

Conductivity of Underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$: Evidence for Incoherent Pair Correlations in the Pseudogap Regime

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A two-channel scenario for the conductivity of underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ is proposed. One is the single-particle excitations channel, which dominates in the optimally doped material, whose resistivity is linear as a function of temperature. The other one gives a contribution which merges the 3D Aslamazov-Larkin fluctuation conductivity at low temperature and obeys a power law at high temperature, depending on two superconductive parameters (T_c and the zero temperature coherence length ξ_{c0}) and an energy scale Δ^* . This allows one to address the nature of the pseudogap in favor of incoherent pairing.

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The variation of the resistivity (ρ) as a function of temperature in high- T_c superconductors (HTSC) remains to be elucidated and, in particular, why it is so different in the underdoped and in the overdoped regions. Most of optimally doped high- T_c cuprates exhibit linear ρ from T_c (as low as 10 K in $\text{Bi}_2\text{Sr}_2\text{CuO}_6$) up to very high temperatures [1]. Among other explanations, it has been proposed that the scattering rate is proportional to the temperature [2]. When the material is underdoped, ρ exhibits no linear dependency anymore but marked downward curvature above T_c . For strong underdoping, an upward curvature usually attributed to localization appears at low temperature. On the opposite, in the overdoped region of the phase diagram, ρ follows a rather more Fermi-liquid behavior. Another peculiarity of most of the underdoped cuprates lies in the density of states in the normal state, which exhibits a depression first discovered by NMR measurements [3], confirmed among others by specific heat [4], scanning tunneling microscopy [5], angle-resolved photoemission spectroscopy [6] measurements, and named pseudogap. The energy scale on which this depression takes place is about the superconducting gap energy, which enables one to think that the pseudogap could be related to superconductivity itself. This is the hypothesis of the particle-particle channel correlations [7–11]. However, other theorists attribute it to correlations in the particle-hole channel such as antiferromagnetic correlations [12], charge stripes [13], and quantum criticality [14]. The physical origin of the pseudogap is thus still highly controversial. In resistivity measurements, the above-described downward curvature is generally ascribed to the pseudogap [15].

The aim of this Letter is to propose a quantitative analysis for the resistivity of underdoped and optimally doped cuprates in the pseudogap regime which strongly suggests that particle-particle correlations survive well above T_c . This analysis lies upon the observation of a continuous behavior from the fluctuation regime of the optimally

doped materials to the high temperature conductivity of the underdoped ones.

The conductivity due to the superconducting fluctuations just above the critical temperature of a superconductor—usually referred to as “paraconductivity” ($\Delta\sigma$)—is strongly related to the coherence length ξ and therefore to the thickness t of the sample with respect to ξ . It was shown by Aslamazov and Larkin (AL) [16] that when t does not exceed ξ , the typical size of the superconducting fluctuating domains is limited by t and $\Delta\sigma$ is given by $\Delta\sigma_{2D}^{AL} = (e^2/16\hbar t)\varepsilon^{-1}$, where $\varepsilon = \ln(T/T_c)$ is the reduced temperature. If no dimension is smaller than ξ , then the fluctuating domains are three dimensional and $\Delta\sigma$ is given by $\Delta\sigma_{3D}^{AL} = (e^2/32\hbar\xi_0)\varepsilon^{-1/2}$, where ξ_0 is the zero temperature coherence length. In 1970, Lawrence and Doniach (LD) [17] proposed a model describing lamellar superconductors, for which $\Delta\sigma$ is three dimensional close to T_c , where ξ is large compared to the layer spacing s , and two dimensional at higher temperatures ($\xi < s$).

In high- T_c cuprates, the range over which $\Delta\sigma$ is important is rather large, because ξ is small and the materials generally have a strong 2D character. Therefore the study of the exact variation of $\Delta\sigma$ constitutes an alternative to evaluate ξ_{c0} (the c -axis coherence length) and to probe the dimensionality (in the LD meaning) of superconductivity in such materials. The early work of Freitas *et al.* [18] and Ausloos and Laurent [19] showed evidence for 3D behavior of $\Delta\sigma$ close to T_c in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ceramics. Oh *et al.* [20] observed the same behavior in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin films with $\xi_{c0} = 1.85 \text{ \AA}$. It is to be noted that similar studies in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ showed 2D behavior in the whole experimental range with no crossover or 3D tendency close to T_c as expected for a more anisotropic material [21]. An open question was the existence of a Maki-Thompson contribution in such materials, and its temperature dependence. Such a contribution was originally proposed in low temperature superconductors [22]

and attributed to the scattering of normal-state quasiparticles by superconducting carriers. Most studies in HTCS to date show no such effect; furthermore, the experimentally measured $\Delta\sigma$ always falls below the theoretical prediction at high temperatures [18,23,24] and this intriguing feature has motivated the analysis presented here.

Optimally doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin films of thickness about 100 nm were grown by pulsed laser deposition as described elsewhere [25]. ρ was measured from T_c to 325 K. $\Delta\sigma$ being defined by $1/\rho - 1/\rho_N$, the main difficulty in such study lies in the correct evaluation of ρ_N : it was deduced from a linear fit between 200 and 270 K.

The data were first adjusted between $\varepsilon = 0.005$ and 0.05 using the 3D AL model for $\Delta\sigma$ and $T_c = 90.3$ K, which leads to a value for the zero-temperature coherence length of $\xi_{c0} = 1.84 \pm 0.02$ Å, consistent with the values found in the literature. The result of the fit is plotted in Fig. 1 (solid line). As can be seen, the adjustment is very good within the fitting range, but $\Delta\sigma$ falls below the theoretical 3D AL model for $\varepsilon > 0.1$. The high-temperature decrease of $\Delta\sigma$ could evoke a 3D/2D LD crossover, but a fit with the LD model does not give the correct value of s . Furthermore, the crossover reduced temperature resulting from the fit is higher than 0.5, which means that the fit has been performed essentially over the 3D part of the LD formula. Moreover, the fit cannot account for the high-temperature decrease of the paraconductivity (see the dashed line in Fig. 1). A bilamellar model, taking into account both the interplane and the interbiplane coupling, as developed by Ramallo and Vidal [26], was unsuccessful in explaining the data. In order to explain the high-temperature behavior, a cutoff was added in the fluctuation spectrum ($\lambda = 2\pi\xi_{c0}/u$) [27] (see the dotted line in Fig. 1). The best fit gave $\xi_{c0} = 1.77 \pm 0.04$ Å and $u = 6.2$, which was not better than the LD model and could neither explain the high-temperature behavior.

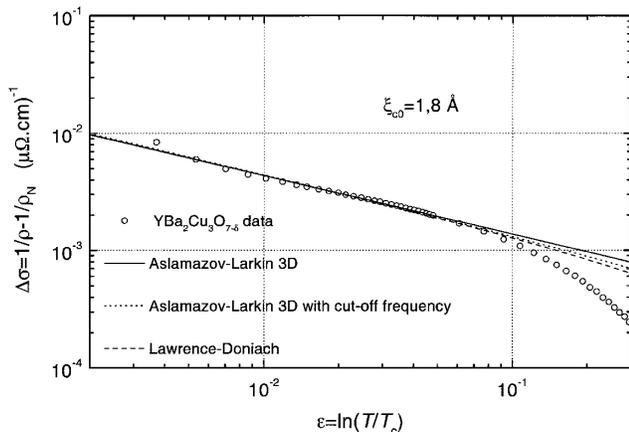


FIG. 1. $\Delta\sigma$ as a function of ε for optimally doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ film. Open circles: experimental data; solid line: AL 3D model with $\xi_{c0} \approx 1.8$ Å; dotted line: AL 3D with cutoff frequency; dashed line: LD model with layer spacing $s = 5 \pm 1$ Å and $\xi_{c0} = 1.82$ Å.

The high-temperature fluctuation regime itself was then analyzed, and the inverse of $\Delta\sigma$ was plotted as a function of T on a log-log scale in Fig. 2 (see the inset). Surprisingly, it exhibits a linear regime between 100 and 160 K, corresponding to $\Delta\sigma^{-1}(T) = AT^\alpha$, where $\alpha \approx 7.5 \pm 0.05$ and $A \approx 9 \times 10^{-13} \mu\Omega \text{ cm}$. This may also be written as $\Delta\sigma(\varepsilon) = \Delta\sigma_0 e^{-\varepsilon/\varepsilon_0}$, where $\varepsilon_0 = 1/\alpha \approx 0.13 \pm 0.01$ and $\Delta\sigma_0^{-1} = 420 \mu\Omega \text{ cm}$. Actually, the low-temperature 3D AL behavior as well as the high-temperature behavior may be modeled by the same simple interpolating function $\Delta\sigma(\varepsilon) = \Delta\sigma_0/\sqrt{2} \sinh(2\varepsilon/\varepsilon_0)$. The agreement of our data with this purely heuristic function is extremely good (see Fig. 2). Moreover, the fit performed only on the high-temperature (not AL) part of $\Delta\sigma$ gives parameters $\Delta\sigma_0$ and ε_0 such that the value of ξ_{c0} deduced from these is exactly $\xi_{c0} = e^2/16\hbar\Delta\sigma_0\sqrt{\varepsilon_0} \approx 1.8$ Å. Hence this function can be written $\Delta\sigma(\varepsilon) = e^2/[16\hbar\xi_{c0}\sqrt{2\varepsilon_0} \sinh(2\varepsilon/\varepsilon_0)]$, where ε_0 is the only adjustable parameter and affects only the high-temperature part of the curve. ε_0 is of the form $\ln(T^\#/T_c)$, where $T^\#$ is a characteristic temperature governing—with ξ_{c0} —the collapse of the superconducting fluctuations. As $\varepsilon_0 = 0.13$ and $T_c = 90.3$ K, this gives $T^\# = 103$ K.

To this point it is important to note that this high-temperature behavior can by no means be attributed to an incorrect choice for the linear ρ_N . If we artificially include the extra resistivity in ρ_N , the value of $\Delta\sigma$ remaining for the 3D fluctuations is too low to be accounted for by 3D or 2D AL theory. Moreover, ρ_N would depend on the value of T_c and would vary exponentially in the vicinity of T_c . We can thus attribute this high-temperature variation to either the superconducting fluctuations themselves or a phenomenon occurring in the vicinity of T_c , affecting the fluctuation conductivity and controlled by ξ_{c0} , T_c , and ε_0 .

Therefore the overall conductivity of the optimally doped thin film is given by

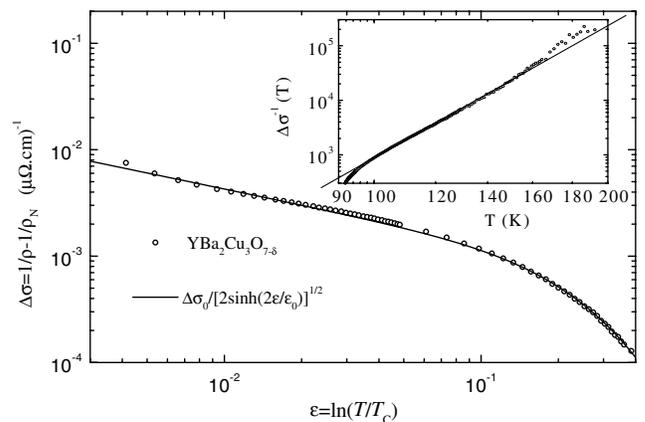


FIG. 2. Fit of $\Delta\sigma$ as a function of ε with the interpolating formula $\Delta\sigma(\varepsilon) = e^2/[16\hbar\xi_{c0}\sqrt{2\varepsilon_0} \sinh(2\varepsilon/\varepsilon_0)]$. The fitting parameters are $\xi_{c0} = 1.84$ Å, $\varepsilon_0 = 0.13$, $T_c = 90.3$ K. Inset: $\Delta\sigma^{-1}$ of an optimally doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin film as a function of T exhibiting linear behavior from 100 to 160 K.

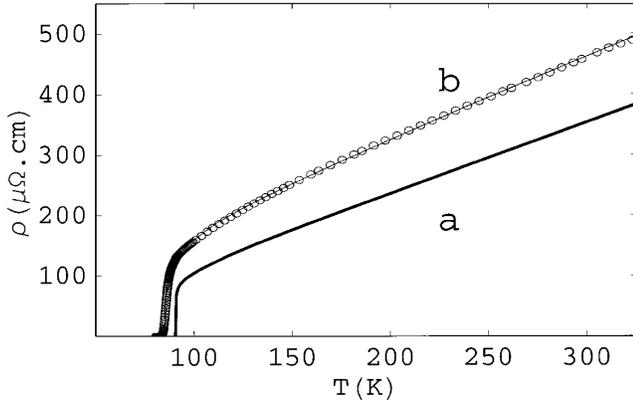


FIG. 3. (a) Resistivity of an optimally doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin film. (b) Open circles: underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin film ($T_c = 85$ K); solid line: interpolating function with parameters $\xi_{c0} = 0.78$ Å; $\varepsilon_0 = 1.34$.

$$\sigma_0 = \rho_0^{-1} = \frac{1}{aT + b} + \frac{e^2}{16\hbar\xi_{c0}\sqrt{2\varepsilon_0}\sinh(2\varepsilon/\varepsilon_0)},$$

where $a = 1.18 \mu\Omega \text{ cm K}^{-1}$, $b = 0$, $\xi_{c0} = 1.8$ Å, and $\varepsilon_0 = 0.13$. It is to be noted that the function $\sigma_0(T)$ admits a linear asymptote only if ε_0 is smaller than 1. For values of ε_0 larger than 1, the second term will dominate at high temperatures and, therefore, there will not be any linear asymptote in the $R(T)$ curve despite the presence of the first term. As a matter of fact, it is a generally established property that the resistivity of underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ shows no linear asymptote. However, it is natural to think that the one-particle excitations with scattering time proportional to $1/T$, which are held responsible for the conductivity of the optimally doped material, remain. This leads one to keep a $1/T$ term and therefore to use σ_0 to describe the overall resistivity of underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ with ε_0 larger than 1.

Let us now consider the case of a slightly underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin film grown by pulsed laser deposition, encapsulated with SiO and Ar-annealed ($T_c = 85$ K). The resistivity of such material is plotted in Fig. 3b (open circles). As usual, ρ from T_c to 300 K shows a slight downward curvature but no linear variation. Nevertheless, it could be fitted with an excellent agreement using the above-defined function $\rho_0(T)$ as is demonstrated in Fig. 3b (solid line). The fitting parameters were $\xi_{c0} = 0.78 \pm 0.01$ Å, $\varepsilon_0 = 1.34 \pm 0.01$, $a = 2.0 \mu\Omega \text{ cm K}^{-1}$, and $b = 60 \mu\Omega \text{ cm}$. This result was extended to the data

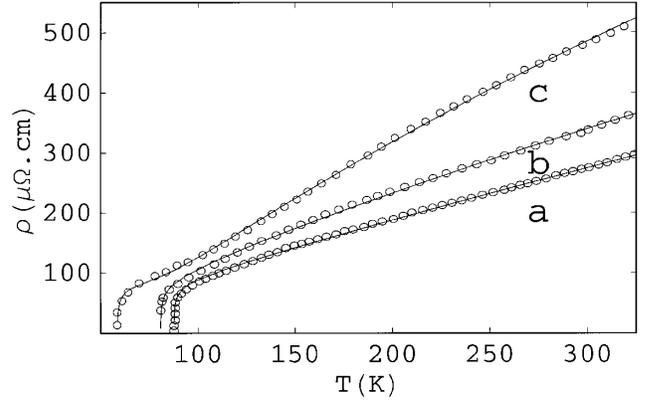


FIG. 4. Open circles: experimental data for underdoped film (from Carrington *et al.* [28]) with (a) $\delta = 0.19$, (b) $\delta = 0.23$, and (c) $\delta = 0.39$; solid line: interpolating function. All parameters are given in Table I.

from Carrington *et al.* on oxygen deficient thin films [28] with an excellent agreement for all the data analyzed as well. Three examples are shown in Fig. 4, for $\delta = 0.19$, $\delta = 0.23$, and $\delta = 0.39$. For the last sample, whose resistivity exhibits upward curvature, a variable range hopping (VRH) term $\rho_1 \exp(T_0/T)^{1/4}$ was added to the linear resistivity component. The fitting parameters are given in Table I. It is to be noted that the sensitivity of the fits to the value of ξ_{c0} and T_c was very high, so ρ has to be determined with great care in order to extract the correct ξ_{c0} . From these results the following three consequences can be outlined:

(i) In optimally doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, the AL superconducting fluctuations are dramatically lowered above 100 K. This is controlled by the parameters T_c , ξ_{c0} , and ε_0 , and the overall resistivity is given by $\rho_0(T)$. The resistivity until room temperature of several underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin films may also be described by $\rho_0(T)$, with a corresponding decrease of ξ_{c0} and an increase of ε_0 . This strong dependency on ξ_{c0} and T_c establishes a link between superconductivity and conductivity in the presence of the pseudogap. This strongly supports the particle-particle correlation channel as an explanation for the pseudogap.

(ii) The fact that the transition itself can still be very well described by an AL model is of great importance: it means that the correlations existing at high temperature (preformed pairs [7] or incoherent pair excitations [9,10,29]) do not alter the system susceptibility with

TABLE I. Fitting parameters for five different samples. The last three are from Carrington *et al.* [28].

Sample	T_c (K)	$a \pm 0.02$ ($\mu\Omega \text{ cm K}^{-1}$)	$b \pm 0.2$ ($\mu\Omega \text{ cm}$)	$\xi_{c0} \pm 0.02$ (Å)	$\varepsilon_0 \pm 0.02$	$T^\#$ (K)	Δ^* (meV)
Opt. doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin film	90.3	1.18	0	1.84	0.13	103	9
Underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin film	85.0	2.02	59.7	0.78	1.34	320	28
$\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin film ($\delta = 0.19$) [28]	87.6	1.15	-0.1	0.79	1.77	510	44
$\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin film ($\delta = 0.23$) [28]	80.7	1.76	-29.8	0.62	2.08	650	55
$\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin film ($\delta = 0.39$) [28]	58.5	4.51	-386.5	0.43	1.97 ^a	420 ^a	28 ^a

^aVRH transport was included ($\rho_1 = 0.41 \mu\Omega \text{ cm}$, $T_0 = 10^5$ K) which might alter the determination of ε_0 .

respect to the order parameter (OP) amplitude fluctuations in the vicinity of T_c . In particular, this possibly rules out particle-particle correlation models based on the *phase fluctuations* of the OP since in the presence of preformed pairs one should not expect standard AL fluctuations of the OP near T_c .

(iii) The third consequence lies in the single-particle excitations themselves. To be able to attribute this nonlinear part of the resistivity to pairing correlations makes it possible to draw conclusions about the single-particle excitations part. The scattering time associated to such excitations still varies as $1/T$ in underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, but their contribution to conductivity is lowered. And for more underdoped samples, an upward variation of $\rho(T)$ attributable to VRH transport indicates that these excitations tend to localize.

The question of the meaning of $\varepsilon_0 = \ln(T^\#/T_c)$ arises naturally. Is $T^\#$ equal to the pseudogap temperature T^* ? The values of $T^\#$ given in Table I are close to usually reported values for T^* [30]. However, there is no general agreement for the determination of T^* , especially as far as transport measurements are concerned. The fit is not very sensitive to a small variation of T^* , but all previous determinations of T^* from resistivity measurements suffer the same uncertainty [15]. It seems here that ε_0 is more to be related to an energy scale Δ^* of about 9–55 meV (see Table I). The orders of magnitude compare nicely to the values of the pseudogap energy estimated by Chen *et al.* [10] in the underdoped region, or to other experimental values (in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8-\delta}$) [5].

Study of the high-temperature part of $\Delta\sigma$ in optimally doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ sheds new light on the resistivity of underdoped cuprates. An expression for the overall *ab*-plane conductivity of the films was proposed from the careful analysis of the high-temperature fluctuations regime in the optimally doped material. It remains valid in the whole underdoping range, with inclusion of localization for the more underdoped sample. It has two components, one presumably due to the single-particle excitations with a $1/T$ scattering time, and one which might describe the particle-particle correlations (with three parameters T_c , ξ_{c0} , and ε_0). The low-temperature limit of this function (close to T_c) is the 3D AL paraconductivity itself.

The analysis developed in the present Letter is consistent with the coexistence of pairing correlations and single-particle excitations such as developed, for example, by Chen *et al.* [10] but not with models for the pseudogap based on particle-hole correlations, as they have no physical reasons to be related to ξ_{c0} and T_c . The presence of AL fluctuations (related to the amplitude of the order parameter) is also more in favor of a three fluids model such as the BCS-BEC (Bose-Einstein condensation) crossover or of modified AL fluctuations than a fluctuating phase model which seems ruled out.

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