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Thermal and voltage activated excess $1/f$ noise in $FeTe_{0.5}Se_{0.5}$ epitaxial thin films

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We report on measurements of electric transport and voltage noise in $FeTe_{0.5}Se_{0.5}$ epitaxial thin films. Detailed structural investigations demonstrate the high quality of the samples, also testified by a superconducting transition temperature of 18 K, larger than the value observed for bulk compounds. An unusual dependence of the $1/f$ noise is found by varying the applied voltage and the temperature. Above a threshold voltage the $1/f$ noise shows a nonquadratic dependence on applied voltage with a temperature-dependent exponent. Conversely, at low voltages the $1/f$ noise amplitude follows always a quadratic law. In the superconductive transition region, a simple percolative model allows one to estimate the dimensionality of the system, which is found to be two-dimensional, with a critical exponent similar to values reported for other high-T_c superconductors. The experimental findings on noise properties give evidence of unusual transport processes, occurring in this new superconducting material, and can provide useful information on possible conduction mechanisms.

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I. INTRODUCTION

Among the recently discovered Fe-based superconductors, the Fe(Te,Se) system has the simplest chemical and crystallographic structure, while sharing some common features with other families, such as square planar sheets of tetrahedrally coordinated Fe ions in the crystal structure and the similar shape of the Fermi surfaces.[1] The critical temperature can be tuned by the Te/Se composition and reaches a maximum $T_c \simeq$ 14–16 K for 50%Te–50%Se.[2,3] High-quality single crystals[4] and thin films[5] are easily obtained.

All the above characteristics make Fe(Te,Se) suitable for the investigation of transport and superconducting mechanisms in this class of new materials. The study of transport in the normal and superconducting state may yield precious information for the understanding of unconventional superconducting mechanisms, which are to date a matter of debate. For this purpose, using different experimental techniques to probe transport properties is particularly useful, due to the complexity of this material band structure. It is believed that four-five electron-type and hole-type Fe d bands contribute to the electronic conduction.[1] They are fairly narrow and almost compensated bands, which determine an overall normal state conduction that can be described as that of a “bad metal,” with weak temperature dependence. Indeed, among the Fe-based superconductors, the Fe(Te,Se) system seems to be the one with the most localized transport properties.[6–8] Evidence for coexistence of intrinsic localized and itinerant states has been found by electron paramagnetic resonance and NMR studies.[8] The localized states are thought to arise from strong electronic correlations within one of the Fe d bands and they couple with itinerant electrons, thus determining distinctive transport and magnetic properties. Substitutional Te/Se disorder and localized magnetic moments associated with excess Fe may be responsible for localization as well. In thin films, also strain due to growing mode, that can enhance the critical temperature up to 21 K,[5] may act as an additional source of disorder.

Noise measurements are sensitive to all the above-mentioned transport processes and may yield insight on these systems. More specifically, $1/f$ noise is usually associated with carrier fluctuations, carrier trapping and detrapping, and generation and recombination of carriers at grain boundaries in semiconductors[9] and interface-based systems.[10] Hooge was the first to give a relation, accounting for the qualitative noise models. He concluded that the mechanism of $1/f$ noise is always of bulk type, and provided an empirical formula useful to estimate the $1/f$ spectral density magnitude in uniform conductors.[11] However, many experiments performed on semiconductors and metals have contradicted the universal $1/f$ noise mechanism predicted by the Hooge relation.[12] The omnipresence of $1/f$ noise and the universality of its properties have been recently revived by Kazakov, who suggested a universal quantum origin for the $1/f$ occurrence.[13] Even in absence of a universally accepted noise theory, specific processes responsible for $1/f$ behavior have been identified and studied in several systems and innovative advanced materials, such as manganites,[14,15] and superconducting cuprates.[16,17]

The potential of the fluctuation spectroscopy for the understanding of electrical conduction processes in condensed matter has lead us to perform dc transport and voltage-noise investigations on $FeTe_{0.5}Se_{0.5}$ [Fe(Te,Se)] epitaxial thin films. Sample preparation and characterization is briefly described in Sec. II. Section III contains the experimental procedures and results. A tentative theoretical interpretation of the experimental data showing an excess $1/f$ noise is reported in Sec. IV, where a comparison with existing models developed for disordered conductors is made. In Sec. V the analysis of noise near T_c is addressed, providing indication on the dimensionality of the conduction system. The conclusions are in Sec. VI.

II. SAMPLE PREPARATION AND CHARACTERIZATION

The Fe(Te,Se) films were grown by pulsed laser ablation deposition (PLD) in an ultrahigh vacuum system, described in Ref. 18, using a $FeSe_{0.5}Te_{0.5}$ stoichiometric pellet with $T_c =$ 16 K as a target. The samples were deposited at a substrate temperature of about 550 °C and at a pressure of 5×10^{-9} mbar on lanthanum aluminate (LAO) single crystal substrates, using the optimized parameters reported in Ref. 19. The laser (248-nm wavelength) repetition rate was 3 Hz, its fluency 2 J cm^{-2} (2-mm^2 spot size), and the target-substrate distance was fixed to 5 cm. The thickness of the obtained films was about 150 nm.

X-ray diffraction (XRD) analysis showed only the (00l) reflections of the film, with neither traces of elementary oxides nor of the hexagonal phase, indicating the excellent purity of the phase and the optimum c-axis alignment of the growth. *In situ* reflection high-energy electron diffraction analyses and *ex situ* XRD Φ scans indicated the epitaxial growth of the Fe(Te,Se) on LAO with the a and b axes parallel to those of the substrate, without evidence of any other orientation.

The samples investigated here have a critical temperature of 18 K, significative larger than the bulk value. It has been demonstrated that, during the growth and after the coalescence of the islands, a compressive strain develops and enhances the critical temperature,[5] similarly to the effect of an external pressure in bulk samples.[20] This compressive strain is closely related to the film thickness and indeed 18 K is the value expected for a thickness of 150 nm. Overall the films produced were of high quality and with epitaxial orientation.

III. ELECTRICAL TRANSPORT AND NOISE MEASUREMENTS

All the experimental investigations were carried in a closed-cycle refrigerator, operating in the 9- to 300-K range. Temperature stabilization was obtained through a GaAlAs thermometer and a resistance heater controlled in a closed feedback loop to better than 0.1 K. A Cernox resistor thermometer, in contact with the sample holder, was used to measure the sample temperature.

Electrical transport and noise properties of the films were investigated by four-probe contact technique. Two methods were used for the contacts: aluminum wire ultrasonic bonding (see Fig. 1, sample A) and pressed indium (see Fig. 1, sample B). We did not observe any difference in the transport and noise properties with both configurations, thus excluding possible effects due to the contact interfaces. In order to prevent any structural and surface damage, no photolithographic processes were performed. Two thin films with almost the same T_c and thickness, but with different geometry of the contacts and contact pads, were investigated. The phenomenologies observed in the two films were very similar, showing that extrinsic effects could be neglected. All the results reported here are therefore referred to sample A.

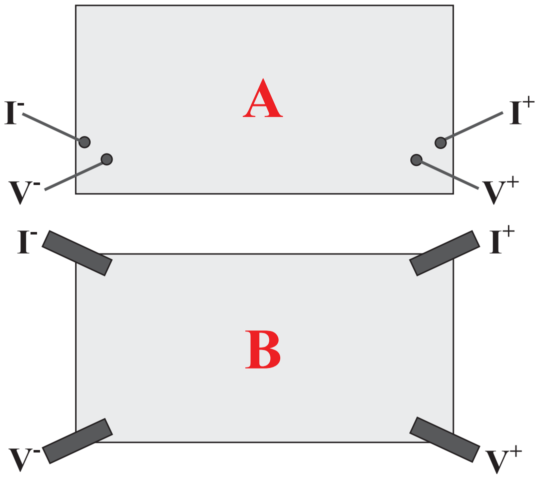

FIG. 1. (Color online) Sample size, 5×10 mm^2. The contacts are made by 25-μm aluminum wire with ultrasonic bonding in sample A, and by 1-mm pressed indium wires in sample B.

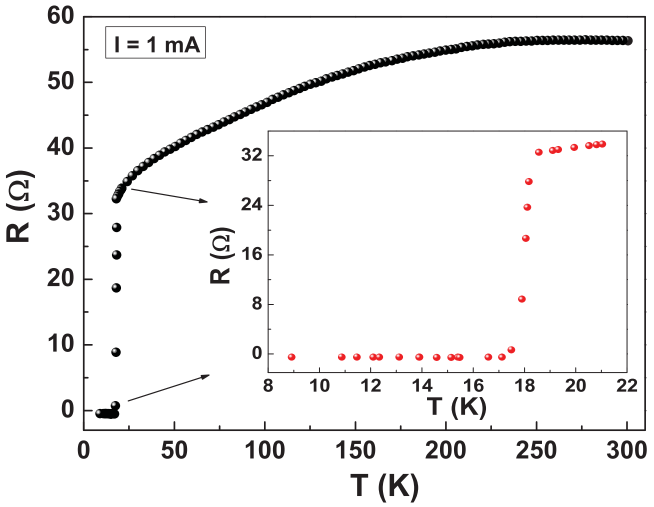

FIG. 2. (Color online) Temperature dependence of the sample resistance R. The full superconductivity is reached at a critical temperature $T_c = 18.0$ K, as shown in the inset.

Resistance versus temperature $R(T)$ curve, shown in Fig. 2, was measured in current-pulsed mode by biasing the samples with a current of 1 mA. As clearly visible in the inset to Fig. 2 , a superconducting transition is found with a critical temperature $T_c = 18.0$ K (defined at 50% of the normal state resistivity). As stated before, this value is very close to the highest values reported in literature for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ thin films, and is larger than 16.2 K observed for bulk compounds.[5,19,21]

Linear I-V characteristics were measured in the whole temperature range from T_c to 300 K and the linearity of the curves was verified by a linear fit of the experimental points, obtaining values of the coefficient of determination larger than 0.998. The data were collected using pulsed current biasing with different pulse duration (from 0.5 to 1000 ms), with identical results. This rules out the possible presence of Joule heating effects.

After the characterization of the dc transport properties, low-frequency voltage-noise analysis was performed at various bias currents and temperatures. All the noise measurements were done using a low-noise dc current bias source and the resulting voltage was sent to a dynamic signal analyzer (HP35670A), through a low-noise preamplifier (PAR5113). The spectra were acquired in the 1- to 100 000-Hz frequency range and the overall instrumental background noise was typically 1.3×10^{-17} V^2/Hz. In order to rule out the existence of electrical noise generated by the contact probes, the experimental technique described in Ref. 22 was used.

The typical frequency dependence of the spectral density S_V of voltage fluctuation processes in the investigated thin films

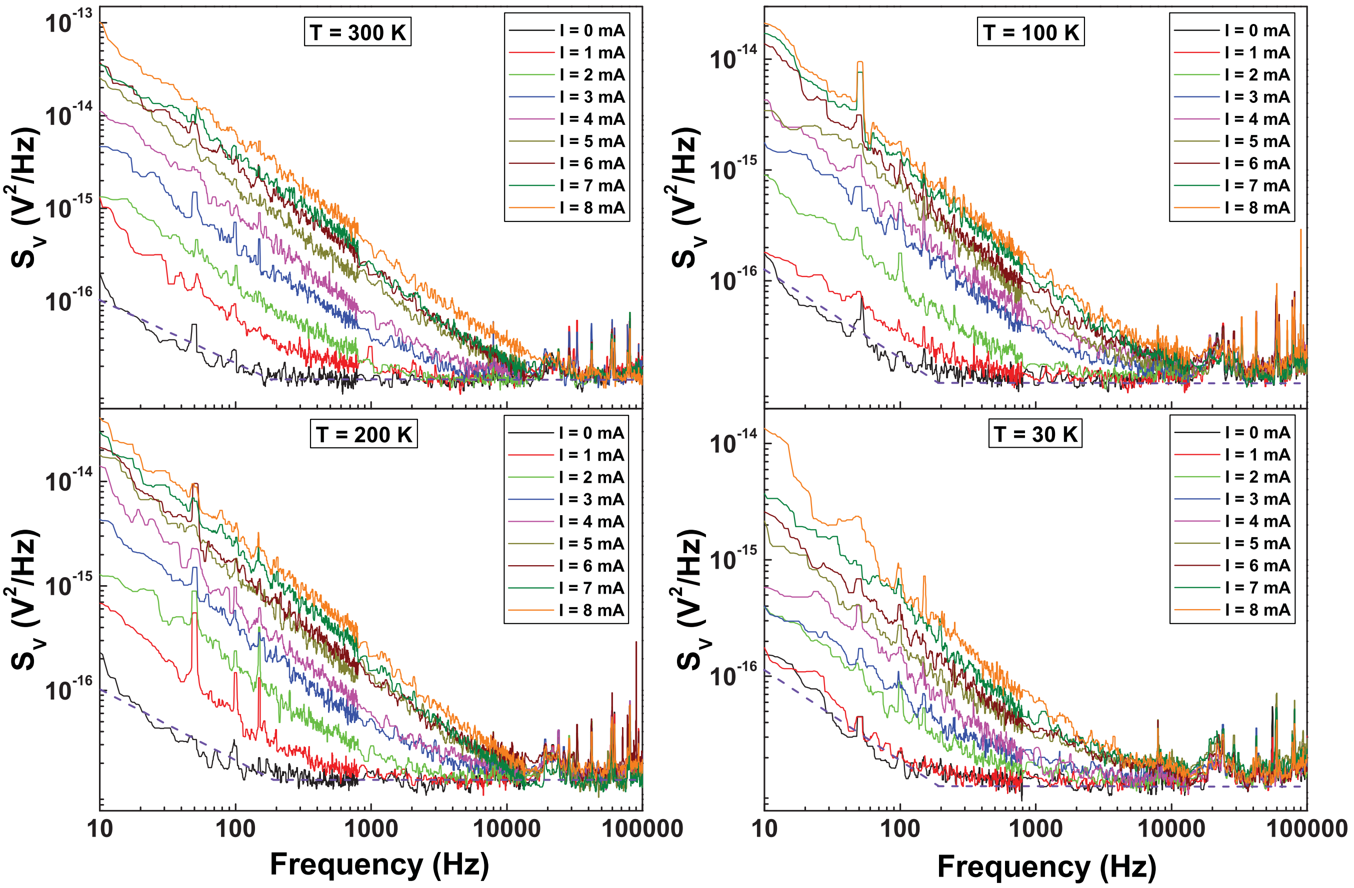

FIG. 3. (Color online) Frequency dependence of the voltage-spectral density at four temperatures and at several currents, in the range 0–8 mA. The voltage-noise background, due to the experimental measurement setup, is also shown as a dashed line.

is shown in Fig. 3. Apart from a number of peaks at definite frequencies due to external noise sources, all the spectral density traces are characterized by two main components. The first one, at low frequencies, has a $1/f$ dependence, and the other is a constant amplitude spectrum, corresponding to our electronic chain noise.

In Fig. 4 the measured $1/f$ noise amplitude at 90 Hz is reported for different bias voltages and temperatures. The data show a clear dependence on the applied bias voltage. In order to better understand this phenomenon, we have fitted the voltage-spectral density with two different functional forms,

$$S_V(90\ \text{Hz}, V, T) = a_2(T)V^2 + a_0, \tag{1a}$$

and

$$S_V(90\ \text{Hz}, V, T) = A(T)V^{B(T)} + C, \tag{1b}$$

where the fitting parameters a_2, A, and B are temperature dependent, while a_0 and C represent the zero-bias constant background noise level. The quadratic behavior, described by Eq. (1a), is usually associated with a “standard” resistance fluctuations model (RFM).[12] Conversely, the power-law dependence of Eq. (1b) can be associated with an excess $1/f$ noise model (ENM).[12] While Eq. (1b) is capable to adequately fit the experimental data for all examined temperatures (see blue solid curves in Fig. 4), the parabolic dependence of Eq. (1a) cannot fit well the data at high temperatures. In this region, the best fit, shown in Fig. 4 as red dashed curves, was obtained using only the low-voltage data, starting from zero and by adding higher values until the overall fit quality deteriorates. This process reveals the existence of a threshold voltage V_0, below which the two models are indistinguishable. Above V_0, and at high temperatures, noise in excess with respect to the “standard” quadratic bias dependence is observed. This feature seems to be intrinsic to the investigated system. Indeed it has been found on two samples and with different configurations of the contact pads, implying different current path geometry and lengths.

To better evidence this phenomenon, in Fig. 5 the relative noise difference $|S_V^{\text{ENM}} - S_V^{\text{RFM}}|/S_V^{\text{RFM}}$ has been plotted versus the applied voltage. A thermally activated process is evident, since at low temperatures (see the 30-K curve in Fig. 5) the two mechanisms give the same noise amplitude. A temperature-independent threshold voltage V_0, where an excess $1/f$ noise starts, can be estimated at (0.22 ± 0.01) V. A similar value, (0.23 ± 0.01) V, has been measured in the sample B. Considering that the distance between the voltage probes is comparable for both samples analyzed, V_0 could correspond to a threshold electric field, above which an additional noise mechanism is activated.

The excess noise is strongly temperature dependent and can be described by the power exponent B in Eq. (1b) as

$$B(T) = b_1 T^n + b_0. \tag{2}$$

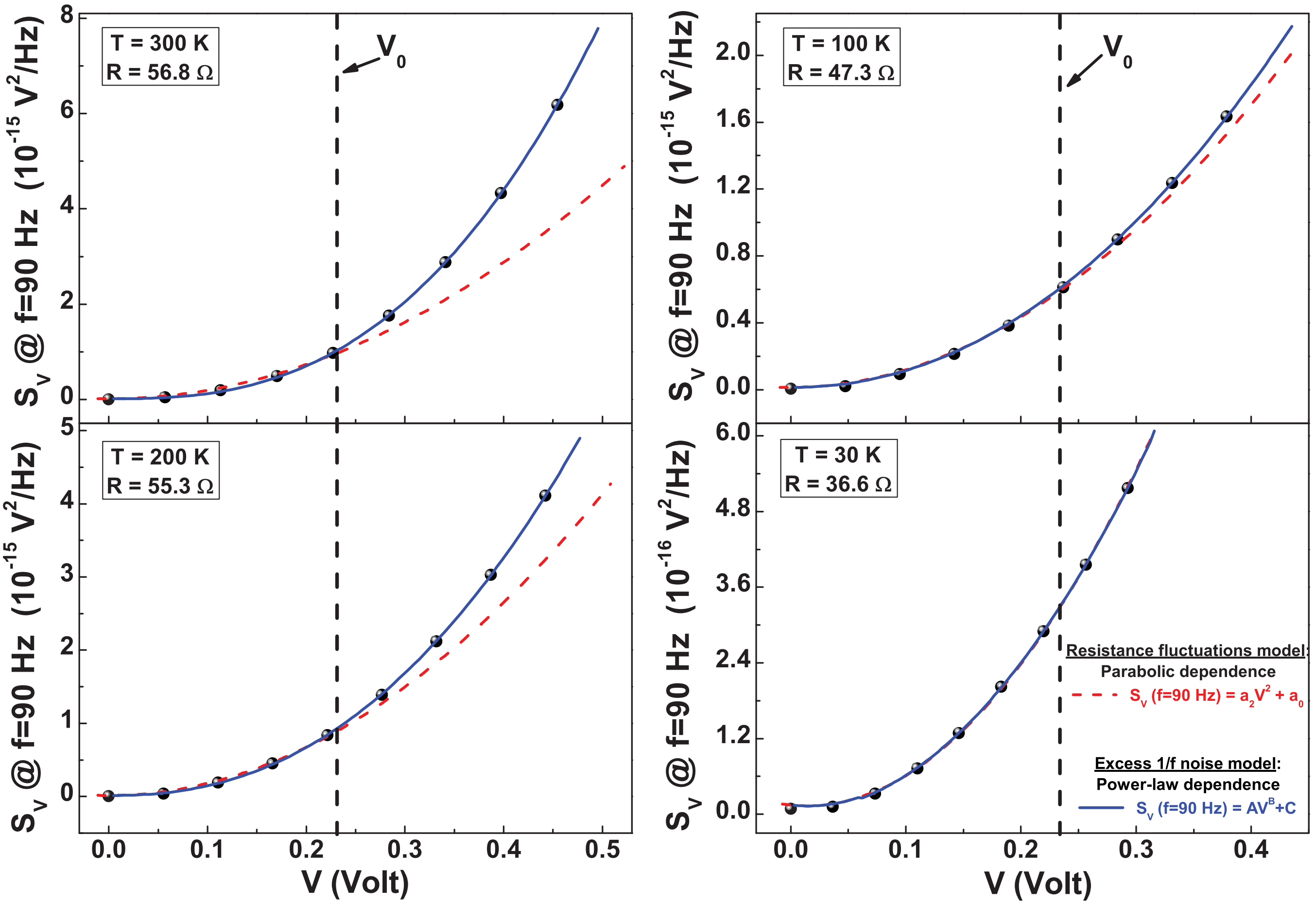

FIG. 4. (Color online) Voltage dependence of the noise-spectral density S_V, estimated at a reference frequency of 90 Hz, for the same temperatures as in Fig. 3. The best fitting curves using Eqs. (1a) and (1b) are also shown as red dashed and blue solid lines, respectively.

The result of the fit with the experimental data is shown in Fig. 6(a). The good agreement is evident and it is worth noting that the best fit values for the temperature exponent n and the constant b_0 are both very close to 2. This has led one to use the following fitting expression:

$$B(T) = \left(\frac{T}{T_0}\right)^2 + 2, \tag{3}$$

where T_0 can be interpreted as a characteristic temperature of the system, whose value is estimated in (367 ± 2) K by the fitting procedure shown in Fig. 6(b). Parenthetically, we note that T_0 may determine an activation energy $E_0 \equiv k_B T_0 \simeq (31.6 \pm 0.2)$ meV. This value is close to the pseudogap energy recently found in iron-based layered superconductors: La($O_{1-x}F_x$)FeAs by Sato *et al.*,[23] LaOFeP by Lu *et al.*,[24] $FeSe_{1-x}$ by Hsu *et al.*[25]. However, to date we are not able to directly relate the scaling temperature resulting from the fit of our experimental data with the occurrence of a pseudogap in the material considered. Other possible mechanisms could explain the observed thermally activated and voltage-induced electric excess noise. A hypothesis involving a superposition of two separate conduction channels is analyzed and discussed in the following.

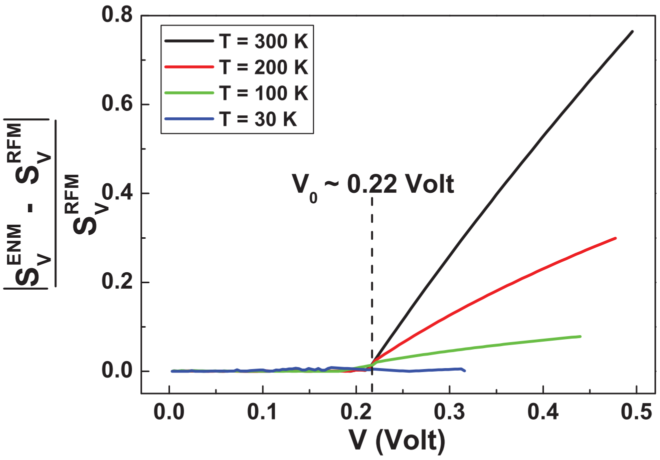

FIG. 5. (Color online) Voltage dependence of the relative difference between the fitting curves of the two theoretical noise models reported in Fig. 4. The threshold voltage V_0, over which the excess $1/f$ noise is activated, is also shown.

IV. DISCUSSION OF EXCESS NOISE

As a rule, in uniform Ohmic conductors the spectral density of $1/f$ noise S_V is proportional to the square of the mean

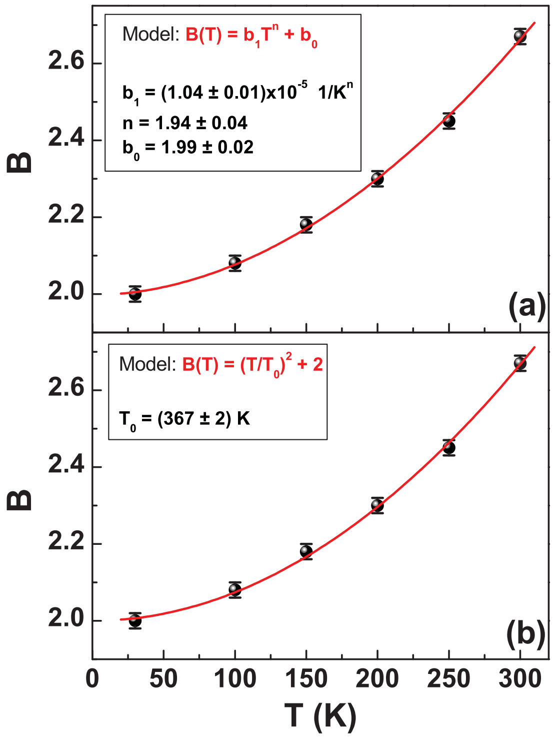

FIG. 6. (Color online) Temperature dependence of the power exponent B in Eq. (1b). A general fitting curve is shown in the upper panel (a), while the specific expression of Eq. (3) is used to reproduce the experimental behavior in the lower panel (b).

voltage V^2, or equivalently the mean current I^2, and is originated by "standard" resistance fluctuations.[12] However, an Ohmic relationship between the mean current and voltage does not always correspond to a linear dependence of the noise on V^2 and different experimental observations, with their specific explanations, have been reported in literature.

Williams and Stone[26] reported experiments on island Pt films where they found spectral density $S_V \propto V^\beta$, with $2 < \beta < 4$. This behavior was interpreted by the existence of very intricate current paths in disordered conductors, which affects the noise much more strongly than the mean quantities (e.g., the current-voltage characteristic).

Nonequilibrium conductivity fluctuations may also result in deviations of the $1/f$ noise power-spectral density from a quadratic current dependence. Nonequilibrium $1/f$ noise measurements on Cr films with high defect concentrations, and on Al/Cu alloy films showed a voltage-spectral density $S_V \propto I^\beta$, with $2 < \beta < 4$ depending on the bias current value.[27] The nonequilibrium component of the noise, activated at high currents, is supposed to be caused by changes in the number of vacancies.[27]

These two mechanisms do not seem to particularly suit the case of our investigated Fe(Te,Se) samples. As reported in the scientific literature, all Fe-based superconductors are characterized by multiple, almost compensated bands contributing to transport, and they can be described as bad metals.[8,28] In iron chalcogenides, the bands crossing the Fermi level are thought to be narrower and with a more localized character than those of other Fe-based superconductor families.[7] Hence the balance of the contributions of each band to the transport properties is even subtler. It has been observed that the Hall effect even changes sign as a function of temperature,[3] indicating that the predominant band is electron-like at low temperature and hole-like at high temperature. Similarly to the existence of a temperature threshold for the hole conductivity to dominate, we suggest that a voltage threshold can also be associated with the onset of a rearrangement in the balance of the contribution to the transport of different channels. Hence we suggest that nonequilibrium fluctuations could arise from the additional hole channel contributing to transport at high temperatures and/or high voltages.

Although a solid theoretical justification of this phenomenon is still missing, we underline that the observed specific and unusual noise properties could represent a signature of the appearance of an additional conductivity channel, controlled by temperature and/or voltage. In this context, we believe that the significant experimental results presented here could be helpful in developing new theoretical models concerning a multiband electronic transport in this class of compounds.

V. NOISE PROPERTIES NEAR T_c

The measured noise increases as the transition temperature is approached, reaches a maximum at T_c, and decreases to zero when the full superconductivity is reached. In Fig. 7 the $1/f$ noise level is shown in the whole temperature range, evidencing a large peak near T_c. This behavior was already observed and reported for other high-T_c compounds.[29] In the transition region the superconductor is a mixture of normal metallic and superconducting phases. The portion of the superconductive phase increases as the temperature is lowered. The simplest model of such a mixture is a lattice of normal resistors, a definite but variable portion p of which are short-circuited. Noise is generated by fluctuations of normal resistors. The relative number of short-circuited links p is

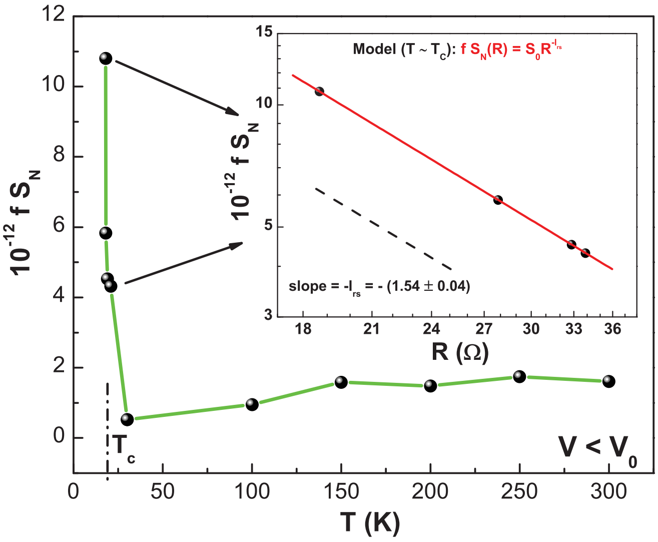

FIG. 7. (Color online) Temperature dependence of the resistance noise level, evaluated for bias voltages $V < V_0$. In order to estimate the critical index l_{rs} of Eq. (4), the resistance dependence of the noise level is shown in the inset above using logarithmic scales.

assumed to increase as the temperature is decreased. In this simple percolation model, the normalized spectral density can be expressed as a function of the resistance R as[12]

$$f S_N \propto R^{-l_{\mathrm{rs}}}, \tag{4}$$

where l_{rs} is a critical index related with the dimensionality of the system. However, in a normal-metal-superconductor mixture this simple percolation model cannot always be directly applied. In particular, Kiss and Svedlindh found that if the portion p of short-circuited resistors is also fluctuating, values of l_{rs} much higher than in the classical percolation framework are obtained.[30] The resistance dependence of the noise level, shown in the inset to Fig. 7, allows us to estimate the critical index $l_{\mathrm{rs}} = 1.54$. This value is in agreement with that reported in Ref. 30, indicating that our Fe(Te,Se) samples behave as two-dimensional systems (the film thickness is smaller than the percolation length) near the superconducting transition.[30]

VI. CONCLUSIONS

We have investigated the dc electrical transport and noise properties of $\mathrm{FeTe_{0.5}Se_{0.5}}$ thin films deposited by pulsed laser ablation technique on lanthanum aluminate single crystal substrates. The good quality of the samples is testified by detailed structural investigations. The superconducting transition is observed at a critical temperature of 18 K, larger than the value reported for bulk compounds. The noise-spectral density traces, recorded at various temperatures and bias currents, show a clear $1/f$ dependence in the low-frequency region. In particular, this $1/f$ noise has unusual dependencies on the applied voltage and/or the temperature. "Standard" resistance fluctuations seem to be the source of the electric noise for low bias voltages and temperatures. Conversely, an excess $1/f$ contribution is clearly found in the high-temperature region above a given voltage, or an equivalent electric field, threshold. This threshold is the same for both samples analyzed, and seems to be an intrinsic feature of the system. The excess noise is characterized by an exponent with a quadratic temperature dependence. The fit of experimental data determines a scaling temperature of 367 K, which could be related to the opening of a pseudogap. Other systems showing similar excess noise do not provide hints for the mechanism responsible for the observed behavior. Alternatively to the pseudogap explanation, theoretical models on multiband electronic transport in iron chalcogenides could give a qualitative description of the investigated phenomenon, by considering an additional hole channel that contributes to conduction, thus enhancing the noise level. However, a well-defined model that accounts for the experimental data is missing. In the superconducting transition region, the noise level can be explained in terms of a "standard" superconductor percolation model. The percolation theory allows one to evaluate a critical index, whose experimental value indicates a two-dimensional behavior of the analyzed samples near the superconducting transition.

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