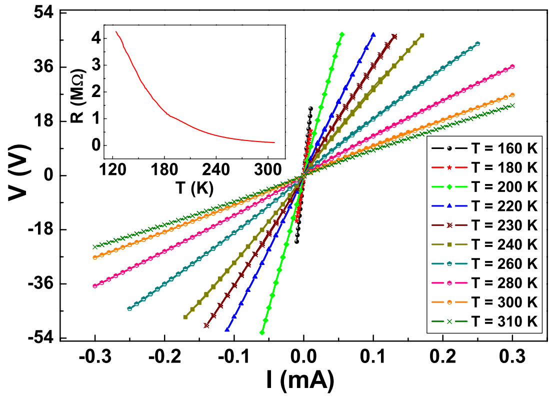

FIG. 2. (Color online) I-V curves in range $160<T<310$ K. Inset: R vs T for the same sample.

The transport measurements were carried out in the standard four-probe configuration with inline geometry. The electrical contacts were obtained by bonding gold wires on silver stripes, deposited at 2 mm one from each other so that the lines of current were always at 45° with the a axis. The superstructure in the charge-ordered phase, on the other hand, is known to modulate the bulk properties along the a axis. We expect this also holds for our films, due to the low strain of the structure. Only a few reports regard structural investigations of charge ordering in films; a modulation along the a axis is reported in the similar compound $La_{0.5}Ca_{0.5}MnO_3$ films.[15] The resistance vs temperature $R(T)$ (Fig. 2) was measured in the current-pulsed mode, by biasing the samples with an active current source. No evident feature is present at the expected charge-modulation temperature as reported in bulk ($T_{CM}\approx 230$ K) while a small feature is found at 180 K, corresponding to the Neel temperature T_N.[8] A signature of the charge modulation taking place below 230 K is provided by the temperature-dependent I-V characteristics reported in the same picture. The data were collected by resorting to 1 ms current pulses to avoid Joule heating. Below 230 K, a slight nonlinearity sets in. No hysteretic features were observed, that we attribute to the limited value of the applied electric field.

Voltage-noise spectral-density measurements were carried out both in the four-probe and in the two-probe configurations. A major effort was dedicated to minimizing the electrical noise generated by the contacts. The problem was circumvented by resorting to an improved measurement technique, described in Ref. 26, aimed to eliminate artifacts due to the contact noise contribution. The normalized voltage spectral density $S_N(f)=S_V(f)/V^2$ reveals a unique $1/f$ component between 160 and 310 K [Fig. 3(a)]. The most striking feature [Fig. 3(b)] is a steplike increase in the S_N amplitude

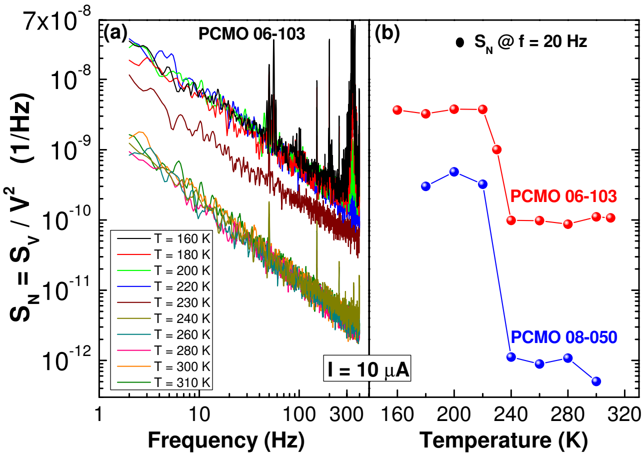

FIG. 3. (Color online) (a) Normalized voltage spectral density S_N at fixed current $I=10\ \mu$A between 160 and 310 K. The peaks superimposed to the $1/f$ component are due to external noise sources. (b) S_N at $f=20$ Hz for two distinct samples.

by 1–2 orders of magnitude, at about 230 K.[27] Several performed checks exclude that such unusual behavior is an artifact. Figure 3(b) shows, for example, the comparison of data regarding a fresh sample (PCMO 08–050) and a two years old one (PCMO 06–103), the latter showing a much higher noise intensity, probably due to development of structural defects or to the interaction with air. In spite of the overall different quantitative behavior, the two samples share the essential feature which is under investigation in this work.

In order to get a further insight into the physical properties of $Pr_{0.7}Ca_{0.3}MnO_3$ below 230 K, we compared the electric field dependence of the differential conductivity and of the normalized spectral density (Fig. 4). Above 230 K, the normalized differential conductivity [defined as $(dI/dV)/\sigma_0$, where σ_0 is the conductivity at zero voltage] is constant vs the electric field E, that is, the sample exhibits an Ohmic behavior. In this regime, the normalized voltage spectral density is independent of E, as well. In the low-temperature phase, on the contrary, both plots are characterized by a threshold $E_c \simeq 1000$ V/m at which a further very strong steplike increase in the S_N amplitude is found.

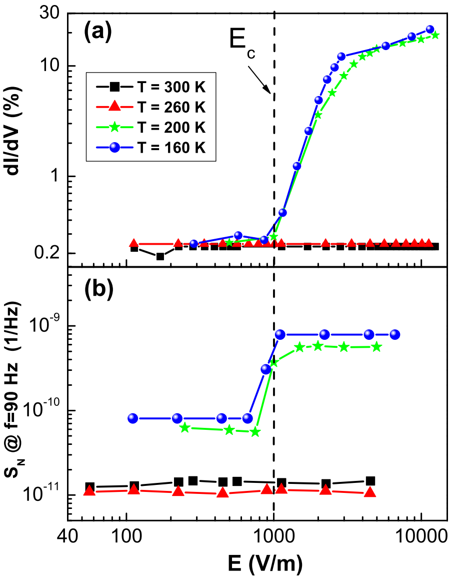

FIG. 4. (Color online) Electric field dependence (a) of the normalized differential conductivity and (b) of the normalized spectral density at a frequency $f=90$ Hz.

III. DISCUSSION

The absence of any sign on the $R(T)$ curve and the steplike noise increase are not easy to reconcile with a "strong coupling" CO picture in which the ordered network of charges is tightly localized on Mn sites. In such a case we would rather expect (i) a step increase in resistivity and (ii) a noise reduction when the "charge crystal" condenses from the "liquid" phase. Phenomenologically, the CDW model can instead explain these findings: (i) the continuity of R at T_{CM} can be understood in the framework of a phenomenological two-fluid model (we remind, to this aim, the noticeable, formal similarities between the CDW state and the superconducting state). In such model, the relative weight of the ordered phase (that in the classical view is not spatially separated) starts from 0 at the thermodynamic transition temperature and it approaches 1 only at $T=0$. In the microscopic description of the CDW state, the same gradual effect on resistivity can be envisaged as due to the temperature dependence of the thermal excitations above the gap. The low-temperature value $2\Delta(T=0)$ in the excitation spectrum of the CDW state has been estimated $\approx 100-200$ meV in Ref. 16. We note that the quoted mechanisms are related to dimensionality issues, namely, they may have different impact in bulk and in thin films. Actually, PCMO bulk samples generally show sharper signatures of the ordering transition in $R(T)$ plots with respect to thin films. (ii) Intuitively, the transition to an ordered state should reduce the electronic noise. Even if the conduction mechanism is due to creep of charges at the border of the charge-crystal region, one may imagine that the noise is at most unchanged since the creep is not qualitatively different from the polaron hopping characterizing the melt state. On the contrary, the presence of an excess broadband $1/f$ noise is a well-established feature of CDW systems.[28,29] Such $1/f$ noise stems from the many metastable states of the CD condensate, that may be seen as long-wavelength phase modulations of the CDW order parameter. Each such state suffers from different pinning forces due to the lattice defects and it is therefore characterized by a different electrical resistance. At a given value of current flow, the thermal activation of transitions between such states determines a resistance fluctuation that results in the extra voltage noise.[30]

Once the CDW model is accepted, more details that connect the noise spectra to the dc transport properties are better understood. The onset of nonlinearity above a critical field (in the I-V characteristics below T_{CM}) is also a typical signature of CDW systems. In the Lee-Rice model[31] E_c is the depinning field, above which the CDW can slide. Our value of 1000 V/m compares well with other reported data.[15,32] More importantly, we observed that the enhancement of

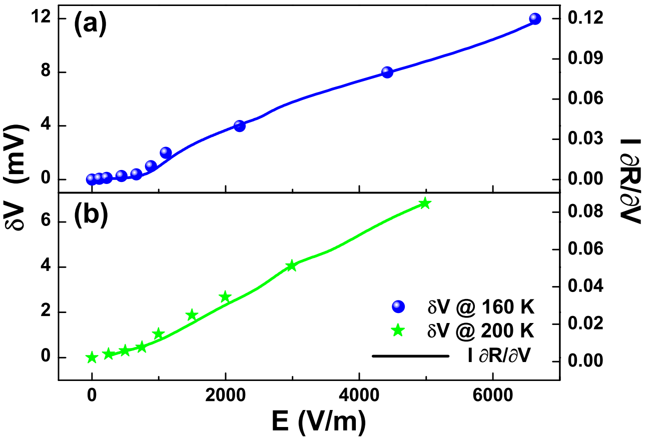

FIG. 5. (Color online) δV (points) and $I\partial R/\partial V$ (solid lines) vs E, at (a) 160 K and (b) 200 K. Right scales have been adjusted for best fitting with noise data.

noise starts just below the threshold, and it is completed at E_c, that is, the same scale of fields determines both the nonlinearity and the noise enhancement. This is just what one expects in a CDW state.[29]

A further check is based on the comparison between our data and the theoretical expectations for the behavior of $1/f$ noise. According to Ref. 29, assuming that the pinning force fluctuations are the source of noise in the chordal resistance $R=V/I$, the voltage noise is given by $\langle \delta V^2 \rangle = I^2(\partial R/\partial V)^2 \langle \delta V_c^2 \rangle$, where V_c is the threshold voltage associated with E_c. This expression was employed in Ref. 29 to establish a quantitative relation between the nonlinear conductance and the noise level. By applying the same analysis to our data below $T_{CM} \approx 230$ K, a quantitative agreement is found. In Figs. 5(a) and 5(b), the average voltage noise $\delta V = \sqrt{\langle \delta V^2 \rangle}$, where $\langle \delta V^2 \rangle = \int_{1\ \mathrm{Hz}}^{100\ \mathrm{kHz}} df S_V(f)$, is plotted vs the electric field E; the behavior is compared to the values of $I\partial R/\partial V$ vs E, as determined by the corresponding I-V characteristic of the sample. The data fairly overlap. In the Ohmic regime above T_{CM}, on the other hand, no such correlation is possible because $I\partial R/\partial V \simeq 0$ while δV grows linearly with the electric field [see, e.g., Fig. 4(b), where $S_N = \mathrm{const}$ at 260 and 300 K implying $\delta V \propto E$). Furthermore, we can evaluate the coherence volume λ^3, that is, the typical volume over which the CDW phase is deformed in going from one metastable state to another, by the equation

$$S_V(f,T) = I^2 \left\{ S_R(f,T) + \left[\frac{\partial R}{\partial V} \right]^2 E_c^2 \lambda^3(T) \frac{l}{A} S(f,T) \right\} \qquad (1)$$

that we adapted from Ref. 29 to include the fluctuations in the normal state. Here l and A are sample length and area, $S_R(f,T)$ is the spectral density of resistance fluctuations and $S(f,T)$ is the normalized spectral weight function of the transition rate between the metastable states of the condensate. By integrating Eq. (1) over the investigated range of frequency (1 Hz–100 kHz) and by neglecting the fluctuation term related with S_R, which is more than one order-of-magnitude lower than the measured noise, λ is determined in terms of the effective voltage noise $\langle \delta V \rangle$. By using our estimation $E_c = 1000$ V/m, we get $\lambda \simeq 400$ nm at $T=160$ K. This is close to the value of the Lee-Rice coherence length $\lambda_{LR} \simeq 300$ nm, based on the relation of Ref. 31, for $E_F \simeq 1$ eV and $Q \simeq 10^{10}$ m^{-1} (see, e.g., Ref. 15 for La$_{0.5}$Ca$_{0.5}$MnO$_3$). Moreover, we find $\lambda \simeq 100$ nm at $T = 200$ K. The decrease in λ, when the temperature approaches T_{CM}, is consistent with the Lee-Rice result $\lambda_{LR} \to 0$ as $T \to T_{CM}$, when assuming that λ scales with λ_{LR}.

Finally, we would like to mention that other physical mechanisms, that may be invoked as source of the extra noise in the charge-modulated phase, do not lead to the observed broadband $1/f$ behavior. As an instance, the tunneling of charges through the grain boundaries between separate regions (may take place in the case of phase separation) results in either a Lorentzian spectral density or a white spectrum.[33] The random telegraph noise, that also may play a role in phase-separated compounds, is related to the occurrence of strongly nonlinear electrical transport and to a Lorentzian spectral density in the very low-frequency region[34,35] at odds with our findings of weak nonlinearity and pure $1/f$ behavior.

IV. CONCLUSIONS

In conclusion, we investigated the nature of the charge-modulated state in Pr$_{0.7}$Ca$_{0.3}$MnO$_3$ thin films by measuring the properties of its current-carrying excited states. The principal conclusion of this work is that the voltage-noise measurements proved to be a unique probe of the transport mechanism that is activated below the ordering transition temperature. The observed steplike increase in the broadband $1/f$ noise level at lowering the temperature is peculiar and calls for a specific interpretation. In most conventional materials, the $1/f$ component is, in fact, either temperature independent or slowly decreasing at decreasing temperature.

Our results are in fact consistent with the expectations based on the CDW model, formerly adopted for one-dimensional metals and recently extended to the description of narrow-band manganites. Qualitatively, it is hard to reconcile the observation of the $1/f$ extra noise with the picture of a rigid lattice of charges crystallizing from a melt phase of polarons, where a noise reduction may well be expected. Moreover, it is still more difficult to understand why the subsequent application of electric fields above some threshold (that in the CO scenario should contribute to melt the lattice again, coming back to the pristine conditions) should further increase the noise.

We also could verify that the extra-noise mechanism is connected with the onset of nonlinearity in the I-V characteristic of samples. Actually, the detailed description of the voltage-noise features fully agree with current models of voltage-noise generation in the CDW state.

This is, in our opinion, a significant result, in view of the still open debate on the CDW state in manganites. The narrow-band oxides are such a complex system, highly sensitive to doping, stress, etc., that more data on the issue of the CDW state and of its implications are actually welcome, to assess under what circumstances and in what samples the model is applicable. We stress, however, that the model itself may need some revision in its theoretical foundations, to account for the specific features of perovskitic, narrow-band oxides, as compared to the simple, one-dimensional metals that are considered in the original work of Peierls.

The implications of the CDW model are of great interest in connection with the fundamental properties of the charge-modulated state such as its commensurability with the lattice. In this respect, we observe that the investigated PCMO samples, with $x=0.3$ doping level, are far from the ideally commensurate $x=0.5$ value. The consistent applicability of the CDW model to this case, in connection with other published results on different doping levels (and materials), is an indication of the robustness of such state, meaning that it seems to extend its range of application on a wide portion of the phase diagram.

The emerging picture suggests that in manganites the charges may be displaced with more easiness then formerly believed, thus envisaging a connection with the CDW dynamics, that requires a deformable charge lattice. Such a finding is relevant to many aspects of the physics of manganites and, in general, of correlated oxides. While in a CO state a phase-separation picture between localized CO regions and ferromagnetic delocalized regions is expected, novel "electronically soft" phases,[36] where the two order parameters (charge modulation and magnetization) coexist within the same domain, have been proposed to be compatible with the CDW case. This is pertinent to the physics of our samples, where both transitions are found. Together with recent claims of sliding charge density waves in cuprates,[37] these results suggest that the CDW mechanism should be introduced as one of the bricks necessary to build up our comprehension of strongly correlated transition-metal oxides. Such interpretation also allows to bridge the established approach that envisages CDW compounds as chaotic systems to the recent claims that the understanding of correlated oxide properties should be pursued in the broader context of complexity.[38]

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