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INFLUENCE OF THE POLYCRYSTALLINITY ON THE PARACONDUCTIVITY IN COPPER-OXIDE SUPERCONDUCTORS

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Abstract

The influence of long length scale structural inhomogeneities on the electrical resistivity of polycrystalline $YBa_2Cu_3O_{7-\delta}$ samples is analyzed on the grounds of an empirical picture proposed by our group some time ago. For that, the electrical resistivity, $\rho(T)$, has been measured in different polycrystalline $YBa_2Cu_3O_{7-\delta}$ samples, all single phase to 4% and with $\delta \lesssim 0.15$, and the results compared with $\rho_{ab}(T)$, the intrinsic resistivity in the ab plane of single crystals with the same nominal composition. Our analysis fully confirms our earlier proposal that the structural inhomogeneities at scales much larger than the superconducting correlation length strongly affects the paraconductivity amplitude, but its reduced temperature behaviour is not affected.

1. Introduction.

In summarizing, in 1978, the effects of fluctuations on the measured electrical resistivity $\rho(T)$ above the superconducting transition in metallic films - effects which had been actively studied for the last 10 years - Kosterlitz and Thouless concluded that the onset of the observed rounding of $\rho(T)$ "may alternatively be a result of film inhomogeneities".[1] However, no quantitative or qualitative justification of that alternative was presented then. In fact, in conventional low temperature superconductors (LTSC's), these possible inhomogeneity effects on $\rho(T)$ above T_{Cl}, the temperature at which $\rho(T)$ around the transition has its inflexion point (see below) have received relatively little attention, although they are often invoked in many works on the critical behaviour around T_{Cl}. This is in contrast with the continued attention to the interplay between other superconducting aspects and inhomogeneities in LTSC's, including the behaviour of the $\rho(T)$ offset (below T_{Cl}).[2,3]

In high-temperature copper oxide superconductors (HTSC), the dilemma between sample inhomogeneities and thermodynamic fluctuations above T_{Cl} was stated by Bednorz and Müller in their seminal work,[4] although they formulated the alternative a way opposite to that done by Kosterlitz and Thouless for LTSC's: after indicating that the observed rounding of $\rho(T)$ above T_{Cl} in La-Ba-Cu-O compounds may be of percolative nature, Bednorz and Müller conclude that "the onset (of the $\rho(T)$ drop) can also be due to fluctuations in superconducting wave functions". Since then, the rounding of $\rho(T)$ above T_{Cl} in HTSC's has been measured in a wide variety of polycrystals, films and single-crystal samples, and the results are fairly well explained in terms of fluctuations of the superconducting order-parameter amplitude (SCOPF) in layered superconductors.[5,6] However, as is still the case for LTSC's, very often it is suggested that inhomogeneities may play an important role in $\rho(T)$ rounding, mainly in polycrystalline samples. In fact, because of the smallness of the superconducting-correlation-length amplitude $\xi(0)$ of HTSC's, of the order of interatomic distances, all the magnitudes may be sensitive near T_{Cl} to different types of inhomogeneities, even when these inhomogeneities exist at small scales. Indeed, as $\xi(0)$ in HTSC's is typically two orders of magnitude smaller than in LTSC's, SCOPF effects will be correspondingly much more important in the former materials.[5-7] It thus seems evident that the interplay between inhomogeneities and SCOPF effects is a topic of considerable interest, the resistivity above T_{Cl} being probably one of the best magnitudes to probe such an interplay.

In previous works we have proposed an empirical picture to account for the influence on $\rho(T)$ above T_{Cl} in copper oxide superconductors of spatial inhomogeneities at scales larger than $\xi(T)$ (here with its geometrical meaning) and temperature independent.[5,6] The influence on $\rho(T)$ of temperature dependent long length inhomogeneities was also analyzed by using effective-mean approaches.[8] In this work we present $\rho(T)$ measurements in single phase (to 4%) poly-

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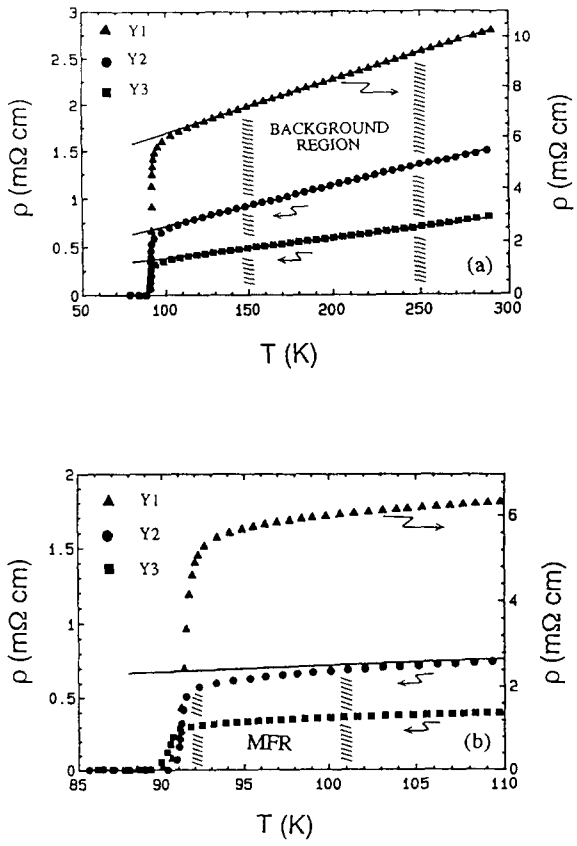

Fig.1.Temperature behavior of the measured electrical resistivity of the three samples studied here. The solid lines represent the background resistivity (of sample Y2 in (b)) fitted in the indicated region. (a) Over all the temperature region measured. (b) Near the transition, showing the mean field region (MFR) of sample Y2. Note that sample Y1 is scaled to the right axis in both figures.

crystalline $YBa_2Cu_3O_{7-\delta}$ samples, and we will show that our empirical picture for temperature independent structural inhomogeneities allows a direct relationship between $\rho(T)$ and $\rho_{ab}(T)$, the intrinsic resistivity in the ab plane of a single crystal of the same chemical composition.

2. Experimental details and results.

Three batches of granular $YBa_2Cu_3O_{7-\delta}$ ceramic samples with $\delta \lesssim 0.15$ were used and they were prepared by usual solid-state synthesis procedure.[9] All the samples are single phase within 4% as shown by X-ray analysis. Optical microscopy measurements and SEM show that the typical grain and crystallite size of our polycrystalline samples are 1 μm to 20 μm. The crystallites show also a high density of twin boundaries at a length scale larger than 1000 Å. The main structural difference between the various samples concerns the pores between the grains. In some of the samples these pores are relatively important, and they gave the same length scales as the grains and crystallites. This porosity reduces the sample average density to 80% of the ideal one.

Figures 1(a) and (b) show the temperature dependence of the measured electrical resistivity of the three samples studied here. The most relevant general characteristics of these $\rho(T)$ curves are summarized in Table I. The notation is the same as in Ref. 5 and 6. In particular, $d\rho/dT$ is the slope of $\rho(T)$ between 150 K and 250 K, a temperature range where $\rho(T)$ may be fairly well approximated by a straight line, with $d\rho/dT > 0$, i.e., where the resistivity of the three samples shows metallic behavior. T_c is defined by $\rho(T_c)=0$, to within the measurement resolution. T_{cI} is the temperature where $\rho(T)$ around the transition has its inflexion point, and ΔT_{cI} is the upper half-width of the resistive transition.[5] Note that whereas T_c will be appreciably affected by the granular nature of our samples, T_{cI} is expected to be close to the mean-field-like normal-superconducting transition temperature of the grains.[5,6,8,10] So, the differences in T_{cI} for the various samples in Table I are probably due to small differences in their oxygen content. Much more important are the differences in their normal dc resistivity, ρ(300 K), or in the temperature slope of $\rho(T)$ in the normal region far away from T_{cI}.

3. The influence of the structural inhomogeneities.

The differences in $\rho(T)$ indicated in § 2 may be easily explained in terms of a phenomenoogical picture, similar to that first proposed by Kirtley and coworkers for granular LTSC's[11] that takes into account the presence in our granular samples of structural inhomogeneities at length scales much bigger than any characteristic length relevant for SCOPF as, for instance, the superconducting correlation length in all directions, $\xi(T)$, the effective interplane (CuO_2) distances, d_e, or the electrical carriers mean free path, ℓ.[5,6] In this picture, above T_{cI} each crystallite is supposed then to behave as a (random oriented) single crystal. The measured resistivity above T_{cI} and in zero applied magnetic field, $\rho(T)$, is related to the intrinsic resistivity in the ab-plane of an ideal single crystal, ρ_{ab}, by[5,8]

$$\rho(T) = \frac{1}{p}(\rho_{ab}(T) + \rho_{ct}) \qquad (1)$$

The fact that $\rho(T)$ depends only on $\rho_{ab}(T)$ is because the resistivity in the c direction is orders of magnitude larger than in the ab plane.[12,13] In

Eq.(1), p ($0 < p \leq 1$) is associated both with a reduction of the cross section area of the sample, due for instance to sample porosity, and with a path lengthening due to the random orientation of the ab planes of the different grains or twinning domains. ρ_{ct} is an average resistivity that accounts for the contact resistance between different long-scale sample domains (grains, untwinned domains, etc). The differences in p in Table I are, therefore, mainly correlated, as indicated before, with sample porosity differences. The way we have rewritten Eq. (1) (compare with the equivalent expression in Refs. 5 and 8) is to emphasize the fact that the intergrains electric contact resistance of the three samples studied here must be relatively similar. This qualitative conclusion is in good agreement with the ρ_{ct} values of Table I. Indeed, ρ_{ct} below T_{CI} must become temperature dependent, in order to get $\rho(T_C) = 0$. In that case, Eq. (1) clearly shows the distinct nature of T_C and T_{CI} in granular samples: whereas T_{CI} concerns only the term $\rho_{ab}(T)$ associated with the grains, T_C concerns $\rho_{ct}(T)$.

The coefficients arising in the measured electrical resistivity and associated with the sample polycrystallinity, p and ρ_{ct}, are extracted for each sample by comparing its normal resistivity far away from the transition (150 K $\leq$ T $\leq$ 250 K), to avoid the presence of critical phenomena,[5] with the resistivity in the ab plane of a single crystal ($p \approx 1$, $\rho_{ct} \approx 0$) of the same composition. The available $\rho_{ab}(T)$ data in different single crystals are well fitted, in this background region, by[12,14]

$$\rho_{abB}(T) = C_1 + C_2(T) \qquad (2)$$

with $C_1 = (5 \pm 15)$ μΩ cm, and $C_2 = (0.5 \pm 0.2)$ μΩ cm K^{-1}, which corresponds to the average values from the data of Refs. 12,13 and 14. It must be stressed also, that the precise functional form of $\rho_{abB}(T)$ has a relatively small influence on the extraction of the critical contributions.[5,6,8] This is because when analyzing critical phenomena, the precise choice of the background should be of small relevance provided that a high-quality fitting in a wide T-region is realized and also that the extrapolation through the transition is smooth.[16] In Figs. 2(a) and (b) we present the $\rho_{ab}(T)$ curves extracted from the $\rho(T)$ data given in Figs. 1(a)and (b) by using Eqs. (1) and (2), the corresponding p and ρ_{ct} values being those given in Table I and with $C_1 = 0$ and $C_2 = 0.5$ μΩ cm K^{-1}. As expected, the $\rho(T)$ measurements in the three single phase samples having the same nominal composition but very different long-scale structural inhomogeneities

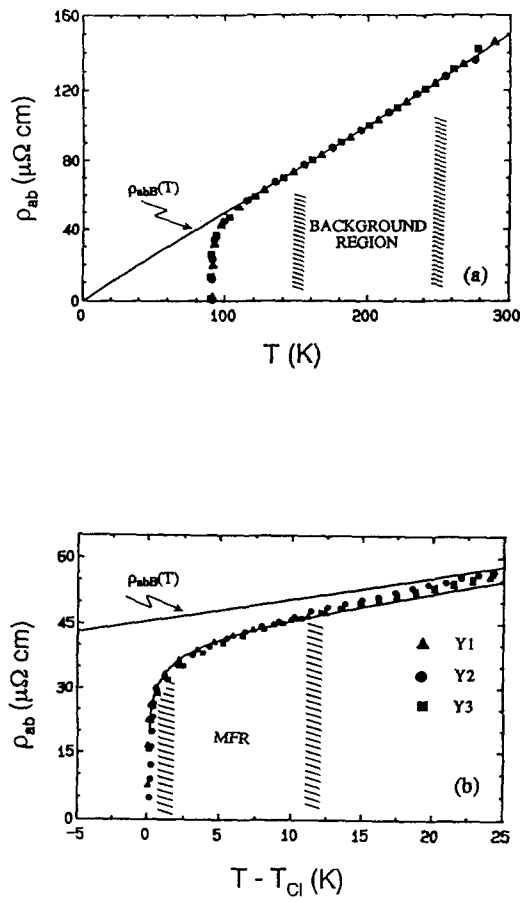

Fig. 2. Temperature behavior of the intrinsic electrical resistivity, extracted from the measured resistivity for each sample used here. The coefficients for the intrinsic background are, $C_1 = 0$ and $C_2 = 0.5$ μΩ cm K^{-1}. (a) General view. (b) Near the transition, showing the average mean field region (MFR) for the three samples. The solid line through the points is the best fit of the Lawrence-Doniach approach.

lead, to within the experimental uncertainties, to the same $\rho_{ab}(T)$ values, which also coincide with those measured in the best single crystal samples,[12,14] over the whole temperature range above T_{CI}.

The solid line in Fig.2(a) correspond to the theoretical Lawrence-Doniach-like approaches[17]

$$\Delta\sigma_{ab}(\varepsilon) = \frac{A_\sigma}{\varepsilon}\left(1 + \frac{B}{\varepsilon}\right)^{-1/2} \qquad (3)$$

where $A_\sigma \equiv g\, N\, A_{AL}$, $A_{AL} = \dfrac{e^2}{16\,\hbar s}$,

and $B \equiv \left(\dfrac{2\xi_C(0)}{d_e}\right)^2$.

Sample	ρ(300 K) (mΩ cm)	T_c (K)	T_{Cl} (K)	$\rho(T_{Cl})$ (mΩ cm)	ΔT_{Cl} (K)	$(d\rho/dT)_{T>150K}$ ($\mu\Omega$ cm K^{-1})	p (10^2)	ρ_{ct} ($\mu\Omega$ cm)
Y1	10.5	89.6	91.3	2.3	0.4	21.5	2.0	53
Y2	1.56	90.6	91.1	0.3	0.3	4.2	11.9	36
Y3	0.81	89.5	90.5	0.2	0.2	2.1	22.7	39

Table I. General electrical resistivity (ρ) characteristics of the samples. T_c is the upper temperature where $\rho(T) = 0$, T_{Cl} is the temperature where $\rho(T)$ has its inflexion point, ΔT_{Cl} is the upper half-width of the resistive transition, and p and ρ_{ct} are coefficients associate to the sample polycrystallinity. The meaning of the other parameters is evident.

In these expressions, g is the number of complex components of the superconducting order parameter and N is the number of CuO_2 planes per unit cell length, s, in the c direction, and d_e is the effective distance between CuO_2 planes. We found $A_\sigma = (350 \pm 100)\ \Omega^{-1}cm^{-1}$, $B = 0.15 \pm 0.08$. A plausible scenario, compatible with these values, is g=1 (i.e., conventional $^1s_0$-wave pairing), $N \approx 2$, $d_e \approx s = 11.7$ Å and $\xi_c(0) \approx 2$ Å. Then, from our $\Delta\chi(\varepsilon)$ results, [10] $\xi_{ab}(0) \approx 10$ Å, and $\varepsilon_G \approx 7\times10^{-3}$.

4. Conclusions.

We must conclude here that the dramatic differences for the paraconductivity in the ab plane, $\Delta\sigma_{ab}(\varepsilon)$, among some of these works,[13,14] with one another on one side, and our results on the other, are just due to differences in the extraction of $\Delta\sigma_{ab}(\varepsilon)$ from the same $\rho_{ab}(T)$ curve. These differences, which have been described in Ref. 6, are mainly associated with the arbitrary use, in some of these works, of a free background and a free mean field critical temperature, T_{CO}. When compared with the extensions of the Lawrence-Doniach approach, our $\Delta\sigma_{ab}$ results seem to be compatible with $^1s_0$-wave pairing if $\xi_c(0) \approx 2$ Å and $\xi_{ab}(0) \approx 10$ Å.

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