Weak localization and $1/f$ noise in $Nd_{1.83}Ce_{0.17}CuO_{4+\delta}$ thin films

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We report on electrical transport measurements and voltage-noise analysis in unreduced $Nd_{1.83}Ce_{0.17}CuO_{4+\delta}$ thin films. At low temperatures ($T \leq 100$ K) the resistivity behavior is characterized by a metal-insulator crossover, which in these materials is usually interpreted in terms of weak localization induced by excess oxygen ions randomly distributed on apical impurity sites. The low-frequency voltage-spectral density reveals the presence of different conduction mechanisms in the metallic and in the insulating regions. Standard resistance fluctuations explain well the $1/f$ noise at temperatures above the resistance minimum while an unusual linear dependence of the $1/f$ noise on the applied bias current is found at lower temperatures, which could be interpreted as a signature of the occurrence of weak localization.

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I. INTRODUCTION

The normal-state transport properties of the high-T_c superconducting cuprates show unusual features which are still a matter of intense debate as far as their interpretation is concerned. In particular, they have been deeply investigated below T_c by destroying the superconducting phase with the application of a suitably strong magnetic field, in hole-doped[1] as well as in electron-doped cuprates.[2–4] In many cases it has been found that the resistivity in the CuO_2 planes shows no monotonic behavior as the temperature is decreased across T_c but it rather exhibits a characteristic upturn following a logarithmic dependence.

This low-T behavior of the normal-state resistivity seems to be rather general since it is also typical of cuprates which do not superconduct at any temperature because of an insufficient doping level. This is particular evident in the electron-doped cuprates $Nd_{2-x}Ce_xCuO_4$ (NCCO) and $Pr_{2-x}Ce_xCuO_4$ (PCCO) where the upturn is seen not only in the nonsuperconducting underdoped case[5–7] but also in samples which, despite a cerium concentration sufficient, in principle, to allow superconductivity, remain normal at any T because of the presence of excess oxygen preventing the formation of the superconducting phase.[8–10]

A resistivity showing a minimum and then rising logarithmically as $T \to 0$, together with the experimental evidence that charge dynamics is essentially confined to the CuO_2 planes, has led some authors[3,5,6,11,12] to conclude that the crossover from a metallic ($d\rho/dT > 0$) to an insulating ($d\rho/dT < 0$) regime may be due to a two-dimensional (2D) weak localization[13] producing a logarithmic quantum correction to the Drude conductivity associated with coherent carrier backscattering.[14] This conclusion has been drawn also on the basis of a strongly anisotropic magnetoresistance $\Delta\rho = [\rho(H) - \rho(0)]/\rho(0)$ which at low temperatures is negative for fields applied perpendicularly to the *ab* plane, and practically vanishing for parallel fields.[5,6,11]

Though this appears to be the more plausible explanation, other conjectures on the origin of the upturn have also been made. In the study performed on nonsuperconducting NCCO films by Sekitani *et al.*,[7] the authors ascribe the observed resistivity upturn to the scattering of the carriers by Cu^{2+} Kondo impurities induced by the presence of residual apical oxygen ions. A spin-scattering mechanism, possibly related to the antiferromagnetic fluctuations persisting in a large region of the parameter space, has also been invoked to explain a similar resistivity behavior observed in PCCO.[15] Finally, a further attempt[16] to explain the metal-insulator crossover in the magnetic field driven normal state of NCCO single crystals relies on the theory of the electron-electron interaction in metallic granular systems.[17]

Regardless of the mechanism leading to the logarithmic increase in the resistivity at low T, it should be noted that the large amount of experimental transport investigations performed on high-T_c cuprates to our knowledge did not include noise measurements in the crossover regime. In this context, natural candidates for the realization of this kind of experiment may be nonsuperconducting electron-doped compounds not optimized in the oxygen content through a suitable annealing process. Indeed, they give the possibility to analyze in detail the key role played by the disorder associated with excess nonstoichiometric oxygen, which, as mentioned above, in this kind of systems is likely to give rise to 2D weak-localization effects.

We also point out that besides high-T_c cuprates, other classes of compounds exhibit a metal-insulator crossover at low temperatures, possibly originated by similar microscopic mechanisms. Among them, those showing evidence of Anderson localization are characterized by unusual voltage-noise processes which apparently are difficult to reconcile under a unified point of view. Prasad *et al.*[18] investigated Anderson localization effects in $La_{1-x}Sr_xVO_3$ samples, interpreting the results of their analysis on conductivity and $1/f$ noise in terms of the pseudogap theory of localization. Cohen and Ovadyahu[19] observed high noise levels in polycrystalline indium-oxide films and ZnO accumulation layers. They interpreted these results as effects of incipient localization near a metal-insulator transition, ruling out classical percolation phenomena. Ghosh and Raychaudhuri[20] found two different sources for the noise processes above and below the Anderson transition in silicon single crystal doped with phosphorus and boron. These results seem to indicate that the dc transport and the noise behavior in the crossover regime may not be universal. The nature of these effects has not been clarified yet, and, as a consequence, more studies on materials which are likely to exhibit Anderson localization phenomena are necessary.

Triggered by this interesting scenario, we performed dc transport measurements and voltage-noise analysis on nonsuperconducting $Nd_{1.83}Ce_{0.17}CuO_{4+\delta}$ thin films deposited by sputtering dc technique. The experimental procedure, the preparation process, and the structural properties of the investigated samples are described in Sec. II while Sec. III contains all the experimental results. A qualitative interpretation of the unusual voltage-noise behavior observed near the metal-insulator crossover is proposed in Sec. IV. A short summary of the paper content is then given in Sec. V.

II. SAMPLE PREPARATION AND EXPERIMENTAL PROCEDURES

The NCCO thin films were grown on (001)-oriented $SrTiO_3$ substrates by dc sputtering technique in on-axis configuration. A single stoichiometric target, prepared by standard solid-state reaction, was used as sputtering source.[21] The samples were deposited at 1120 K in a mixed atmosphere of Ar and O_2 at a total pressure of 170 Pa. With this procedure 250-nm-thick films were fabricated at a growth rate of 1.2 nm/min. After deposition, an annealing process in vacuum for about 30 min at the same temperature was performed but no sign of a superconducting transition was observed down to 4.2 K. Rather, these as-grown samples showed a clear metal-insulator crossover with a minimum in the resistivity at a given temperature $T_{\min}$. As widely reported in the literature, this behavior is usually assumed to be due to the disorder induced on the atomic length scale by a small percentage of excess oxygen ions, randomly occupying apical sites which are nominally vacant in the ideal layered T' structure in which NCCO and PCCO tend to crystallize.[22] These excess ions inhibit the formation of the superconducting phase, even in the case of optimal cerium doping, and thus need to be removed by means of an additional thermal treatment in order to make superconductivity appear.[23]

Film quality was monitored by x-ray diffraction measurements, scanning electron microscopy, and wavelength dispersive spectroscopy (WDS).[24] θ-2θ x-ray spectra showed strong (00l) peaks indicative of a c-axis orientation normal to the substrate surface. The full width at half maximum (FWHM) of the (004) Bragg-peak results, on average, the same ($\sim 0.05°$) even changing the oxygen content in the as-grown samples, this implying that the grain size is not affected by the oxygen reduction process. Moreover, the FWHM values of the rocking curve around the same (004) diffraction peak are $\sim 0.38°$ for films 250 nm thick, in agreement with the values reported for other cuprates of similar thickness.[25] The WDS analysis revealed a slight excess of Ce content (x=0.17) with respect to the optimally doped value (x=0.15). Finally, we point out that the value of $T_{\min}$ decreases with decreasing the oxygen partial pressure in the chamber, indicating that the transport properties are strongly affected by the disorder induced by the excess oxygen. Further details about the growth technique and structural characterization of the films will be described elsewhere. We stress that as-grown nonsuperconducting samples were the only ones used for the experimental investigation described here. After noise measurements, an additional *ex situ* thermal treatment in ambient atmosphere of Ar at 1170 K made the films superconducting with a critical temperature T_c around 10 K.[26] This is a further confirmation that our samples are slightly overdoped ones as far as Ce concentration is concerned.

The analyzed thin-film samples were patterned into 30-μm-wide and 1-mm-long strips, using standard photolithography and wet etching in 20% HCl solution. Standard four-probe technique with in line geometry was used to investigate the dc electrical transport and ac voltage-noise properties. All the measurements were carried in a closed-cycle refrigerator. The temperature was stabilized in the 9–300 K range with a stability better than 0.1 K using a GaAlAs thermometer and a resistance heater controlled in a closed feedback loop. The sample temperature was measured by a Cernox resistor thermometer in contact with the sample holder.

Resistance and *I-V* curves were measured using both dc and pulsed current of different duration (in the range 0.5–1000 ms), with current values going from 10 μA to 8 mA. The resulting values were identical. Although we cannot completely rule out possible Joule heating effects, we consider them highly improbable.

The noise measurements were performed using a low-noise digital current supply for the biasing of the samples and the output voltage signal was analyzed by a dynamic signal analyzer (HP35670A), through a low-noise (PAR5113) preamplifier. The equivalent input voltage-spectral density due to the electronic chain is typically $S_{Vn} \simeq 1 \times 10^{-17}$ V^2/Hz from 10 Hz to 1 MHz with a $1/f$ rise below 10 Hz. The details of the measurement setup are reported in Ref. 27, where the experimental technique used to minimize the electrical noise generated by the contacts is also described.

III. EXPERIMENTAL RESULTS

The resistance temperature dependence of a typical investigated nonsuperconducting NCCO sample is shown in Fig. 1, where, as mentioned above, the most prominent feature is a characteristic upturn signaling a metal-insulator crossover at $T_{\min} \sim 80$ K. This behavior is satisfactorily reproduced in terms of the following expression:

$$R(T) = \frac{1}{G_0 + C \ln T} + AT^2, \qquad (1)$$

where the first term is a contribution describing the tendency to 2D weak localization,[13] and the other one is a contribution associated with a standard metal-like behavior. The same expression has also been used to account for weak-localization effects in ultrathin films of $La_{0.7}Sr_{0.3}MnO_3$.[28] The experimental data points in Fig. 1 have been measured at decreasing temperatures, from 300 to 9 K, with a bias current value of 1 mA. The solid line is the best fit to the experimental behavior obtained using Eq. (1), with G_0, C, and A as free-

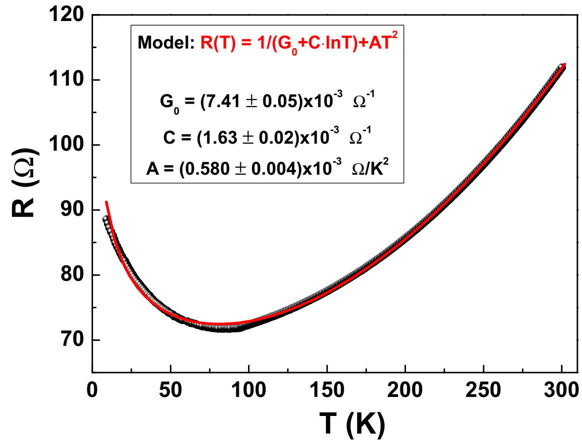

FIG. 1. (Color online) Temperature dependence of the resistance R for a typical nonsuperconducting NCCO investigated sample. Solid line represents the best fit to the experimental data points using Eq. (1). The values of the fitting parameters G_0, C, and A are reported in the inset. The coefficient of determination of the fit is $r^2=0.998$.

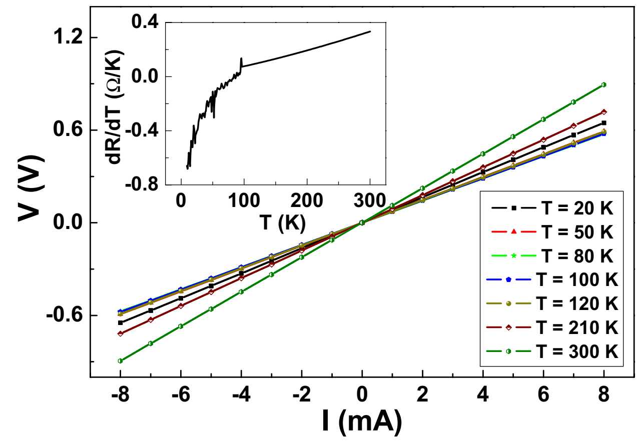

FIG. 2. (Color online) I-V curves, in the temperature range 20–300 K, for the same sample as in Fig. 1. The temperature dependence of the differential resistance is reported in the inset.

fitting parameters. The good agreement between the experimental data and the theoretical expression in Eq. (1), clearly visible from Fig. 1, is testified by a large value of the coefficient of determination. Using the fitting values reported in the inset of Fig. 1, we have verified that the two resistance contributions in Eq. (1) due to 2D weak localization and to a metal-like behavior are dominant at temperatures below 60 K and above 140 K, respectively. Between 60 and 140 K the two conduction mechanisms are comparable, giving rise to the crossover observed around T_{min}. We also point out that in this region the resistivity of our sample is approximately equal to 80 $\mu\Omega$ cm, corresponding to a sheet resistance of $0.026\ 2h/e^2$. This value leads in turn to $k_F l \sim 19$, thus justifying the use in Eq. (1) of the logarithmic correction theoretically predicted in the case of 2D weak localization.[12]

Linear I-V curves have been found in the whole temperature range with an evident Ohmic behavior, as shown in Fig. 2. The linearity of the curves has been verified by fitting the experimental data with a linear function, always obtaining values of the coefficient of determination higher than 0.997. Despite the Ohmic I-V characteristics, a change in the electrical transport mechanisms occurs at T_{min}, as visible in the temperature dependence of the differential resistance reported in the inset of Fig. 2. In particular, we find a metallic behavior ($dR/dT \propto T$) for $T > T_{min}$, and a decrease ($dR/dT < 0$) below T_{min}. The performed dc electrical measurements thus clearly suggest the existence of two distinct transport regimes: a metallic one above T_{min}, exhibiting conventional features, and an "insulating" one at low temperatures, most probably developing as a consequence of weak-localization effects.

In order to better understand the physical mechanisms behind the transport properties in our unreduced NCCO films, voltage-noise analysis at different bias currents and temperatures has been carried out. Figure 3 shows the voltage-spectral densities S_V, at four temperatures, for the same NCCO sample of Figs. 1 and 2, for different current levels. Apart from a number of peaks at definite frequencies due to external noise sources, a general trend is observed in all spectra: as the frequency is increased from few Hz to 100 kHz the spectral-noise density amplitude first decreases with a linear slope and then becomes constant. We have fitted all the spectra with the expression

$$S_V(f,I,T) = \frac{G(I,T)}{f^{\alpha(I,T)}} + c(I,T), \tag{2}$$

which corresponds to what is generally expected for $1/f$ noise in conductors. We have verified that all the curves could be well fitted by choosing constant values for α and c ($\alpha=1$ and $c=1.4\times10^{-17}$ V^2/Hz). As a reference, we show in Fig. 3 the fitting curves in the case $I=1$ mA for each of the measurement temperatures. This implies that the whole temperature and current dependences could be ascribed to the $1/f$ coefficient G only. The fitted value of c is comparable with the background readout electronic noise of our measurement system. Only in few spectral traces, obtained at large bias currents, we observe an additional constant noise contribution at high frequencies, e.g., see Fig. 3 bottom left at $I=8$ mA. We attribute this feature to a temporary increase in the environmental noise during the trace acquisition. As the measured low-frequency $1/f$ noise is much larger than this effect, our analysis is not affected, and thus we can state that the investigated sample exhibits a rather "standard" $1/f$ noise behavior. However, the temperature and current dependencies of the measured spectra are not standard.

In particular, the current dependence is markedly nonlinear, with a parabolic behavior in the high-temperature range and a linear behavior at low temperatures. We recall here that, in the hypothesis of $1/f$ noise generated by resistance fluctuations, a quadratic current scaling of the noise-spectral density is expected. In order to better understand this feature, we have fitted the current dependence of the spectra, estimated at a reference frequency of 90 Hz, as

$$S_V(90,I,T) = \frac{G(I,T)}{90} + c \equiv F(I,T), \tag{3}$$

where for F we use two different functional forms

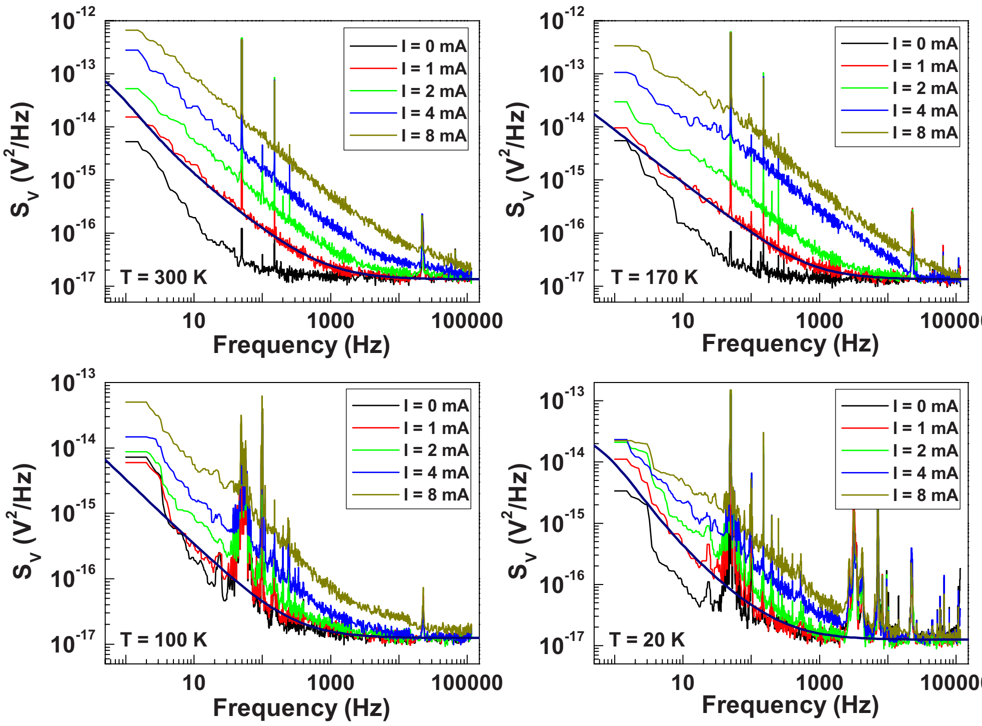

FIG. 3. (Color online) Voltage-spectral traces at four different temperatures (T=20, 100, 170, 300 K) for the NCCO investigated sample. As a reference, best fitting curves (thick solid line) are shown at the bias current value of 1 mA, for each of the considered temperatures.

$$F(I,T) = K(T)I^{\nu(T)} + S_0(T) \tag{4a}$$

and

$$F(I,T) = a_2(T)I^2 + a_1(T)I + a_0(T), \tag{4b}$$

where K, ν, S_0, a_2, a_1, and a_0 are temperature-dependent fitting parameters.

In Table I, the results of the analysis procedure on the spectra recorded at several temperatures are reported in terms of the reduced χ^2 values and of the coefficients of determination r^2. In general, the fits are all satisfactory. However, a closer look shows that above the metal-insulator crossover ($T > 100$ K), the χ^2 values are generally the same for both functional forms of Eq. (4) while at low temperatures ($T \leq 100$ K) the χ^2 values associated with the parabolic dependence [Eq. (4b)] are more than one order of magnitude smaller than those associated with the power law of Eq. (4a). The same effect is also seen with the r^2 coefficients, which similarly show good values in the high-temperature region while the parabolic dependence is the best fit procedure below the resistivity minimum. The temperature behavior of the power exponent ν, shown in Fig. 4, reveals a quadratic current dependence of the $1/f$ voltage-spectral density at $T > 100$ K while a clear change is evident at $T \leq 100$ K, where the values of ν are in the range between 1 and 1.5. In the inset of the same Fig. 4 the temperature dependencies of the other two fitting parameters K and S_0 are reported. While S_0 shows no sign of temperature dependence ($S_0 \simeq c$, the ex-

TABLE I. Reduced χ^2 values and coefficients of determination r^2 resulting from the fit of the spectra with Eqs. (4a) and (4b) at several temperatures for the investigated samples.

T (K)	$\chi^2 \times 10^{-34}$ [Eq. (4a)]	$\chi^2 \times 10^{-34}$ [Eq. (4b)]	r^2 [Eq. (4a)]	r^2 [Eq. (4b)]
20	32.99	1.71	0.962	0.998
50	50.25	1.08	0.956	0.999
80	34.98	2.09	0.969	0.997
100	22.13	2.46	0.972	0.996
120	2.47	1.58	0.996	0.999
170	3.85	3.84	0.989	0.990
210	2.73	2.71	0.994	0.996
300	2.08	2.00	0.997	0.998

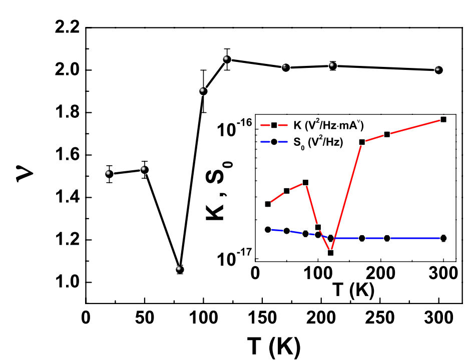

FIG. 4. (Color online) Temperature dependence of the current slope coefficient ν in Eq. (4a); the other two fitting parameters K and S_0 are shown in the inset. The results give a clear indication of an anomalous behavior occurring at the metal-insulator crossover temperature.

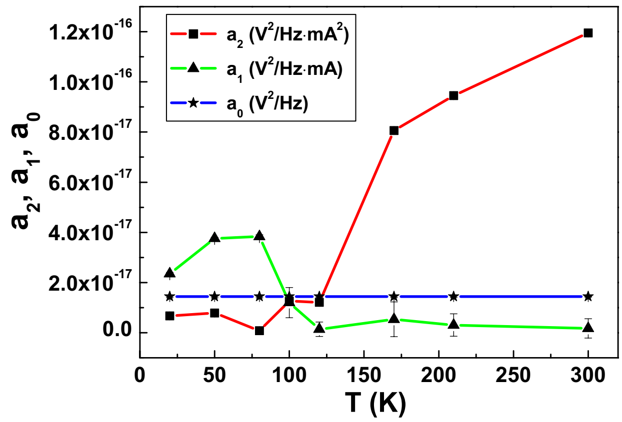

FIG. 5. (Color online) Temperature dependence of the three fitting parameters a_2, a_1, and a_0 appearing in Eq. (4b). A clear sign of a crossover is evident at $T \simeq 100$ K, where weak-localization effects appear producing an upturn of the resistivity.

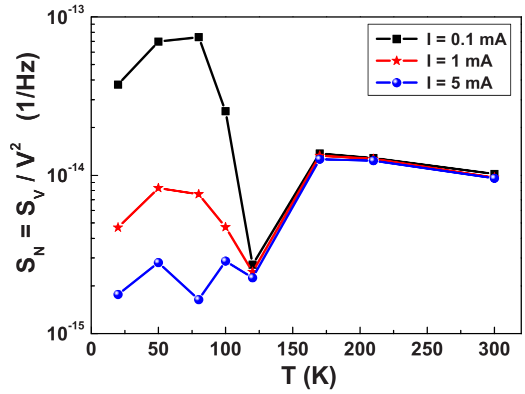

FIG. 6. (Color online) Temperature dependence of the normalized voltage-spectral density S_N at the reference frequency of 90 Hz, for three different bias current values.

ternal noise background), K is clearly temperature dependent with a pronounced dip near T_{min}. These results support the idea that the change in the conduction mechanism associated with the metal-insulator crossover plays an important role in determining also the electrical noise characteristics.

As recalled before, the quadratic current dependence of the noise-spectral density is a standard feature when the $1/f$ noise is originated by resistance fluctuations in Ohmic compounds.[29] However, from the temperature dependence of the three fitting coefficients a_2, a_1, and a_0 of Eq. (4b), an unusual noise behavior emerges in the region around T_{min}. As one can see from Fig. 5, the contribution from resistance fluctuations (proportional to a_2) is strongly suppressed while a new noise contribution, linear in bias current, arises (proportional to a_1). This indicates that, while a predominant quadratic current dependence of the $1/f$ noise is observed in the metallic region well above T_{min}, below the metal-insulator crossover, in the weak-localization region, the conduction channels in the sample are strongly modified. Also here, the parameter a_0 ($\simeq c$) is not temperature dependent because it originates from frequency-independent external noise contributions. The anomalous behavior shown in Fig. 5 has never been observed in the class of compounds investigated here. Moreover, few theoretical models have been formulated on noise effects originating from weak-localization phenomena, all concerning different physical systems.

IV. DISCUSSION

The experimental results reported here show similarities to electrical transport and noise behaviors found in the scientific literature for other systems, undergoing weak localization. An increase in the $1/f$ noise level associated with weak localization has been observed in indium-oxide films and in ZnO accumulation layers.[19,30] In our case, the peculiarity is that the occurrence of weak localization determines the increase, at temperatures around and below T_{min}, of a specific noise component: the non-Gaussian one. This is evident from Fig. 6, where we show how the measured normalized voltage-spectral density, $S_N \equiv S_V / V^2$, has two distinct dependencies on the bias current. Well above T_{min} the $1/f$ noise level is independent of current and weakly dependent on temperature, as expected in the case of pure resistance fluctuations in a metallic sample. Around and below T_{min} the overall $1/f$ noise level depends on the bias current. For small currents the linear term in Eq. (4b) is dominant, giving rise to a noise increase. At high currents the quadratic term in Eq. (4b) is dominant, resulting in an overall decrease in the total noise.

By comparison with the results reported in Refs. 19 and 30, we can conclude that in nonsuperconducting NCCO samples weak localization leads to a new noise component, linear in the applied dc current, which is responsible of an increase in the overall noise level at low bias currents. $1/f$ noise linear in dc current is not commonly observed and has been related to the existence of fluctuating current paths, giving rise to the so-called "dynamical current redistribution" (DCR).[31] DCR can cause large-amplitude non-Gaussian noise for statistically independent fluctuators. Non-Gaussian effects are predicted to be large near and below the metal-insulator crossover.[31]

A nonquadratic current dependence of the noise spectra has also been found by Mantese *et al.*[32] in granular composites of Ni particles in an Al_2O_3 matrix and by Parman *et al.*[33,34] in hydrogenated amorphous silicon. In both cases the noise properties were explained by the presence of inhomogeneous fluctuating current paths.

From a microscopic point of view, we can suppose that the upturn in the resistivity developing in unreduced NCCO samples is originated by weak-localization effects associated with coherent backscattering of the carriers.[14] They are generated by nonstoichiometric oxygen ions randomly occupying a small percentage of impurity apical sites within the ideal apical site-free T' crystal structure. At temperatures higher than T_{min} the phonon-mediated scattering is dominant and the effect of disorder is not visible in the sample resistance. In this region we expect, and find, the usual quadratic current dependence of the voltage noise. At T_{min} the phonon-mediated incoherent scattering and the coherent one are comparable and the current transport in the sample becomes inhomogeneous. In this state the current is divided in different current paths that can easily change under the effect of thermal fluctuations. At low temperatures the coherent backscattering due to disorder becomes dominant, the overall sample resistance increases and the current transport is further divided in many percolating paths. We therefore suggest that the peculiar noise properties measured on our NCCO sample are due to the occurrence, at T_{min} and below, of an inhomogeneous transport scenario induced by 2D weak localization.

Although a solid theoretical justification of this phenomenon is still missing, we underline that the specific and unusual noise properties discussed here could represent a significative signature of a system undergoing a weak-localization-induced metal-insulator crossover. In this context, we believe that the intimate connection between noise and localization could be better understood by investigating the effect on noise measurements of an external magnetic field.

V. SUMMARY

We have investigated dc transport properties and voltage fluctuation processes in unreduced NCCO thin films. The presence of a metal-insulator crossover is clearly evident around a temperature $T_{min} \sim 80$ K. In the metallic region, above 100 K, the conduction and electric noise mechanisms are explained in terms of standard and well-known resistance fluctuations theoretical models. Conversely, at lower temperatures weak-localization effects may have a crucial role in determining the origin of the measured $1/f$ noise. The main consequence is the appearance of a linear current dependence of the $1/f$ noise-spectral density below T_{min}. Our experimental observations have been compared with similar behaviors found in different systems, presenting Anderson localization phenomena. All the reported results give a strong indication that the electrical transport and the noise processes are strictly related to this change in conduction regime.

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