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Fluctuation conductivity of YBaCuO epitaxial thin films grown on various substrates

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Abstract

The fluctuation conductivity, in zero applied magnetic field, is analyzed for different high-quality *c* oriented epitaxial thin films of $Y_1Ba_2Cu_3O_{7-x}$ grown on various substrates; one ex-situ grown on single crystal $LaAlO_3$ and one in-situ grown on crystalline $KTaO_3$, and one in-situ grown on polycrystalline YSZ. From the temperature dependence of the thermal fluctuation behavior, critical exponents are extracted, without fitting parameters, and the fluctuation dimensionality is deduced. Our findings are explicitly consistent with the thin-film structure of the sample and the conductivity behavior model proposed by Maki and Thompson. The first two epitaxial films show 2D dimensionality with a weak Maki–Thompson pair-breaking mechanism above T_c, and a cross-over to a 3D dimensionality near T_c. Moreover, the insensitivity to different substrate-induced lattice strains suggests that lattice strains do not alter the fluctuation behavior. The findings suggest a percolative mechanism in the polycrystalline film.

1. Introduction

The crystal structure of the new high-temperature Y cuprate superconductors is composed of CuO planes and linear chains between them. This structure is expected to favor a superconductivity of low dimensionality which may be traced in the thermal fluctuations. Information about the latter can be derived from the study of the temperature behavior of transport properties such as electrical conductivity, and thermoelectric power (for a review of both see Ref. [1]) in the proper fluctuation temperature region. Fluctuations also make themselves manifest in the specific heat and susceptibility (for a review, see Ref. [2]).

In cuprate superconductors the high transition temperature T_c, the shorter coherence length ξ, and high anisotropy ($\xi_c \ll \xi_{a,b}$) favor fluctuations further from the transition temperature T_c [3]. The small coherence length, on the order of a unit cell, has as a result a small coherence volume containing few Cooper pairs. This implies that fluctuations have a higher influence on conductivity here than on the classic superconductors; on the other hand this short coherence length implies that superconductivity will be much more sensitive to structural or chemical imperfections in high-T_c materials. Thus, in these materials, fluctuations change the conductivity behavior from a sharp one as in the conventional materials to a rounding one, resulting in a higher conductivity as T_c is approached from above.

The temperature at which this rounding or lowering of the resistivity (or equivalently, an excess of conduc-

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tivity or paraconductivity) starts may be considered as the sign of a higher-temperature superconducting phase. In this temperature region the measured conductivity σ is equal to $\sigma = \sigma_0 + \Delta\sigma$ where σ_0 is the background normal-state conductivity and $\Delta\sigma$ is the component due to fluctuations (other factors such as inhomogeneities have less influence than might be thought [4]). Hence the excess conductivity or paraconductivity is given by $\Delta\sigma = \sigma - \sigma_0$. The background conductivity σ_0 can be estimated by linear extrapolation from temperatures above $2T_c$, since a linear behavior of conductivity with temperature is expected in that region [5].

Although electron pairing above T_c is not favored, because of the higher energy, there are always pairing fluctuations. These fluctuation pairings, at a temperature above T_c (but not so close to it), increase the conductivity to much higher values as T_c is approached and their presence becomes more prominent. This excess conductivity can be expressed by the Aslamasov–Larkin [6] relations and related theoretical approaches such as those by Maki and Thompson [7] and Lawrence and Doniach [8]. From the above theoretical treatment, the dimensionality of the thermal fluctuations can be deduced, as will be shown below.

According to Aslamasov and Larkin (A–L), the increase of conductivity due to fluctuations or paraconductivity $\Delta\sigma$ can be expressed through different relations [6] according to the dimensionality of the superconducting system. Generally one has

$$\Delta\sigma_D = A_D \epsilon^{-\lambda}, \tag{1}$$

where A_D is a temperature-independent parameter, $\epsilon = (T - T_c)/T_c$ and $\lambda = 2 - D/2$ or

$$D = 4 - 2\lambda; \tag{2}$$

T_c is the transition temperature and D the dimensionality of the fluctuating system. Therefore, using Eq. (1) to extract the numerical value of λ, which can be derived from electrical-conductivity measurements and a log–log representation of the data, allows the dimensionality D to be deduced.

The above theoretical approach is complemented by those of Maki and Thompson [7] and Lawrence and Doniach [8]. In order to justify the high value of $\Delta\sigma$, observed mainly in films, Maki and Thompson (M–T) interpreted this effect as due to a decay of the fluctuation pairs into pairs of quasi-particles which continue to be much accelerated. Under a weak pair breaking mechanism, the pairs decay into quasi-particles and vice versa; this leads to an increase of the electrical conductivity which is otherwise limited by strong inelastic scatterers and other pair-breaking interactions (e.g. magnetic impurities).

So, finally for the 3D case, the M–T model gives a contribution larger than that of Aslamasov–Larkin. For 2D it gives an additional term as follows:

$$\sigma_{2D}(\text{M–T}) = \frac{e^2}{8k\hbar d} \frac{1}{\epsilon - \delta_T} \ln\left(\frac{\epsilon}{\delta_T}\right), \tag{3}$$

with d the film thickness and $\delta_T = 2\xi^2(0)/(d^2\delta)$. Here δ is a pair-breaking parameter varying with field and temperature and $\xi(0)$ is the coherence length at $T=0$. A similar logarithmic relation has been recently derived by Maki and Thompson [9]. Through Eq. (3) it is clear that such a contribution increases logarithmically for $\epsilon \gg \delta_T$, i.e. at a large T (far from T_c). Indeed the influence of a M–T term is greater further from the transition temperature while the A–L term dominates closer to T_c, as has been shown [10].

Another approach by Lawrence and Doniach (L–D) [8], for layered superconductors, considers an anisotropic Laudau–Ginzburg model with Josephson junctions between the layers. Dirtiness and large interlayer spacing tend to favor 2D dimensionality while closer to T_c 3D effects tend to dominate.

2. Data-analysis procedure

As mentioned previously, the value of λ can be deduced from the slope of a log–log plot where the best line fitting allows us to calculate the dimensionality D of the superconducting system. A disagreement has arisen among different authors on the value of λ and hence on dimensionality, specially for YBaCuO, where both 2D and 3D fluctuations have been reported although the same data-analysis method or slightly different ones have been used [1]. Apart from experimental factors, e.g. temperature stability, lack of thermal gradient, etc., accuracy of the experimental data, and the "metallurgical" state of the samples [11,12], the correct evaluation of some features of the analysis method may be the reason of this discrepancy. The main errors in the analysis of the fluctuation data stem from uncertainties in knowing the normal-state background conductivity σ_0 (or ρ_0) and the transition temperature T_c. Some basic aspects on the fluctuation analysis and the method followed in this report for the data analysis must be pointed out.

Various authors use more or less different procedures for their data analysis although all are based on the A–L, M–T or L–D relations. For example, different elaborated expressions from the basic ones of A–L [13] or a parameter-fitting process [14] or similars are employed. In this report, the analysis will be based on the basic A–L expression although, instead of $\Delta\sigma$, $\Delta\rho$ is used and the dependence of $d\Delta\rho/dT$ on ϵ is examined in a log–log representation as previously [15,16] has been done.

For the A–L expressions, we need the quantity $\Delta\sigma(=\sigma-\sigma_0)$. Experimentally ρ and ρ_0 or more exactly R and R_0 are the measured quantities through an arrangement where the sample is fed by a constant-current source and the voltage over the sample is measured. But as has been indicated [17] the change of the measured $\Delta\rho$ into $\Delta\sigma$ and the subsequent analysis could lead to data error propagation since an error $\delta\rho$ in resistivity leads to an error $\delta\sigma=-\delta\rho/\rho^2$ in σ and therefore to a large error when T_c has been approached. Since $\Delta\sigma$ and $\Delta\rho$ have the same kind of singularity [17] near the critical temperature the present analysis uses directly the resistance data instead of conductivity and $\Delta\rho\ (=\rho-\rho_0)$ is examined instead of $\Delta\sigma$. In fact, $d\Delta R/dT$ in relation to ϵ is analyzed in a log $d\Delta R/dT$–log ϵ plot and hence from the slope of the fitted line, λ is defined and consequently D is deduced.

The normal-resistivity background is taken from a temperature region where fluctuations are expected to have a quite small effect. Measurements and associated theory show that the effects of superconducting interactions persist to temperatures very much higher than T_c with a cutoff at about $2T_c$ [18]. So as such the region for $T>2T_c$ is taken. In this region the behavior is considered linear although Anderson and Zhou [19] proposed a $\rho=A/T+BT$ law. Some researchers [14] use the background in the fit itself. Others who consider that in granular materials the background resistivity must be weighted by a factor taking into account the current path resulting of intergrain connections. So the normal-state resistivity in the transition region can be obtained by a linear extrapolation of the resistivity curve. But since the correct choice of ρ_0 (or σ_0) is rather relevant, here we analyse $d\Delta\rho/dT$, as is usual in other transitions (e.g. magnetic), since this method allows for smoothing of the background removal [18].

For any fluctuation analysis the value of the critical temperature T_c, is important as it has been shown [20,21] to have drastic effects as ϵ goes to zero. Since the transition in the new superconductors is broad, there is an ambiguity about T_c. Even more for polycrystalline samples a T_c distribution is possible. As has been shown [20] by simulation a finite width of a T_c distribution results in a finite rounding of a log–log plot near T_c which may result in different exponent values even in the mean-field regime. Some researchers leave T_c as a free parameter for the proper fitting. Though early reports adopted the temperature $T_{1/2}$ at which the resistivity falls to its half its normal-state value, the present report uses the value at which dR/dT has its maximum value (or $d^2R/d^2T=0$). These two values generally coincide or are at least close to each other for the best good-quality samples.

In these new materials, a granular character can easily be recognized as a main feature [22]. This results

(1) from an intrinsic disorder which originates from atomic inhomogeneities of different origin (e.g. local deviations from stoichiometry, oxygen deficiency [23], lattice defects, twin etc.) which features are accentuated by the short coherence length and

(2) from an "extrinsic" disorder, due to an assortment of more or less homogeneous grains coupled together in a random way (i.e., through generally not the same but different junctions). In view of the above, T_c may have different values according to the considered transition.

Upon lowering the temperature, one first approaches a critical temperature T_c at which the grains become superconducting. At this temperature, the material resistance drops sharply, since the grains are supposed to possess the larger volume of the sample. This is expected for a good-quality sample (i.e. homogeneous, of low resistivity and sharp transition). In this case, one expects T_c to coincide with the temperature at which dR/dT gets its maximum value.

Upon further cooling for $T<T_c$, Josephson coupling in the weak links begins to overcome the thermal fluctuations, which leads to a phase-locking between the grains. Since the coupling between neighboring grains is influenced by disorder within grain boundaries, pairs of grains may have their own critical temperature $T_{c(ij)}$ at which their junction becomes superconducting [24].

As the temperature decreases further and approaches the different $T_{c(ij)}$ so that the junctions begin to enter the superconducting state, superconducting islands or superconducting "percolation" clusters of grains are created. (The size of these finite clusters is called the percolation correlation length ξ_p). As the temperature further decreases and more junctions switch to the superconducting state, clusters join each other to form larger superconducting regions and ξ_p becomes higher. As the cluster reaches the size of the sample the observed resistivity will go to zero (this is defined as $T_{c,0}$).

According to the above discussion, we consider the temperature where dR/dT has its maximum value as the proper one for the fluctuation analysis, since we are interested in the region close to (but above) the temperature where superconductivity first manifests itself. Apart from the above, different parametrical fittings [25,26] give T_c values that differ slightly from the mid-point or steepest slope of $R(T)$.

3. The material and experimental procedure

Since the early days of the new high-temperature superconductors when the role of the grain boundaries to the transport current was realized, the importance of thin-film materials has been stressed. Indeed, films have provided good-quality materials for different studies, including paraconductivity investigations. From studies on thin films (see Refs. [27–31] for a short collection) different dimensionalities, 3D, 2D or even 1D, were deduced. In this paper, measurements of conductivity for three different high-quality thin films of $Y_1Ba_2Cu_3O_{7-\delta}$ are presented. The films, grown on three different substrates ($LaAlO_3$, $KTaO_3$ and YSZ), were all *c* oriented.

The experimental method for making these measurements is the standard four-point DC technique. These films which are 1000–2000 Å in thickness with the *c*-axis perpendicular, were photolithographically patterned in a proper bridge arrangement [32] (3 mm × 10 μm). Six gold dots were sputtered onto the contact areas in order to allow simultaneous measurements of resistive and Hall signals to be made. Currents of 1 μA, corresponding to a current density in the range of 5 A/cm^2, were systematically reversed to eliminate thermal emf's. After an oxygen anneal at 550°C in 1 atm O_2 to facilitate gold diffusion into the films the resistances of the contacts were less than 0.1 mΩ. This allowed for easy, solder-free mounting and demounting with the use of Au–In–Au pressure pads and spring loaded contacts [33]. Data around the transition temperature have been taken with a temperature sweeping rate of less than 10 K/h (for this rate no hysteresis was observed between warming and cooling) so that quasi-static conditions may be assumed.

4. Fluctuation behavior and discussion

The fluctuation behavior of each of the three thin films will be discussed below together with the main characteristics of the materials. After a description of the film fabrication for each film the experimental data will be presented followed by a commentary discussion.

4.1. Epitaxial film grown on $LaAlO_3$

This YBaCuO epitaxial thin film, grown on the (100) surface of a single-crystal substrate of $LaAlO_3$ has been fabricated by using the BaF_2 method. This consists of simultaneous coevaporation of BaF_2, Y and Cu precursors followed by an optimized "ex-situ" oxygen anneal process [34]. With a proper annealing temperature of about 900°C, epitaxial thin films of 1000 Å in thickness have been grown with smooth laminar morphology and excellent crystallinity, similar to single crystals. In these films, cation alignment and the low pinning behavior indicate an improved epitaxy, such as those observed in monocrystals. The improved epitaxy is in part due to a low lattice mismatch of only 1.71% (YBCO $a = 3.820$ Å, YBCO $b = 3.888$ Å, and $LaAlO_3$ $b = c = 3.788$ Å). The sample with the lowest flux pinning was chosen for the fluctuations conductivity measurements. In addition, RBS yields are between 2 and 3% indicating good-quality samples. The resistivity at 100 K is 160 μΩ cm and the transition width, defined at the half maximum of the temperature derivative, is 0.9 K.

The critical temperature T_c, which is further used in the data analysis, is inferred from the maximum of dR/dT. Its value is 90.25 K which coincides within the experimental error with the mid-point value. Fig. 1 shows a representative data analysis in a $\log d(\Delta R)/$

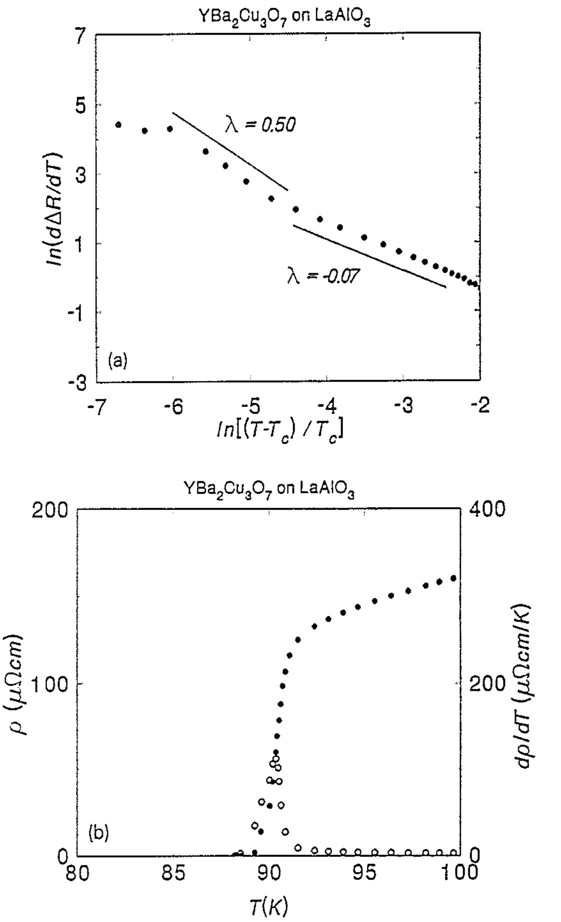

Fig. 1(a). Log–log plot for the ex-situ epitaxial YBaCuO film on $LaAlO_3$, from which values for the critical exponents are deduced. The values of λ are indicated for different temperature regimes. (b) ρ and $d\rho/dT$ vs. T.

ΔT–log ϵ representation. There we get the slope of the fitted line, which according to the relation $d(\Delta R)/dT = A\epsilon^{-(\lambda+1)}$ must be equal to $-(\lambda+1)$; from this the value of λ is deduced and hence through Eq. (2) the value of D. According to this process we distinguish two main regimes of ϵ which correspond to two lines with different slopes or different λ, as is shown in Fig. 1. From $\ln \epsilon = -2$ down to about $\ln \epsilon = -4, 6$, the value of λ is equal to -0.07 i.e. it is equal to zero (the error bar is estimated to be ± 0.003). At about $\ln \epsilon = -4.6$ (or $\epsilon = 0.01$), which corresponds to a temperature interval $\Delta T (= T - T_c) = 1$ K, above T_c, a crossover is distinguished and the slope, down to $\ln \epsilon = -6$, turns to the value $\lambda = 0.50$. For even lower values of $\ln \epsilon$ (temperatures even closer to T_c) a rounding behavior is observed.

Starting from high temperatures, far from T_c, a $\lambda = 0$ value can be deduced from the characteristic of this regime which leads to $D = 4$. This value has been found by other researchers as well in bulk polycrystalline material [16,35] and in single crystals [36], and not only for the fluctuation conductivity but even for the thermoelectric power of this temperature region [37]. As is known from the "singular function analysis", the $\lambda = 0$ value indicates a logarithmic singularity [38] corresponding to a logarithmic variation with temperature. Such a logarithmic behavior, as mentioned above, was foreseen by Maki and Thompson in an elaboration of the Aslamasov–Larkin two-dimensional model by introducing a weak pair-breaking process. The decay of fluctuating pairs into quasi-particles, by pair-breaking mechanisms of intrinsic and extrinsic origins (grain boundaries, lattice imperfections etc.) must be considered, since the lifetime of the pairs is short away from T_c. Moreover in anisotropic superconductors, elastic and inelastic scattering can contribute to pair breaking [39].

Closer to T_c, i.e. for $-6 < \ln \epsilon < -4.6$ the $\lambda = 0.50$ value leads to $D = 3$, a three-dimensional fluctuation superconductivity. Such a dimensionality (3D) for YBaCuO in this temperature region has been found by many researchers. It has been investigated in bulk material of simple YBaCuO composition, in thin films and single crystals [1]. For even lower values of $\ln \epsilon$ ($\ln \epsilon < -6$) or for temperatures still closer to T_c (ΔT less than about 0.2 K), a rounding behavior is observed. Such a picture seems to be common for this temperature regime, since it has been observed or at least can be discerned not only in polycrystal [16,40] and single-crystal material [14], but even in BiCaSrCuO material [17].

4.2. Epitaxial film on $KTaO_3$

Thin films of YBaCuO composition have been also produced epitaxially on $KTaO_3$ (potassium tantalate). These films were grown in situ on the (100) surface of $KTaO_3$ substrates, which are cubic perovskites with an enlarged lattice constant (3.989 Å) inducing lattice strains while maintaining epitaxial growth, by the pulsed laser ablation method (PLA) [41]. The lattice mismatch is now 3.50%. The characteristics of these film include: sharp superconducting transition, transition temperature width 0.3 K, and a T_c defined equal to

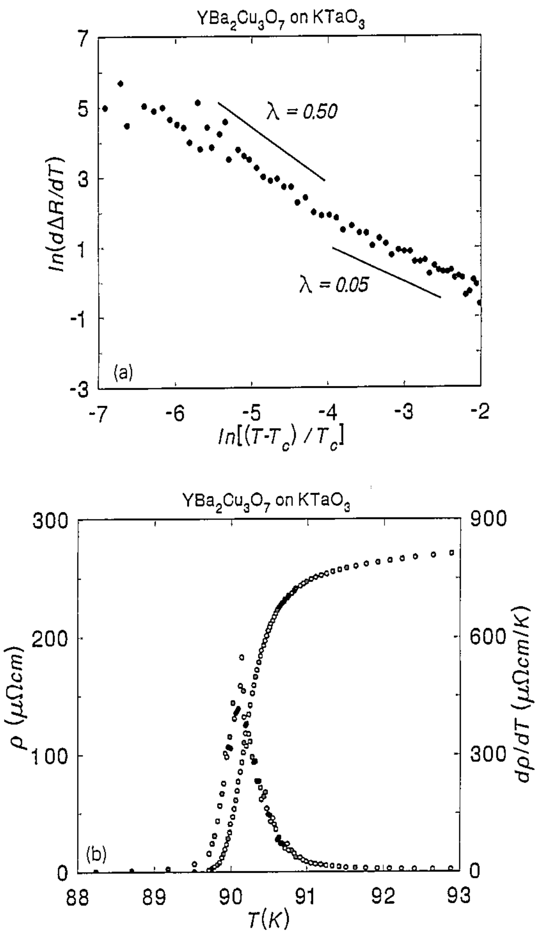

Fig. 2. Same as Fig. 1, but for the in-situ epitaxial YBaCuO film grown epitaxially on $KTaO_3$.

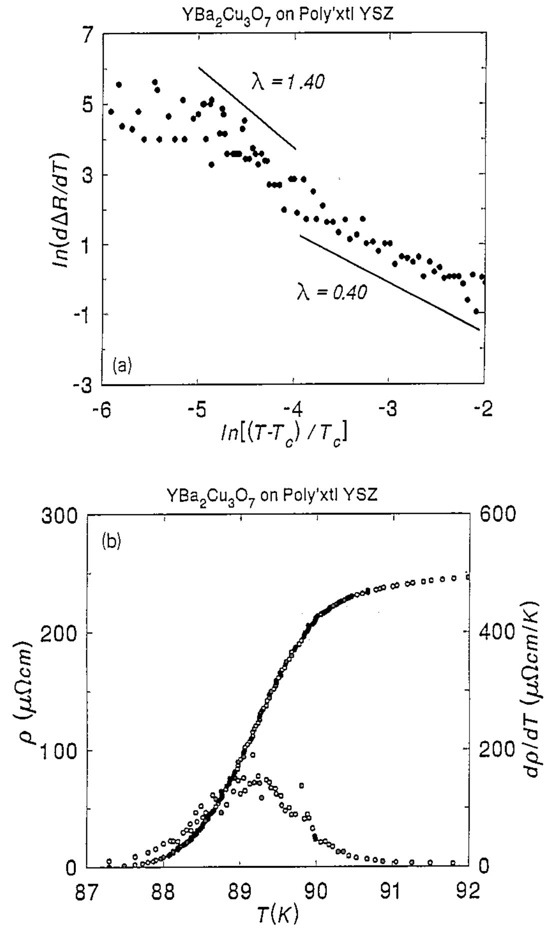

Fig. 3. Same as Fig. 1, but for the polycrystalline film grown on YSZ.

92.15 K (slightly lower than the mid-point value). These features indicate a good-quality thin film in a *c*-axis perpendicular orientation (thickness about 2000 Å). The value of the normal resistivity, $\rho(100\ \mathrm{K}) = 250\ \mu\Omega\ \mathrm{cm}$, comes from slight cracks in the film near the substrate caused from the lattice mismatch (observed in microscopy) leading to errors in geometrical cross-sections and hence to a larger normal-state resistivity.

An identical data analysis, which is depicted in Fig. 2, yields λ values similar to those for films grown on $LaAlO_3$. For $-4 < \ln \epsilon < -2.5$ the value of λ is approximately zero and after a cross-over, which takes place at about the same region ($\ln \epsilon = -4$), the λ equals 0.5 (for $-5.5 < \ln \epsilon < -4$). In general, the behavior of the epitaxial film grown on $KTaO_3$ can be easily compared to that grown on $LaAlO_3$ since they both show a similar fluctuation behavior in the same $\ln \epsilon$ regions. This suggests that lattice strains do not alter the fluctuation behavior. The $\lambda = 0$ value is a sign of a Maki–Thompson logarithmic behavior as pointed out above, and it occurs in about the same temperature interval. Similarly for temperatures closer to T_c, λ crosses over to a value of 0.5 which indicates three-dimensional (3D) behavior. Finally for $\ln \epsilon < 5.5$, the characteristic rounding observed in the "on $LaAlO_3$" epitaxial film can be discerned as well.

4.3. Polycrystalline thin film on YSZ

Unlike the previously mentioned epitaxial films grown on monocrystal substrates, this polycrystalline thin film with *c*-axis perpendicular orientation has been grown on polycrystalline yttria-stabilized zirconia (YSZ) by pulsed laser ablation. This granular film, 200 Å in thickness, consists of large superconducting grains, 1000–10 000 Å in width. Microscopy shows intermittent connections between grains. This film is weak-linked (as demonstrated by a rapid decrease of $J_c(H)$ at low fields [42]) of "SNS-like" type. The normal-state resistivity is higher than the film on $LaAlO_3$, but is similar to that on $KTaO_3$. The transition is more than 2 K wide and the critical temperature, as defined by the maximum of $d\rho/dT$, is equal to 89.2 K, somewhat lower than for the former films. Two different fluctuation regions can also be defined (Fig. 3). One for $-4<\ln\epsilon<-2$ characterized by the $\lambda=0.4$ value and a second for lower temperatures, closer to T_c, i.e. for $-5<\ln\epsilon<-4$, where λ equals 1.4. A cross-over can be distinguished to lie at about $\ln\epsilon=-4$.

Curiously, these values of λ give non-integer values of D. Such values of D can be interpreted as a fractal behavior noticed first in the high-T_c Bi compound [17,43] and later for a different composition of YBaCuO bulk material [12].

5. Conclusions

Generally phase transitions are considered to be blurred by sample inhomogeneities and internal strains. Even more the rounding near T_c has sometimes been thought of as arising from strain contributions, although it is inferred that strains have less influence than could be thought [44]. Such strains are expected to be induced by the substrate in thin films. The results of this report suggests that such strains, different in the two films ($LaAlO_3$, $KTaO_3$), do not alter, at least substantially, their fluctuation behavior.

The first two high-quality epitaxial films, grown on $LaAlO_3$ (3.788 Å) and $KTaO_3$ (3.989 Å), indicate a similar dimensional behavior. For temperatures away from T_c, they both show explicitly a two-dimensional (2D) Maki–Thompson behavior indicative of layered superconductivity with a pair-breaking mechanism present. Such a behavior is consistent with the sample structure (thin film) and the nature of superconductivity. For a temperature regime closer to T_c, their behavior changes to three-dimensional (A–L) which is characteristic of homogeneous superconductors. This trend to higher dimensionality for temperatures closer to T_c, is consistent with an expected increase of the coherence length along c for lower temperatures. As long as the coherence length increases, so that it becomes comparable to the interlayer distance, a 3D conductivity emerges and the superconductor is viewed as homogeneous. Still closer to T_c a common (or "universal") rounding is observed.

The temperature regime of the cross-over for both epitaxial films is similar. For the "on $LaAlO_3$" film it lies at $\ln\epsilon_c=-4.6$ and for the second one at $\ln\epsilon_c=-4$. The first value of ϵ_c is controversial since it corresponds to $\epsilon=0.01$. This is considered by some authors as the low-temperature limit for the mean-field region [45], although there are contradictions on this point [46,47]. Others view this as the low-temperature limit, below which the value of T_c starts to have a drastic influence on the fluctuation analysis [21] (however, the common behavior of these films with different T_c argues against this view). On the other hand such a cross-over in the above temperature region is mentioned in many reports (e.g. Ref. [12]).

The above results are contrasted with previous ones suggesting 2D fluctuations in polycrystal [15] and single-crystal material [48] but others favored 3D dimensionality [49] (in thin films). The results of the present report are closer to those suggesting a Lawrence–Doniach-like behavior characterized by 2D fluctuations with a dimensional cross-over to 3D for temperatures closer to T_c (on single crystals [14] and on highly oriented thin films [27]) as is expected for a quasi-two-dimensional material. The observed behavior in these films, Maki–Thompson 2D fluctuations with a cross-over to 3D, is similar to that observed by Kim et al. [28] and recently by Lang et al. [31]. Concerning the critical 3D XY model suggested recently by Salamon et al. [50]: it is hard to be concluded to here since it is not sure whether the critical region is entered. Although conclusions have been derived sometimes from such measurements [51,52] we believe that other measurements (e.g. under H) will give more reliable results.

In the polycrystalline film grown on YSZ, λ values of 0.4 and 1.40 are obtained for temperatures far and closer to T_c. These awkward values of λ can be justified if the relation (2) is extended to contain non-integer values of D. Indeed, if we accept a percolation mechanism to be effective in this temperature region and consequently we assume that the superconductive system consists of percolation networks, then the system is expected to behave as one of lower dimensionality from the dynamical point of view [53]. Then, for a inhomogeneous state, Eq. (2) may be rewritten as $\lambda = 2 - d/2$ [54] where d is the fracton or spectral dimension [55]. This is valid only if ξ is of the order or less than ξ_p. For larger distance scales the system has to appear homogeneous and fractal effects disappear. In granular material as here, ξ_p should depend on the grain and intergrain size distribution. For temperatures closer to T_c the value 1.4 is well within the average value obtained by Pureur [11] for polycrystal bulk material. Finally, it is remarkable that the crossover temperature (ϵ_c) for this pollycrystalline film is similar to those of the first two films, especially "on $KTaO_3$" thin films.

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