

Disorder and Transport in Cuprates: Weak Localization and Magnetic Contributions

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We report resistivity measurements in underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$ and overdoped $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+x}$ single crystals in which the concentration of defects in the CuO_2 planes is controlled by electron irradiation. Low T upturns of the resistivity are observed in both cases for large defect content. In the Tl compound the decrease of conductivity scales as expected from weak localization theory. On the contrary, in $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$ the much larger low T contribution to the resistivity is proportional to the defect content and might then be associated with a Kondo-like spin flip scattering term. This would be consistent with the results on the magnetic properties induced by spinless defects.

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The origin of the strange metallic properties of the cuprates, among which are the transport properties, is still the key feature to be resolved to progress in the understanding of the physics of these 2D correlated electron systems. As the pseudogap is evidenced both in magnetic and transport properties, some interplay between spin and charge degrees of freedom is required. This interplay can be studied as well from the influence of defects and impurities on the transport properties. Point defects such as Zn impurities induce a strong depression of T_c as expected for d -wave superconductivity. This is often considered to result from pure potential (unitary?) scattering [1–4]. However, it has been shown that such spinless defects induce a local moment in their vicinity [5,6]. One can therefore wonder whether these magnetic degrees of freedom play a role in the charge scattering, and induce, for instance, a (Kondo-like) spin flip contribution to the resistivity.

In the presence of Zn impurities, low T upturns of the resistivity with a $\ln T$ variation have been observed in $\text{La}_2\text{Sr}_2\text{CuO}_4$ (LSCO) and $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) [7,8]. However, these observations have required large Zn contents or low hole doping in order to suppress superconductivity. Data were then limited to large values of the 2D resistivity which approach or exceed $\approx 5 \text{ k}\Omega/\square$ for which deviations from weak localization predictions start to be observed in 2D systems [9,10]. These experimental constraints have up to now prevented any thorough study either of the impurity contributions to the scattering or of the applicability of 2D weak localization theory [11].

Recent accurate NMR measurements in the case of Li impurities, which behave similarly to Zn, have evidenced a Kondo-like variation of the induced local moment susceptibility with a Kondo temperature T_K which increases with hole doping [6]. One might expect that magnetic contributions to the transport properties should then differ markedly in underdoped and overdoped systems while localization effects should always be present.

In this work, we address both questions for the first time by comparing underdoped and overdoped cuprates. We have used electron irradiation to introduce in-plane point

defects which are presumably oxygen and copper vacancies. We have previously shown in Refs. [12] and [4] that these defects behave similarly to Zn substitutions in YBCO_7 . As measurements could be performed on a single sample, the accuracy on the defect contribution to the resistivity was high enough to study thoroughly its variation with defect content and temperature, even when T_c is still high. In the heavily overdoped $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+x}$ (Tl2201), we find here that T_c can be suppressed while the low T resistance is still below the strong localization limit. This enables us to demonstrate that weak localization theory applies and that defect scattering is purely elastic in this system. We shall show that this approach fails in underdoped $\text{YBCO}_{6.6}$. The large magnitude of the low T upturns of the resistivity and its variation with defect content rather indicates the occurrence of a Kondo-like inelastic spin flip scattering. The difference between these two systems will be found consistent with the observed doping dependence of T_K . For overdoped Tl2201, the measurements are performed at $T < T_K$ and defects should behave as normal scatterers, whereas for underdoped $\text{YBCO}_{6.6}$ the data are taken above T_K and Kondo-like behavior could occur.

We have taken data on the underdoped $\text{YBCO}_{6.6}$ single crystal, which has already been used in Ref. [4] and on a highly overdoped Tl2201 sample prepared similarly to those of Ref. [4]. In both cases, special care was taken to put the electrical contacts on the edges of the sample to ensure the homogeneity of the current through the sample. $\text{YBCO}_{6.6}$ crystal displayed the usual pseudogap resistivity temperature dependence with an initial T_c of 60 K. The Tl2201 sample, with an initial T_c of 30 K, exhibited a power law dependence of $\rho(T)$ characteristic of the overdoped state [13]. The samples have been irradiated with 2.5 MeV electrons [the irradiation experiments were performed at the LSI (Ecole Polytechnique, Palaiseau)] in liquid hydrogen to allow high electron flux without sample heating. After a short defect annealing (≈ 10 min) either at $T_a = 100, 150,$ or 300 K, resistivity measurements were performed between 20 K and T_a . In such conditions the resistivity versus T curves plotted in Fig. 1 were found

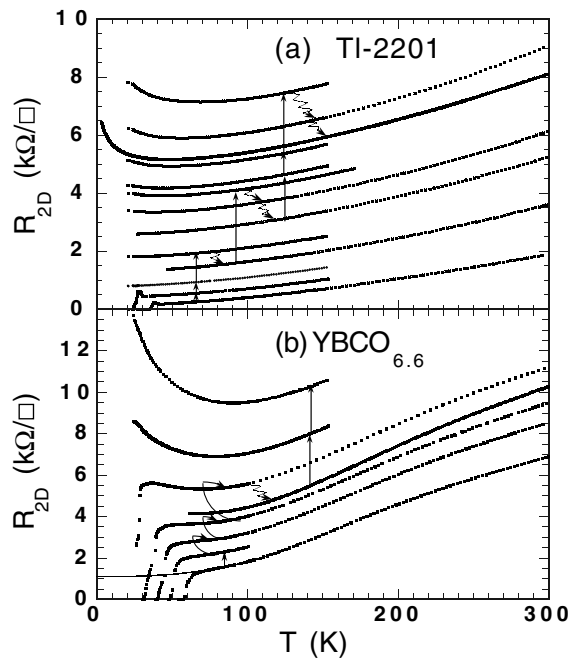


FIG. 1. The measured in-plane resistivity ρ , scaled by the lattice constant c and the number n of CuO_2 planes to give the 2D resistance $R_{2D} = \rho n/c$, is plotted versus temperature for different irradiation conditions. The arrows indicate the sequence of measurements: increasing irradiation for straight arrows and long annealing at room T for wiggled arrows. (a) TI2201: the data down to 3 K has been taken in a different setup. (b) $\text{YBCO}_{6.6}$: the extrapolated low T variation of $R_{2D}^0(T)$ is shown as a full curve.

absolutely reversible, indicating the absence of further evolution of the defect content. The high fluences (10^{20} and $2.6 \times 10^{20} e/\text{cm}^2$, respectively, for $\text{YBCO}_{6.6}$ and TI2201) required to reach large enough resistivities led us to perform several independent irradiation runs. This forced us to remove the probe from the irradiation facility and to let the samples at room T for a few months after each run, which reduced subsequently the concentration of defects [4]. The resistivity curves after annealing at 300 K are quite identical to those observed *in situ* for lower irradiation fluences. This confirms that the measured resistivity is sensitive only to the concentration of in-plane defects. After the last run the samples were removed from the irradiation probe and measurements were then performed down to 3 K in a different setup in which magnetoresistance data could be taken.

As can be seen in Fig. 1 the high T parts of the resistivity curves are nearly parallel to each other in $\text{YBCO}_{6.6}$, i.e., Mathiessen's rule extends to large irradiation fluences [4], which implies also that the hole doping of the CuO_2 planes is not significantly modified. Conversely the variation with T of the resistivity of the TI2201 sample increases with defect content showing that in this case the hole doping changes slightly (less than 10% for the final irradiation) [14]. In both compounds low T upturns of $\rho(T)$ are observed and increase with defect content. They appear at

higher T for $\text{YBCO}_{6.6}$, and correspond to larger contributions to the 2D in-plane resistance R_{2D} than for TI2201 [15].

To see this effect more quantitatively, in Fig. 2 we have reported δR_{2D} , the low T increase of R_{2D} obtained in these two compounds for comparable values of the residual resistances R_{2D}^* [16]. In order to evaluate δR_{2D} the resistivity of the nonirradiated compound had to be extrapolated below T_c . For pure $\text{YBCO}_{6.6}$, two different empirical forms $R_{2D}^0(T) = a + bT + cT^3$ and $R_{2D}^0(T) = a + bT^p$ have been used to fit the data between 80 and 150 K. This lower T limit has been taken to avoid the T range for which superconducting fluctuations contribute to the resistivity. Although their extrapolations differ significantly at low T , the magnitude and T dependence obtained for δR_{2D} are quite identical in the limited experimental range above T_c . For TI2201, superconductivity has been suppressed for this fluence and the low T extrapolation might be more critical than for $\text{YBCO}_{6.6}$. To take into account the slight change of hole doping we have then fitted the high T part of each resistivity curve with $R_{2D}(T) = a + bT^p$. To test the validity of this procedure, two extreme cases have been considered. The fits were performed either between 70 and 150 K or between 150 and 300 K. Although δR_{2D} is naturally found to depart from zero at a slightly higher T in the latter case, the overall variation and particularly the $\ln T$ dependence found at low T is not sensitive to the function chosen for $R_{2D}(T)$. It is clear in Fig. 2 that, whatever the fitting procedure, δR_{2D} is larger and has a steeper T variation in $\text{YBCO}_{6.6}$ than in the TI compound.

We can at this stage compare these data with the expectations of the weak 2D localization theory, which can apply as long as the elastic mean free path l verifies $k_F l \gg 1$ (where k_F is the Fermi wave vector), that is, for $R_{2D}^* \ll 26 \text{ k}\Omega/\square$. The weak localization contributions are expected to become sizable at temperatures for

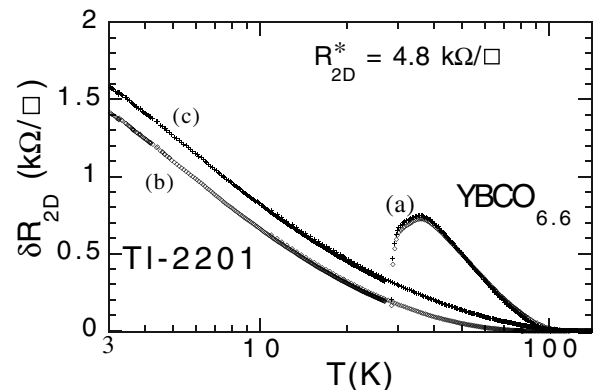


FIG. 2. The low T increase δR_{2D} of the resistivity is plotted for R_{2D}^* of about $4.8 \text{ k}\Omega/\square$ for the two compounds. The different fitting procedures used to estimate δR_{2D} are described in the text: for $\text{YBCO}_{6.6}$ fits with $a + bT^p$ or $a + bT + cT^3$ (a). For TI2201, fit with $a + bT^p$ between 70 and 150 K (b) or between 150 and 300 K (c).

which the elastic scattering rate $1/\tau$ becomes much larger than the inelastic scattering rate $1/\tau_i$. Assuming that the in-plane defects contribute only to the elastic scattering, the ratio τ_i/τ can be estimated from the data, if the inelastic rate is associated with the T dependent contribution to R_{2D} of the starting pure material. For instance, for the residual resistivity $R_{2D}^* \approx 4.8 \text{ k}\Omega/\square$ of Fig. 2, $\tau_i/\tau \approx 10$ – 12 and ≈ 7 , respectively, for Tl2201 and YBCO_{6.6} at 100 K (at 50 K the deduced value would be $\tau_i/\tau \approx 40$ for the two systems). Therefore, within the above assumptions, weak localization conditions are fulfilled even at such high T . Let us recall that the theoretical correction to the 2D conductance has a universal behavior at low T given by

$$\Delta\sigma = \frac{\alpha e^2}{2\pi^2\hbar} p \ln(T/T_0), \quad (1)$$

where p is the exponent of the T dependence of the inelastic scattering rate $1/\tau_i \propto T^p$, and $\alpha = 1$ in the absence of electronic interactions, in good agreement with experimental observations [17–20].

In Fig. 3a $\Delta\sigma = (R_{2D})^{-1} - (R_{2D}^0)^{-1}$ have been plotted for the TI compound. The *in situ* measurements were limited to 20 K, while they could be pursued down to 3 K for the final state of the sample. In this case a good fit with a $\ln T$ dependence extends over a decade. One can notice in Fig. 3a that the curves for the other defect contents are parallel to each other at 20 K, with a slope identical to that found for the lower T run, within experimental accuracy. Finally, at a given T the magnitude of $\Delta\sigma$ increases with defect concentration, as the upturn in the resistivity curve occurs at higher T as can be seen in Fig. 1a. These qualitative observations are more striking in Fig. 3b where we have performed the scaling of these curves in $\ln(T/T_0)$. Here T_0 is found to increase roughly linearly from 70 to 120 K as R_{2D}^* increases from 1 to 7 $\text{k}\Omega/\square$. The slope obtained in Fig. 3b yields the numerical value $\alpha p = 1.3$, when compared to (1). The value of p can be independently estimated from the T dependence of $R_{2D}^0(T)$ and varies from 1.7 to 2.3. This would correspond

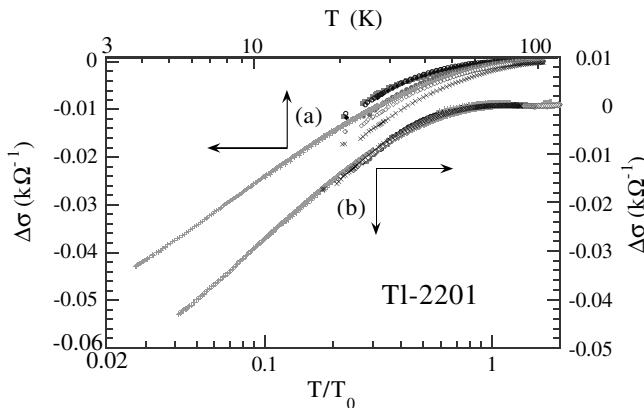


FIG. 3. (a) Plot of $\Delta\sigma$ versus $\ln T$ for different defect concentrations in Tl2201. (b) These curves are scaled by adjusting T_0 in Eq. (1).

to $\alpha \approx 0.7$. These results provide a strong experimental evidence that 2D weak localization theory explains the Tl2201 data.

Transverse magnetoresistance measurements performed up to 8 T after the last irradiation provide an independent evidence which confirms this analysis. It, indeed, exhibits a *negative contribution* which decreases with increasing T as expected for weak localization. The field variation was accurate enough at low T to allow an analysis along the lines of Ref. [18], which allowed us to deduce $\alpha \approx 0.5$ and $\tau_i/\tau \approx 60$ at 4.2 K. While the value of α is in good agreement with the T dependence of R_{2D} , the value of τ_i/τ is smaller than that obtained from the extrapolation of the T dependent part of the resistivity of the pure sample. Such a trend is quite commonly obtained for the magnetoresistance data in various disordered metallic systems [17,19,21]. Further experiments are needed to determine whether interaction effects contribute to $\Delta\sigma$ [22].

Let us consider now the data for the underdoped compound YBCO_{6.6}, for which we have seen that τ_i/τ is weaker or similar, whereas δR_{2D} is larger than for the overdoped compound. In this case superconductivity prohibits the investigation of the low T range, and weak localization contributions are not expected to be fully developed. However, as can be anticipated from Fig. 2, a plot similar to that of Fig. 3 gives much larger values of $\Delta\sigma$ than those found for Tl2201. The slope found at the lowest T for which T/T_0 is only 0.5 would extrapolate at low T to a value $\alpha p > 5$, which is absolutely not sound physically. We could obtain a similar conclusion for electron irradiated YBCO₇ for which $p = 1$ for the well known T linear dependence of $\rho(T)$ [23]. Therefore all these data cannot be explained by weak 2D localization theory, as was also concluded for Zn substitution in YBCO_{6.7} [7].

Alternatively, we can wonder whether spin flip scattering effects can account for these large low T contributions. For weak localization δR_{2D} scales with R_{2D}^2 from (1) as long as $\delta R_{2D}/R_{2D} \ll 1$. On the contrary, a Kondo-like contribution to R_{2D} is expected to scale linearly with the defect content, that is, with R_d , the increase of the residual resistivity. Indeed, one can see in Fig. 4 that $\delta R_{2D}/R_d$ follow the same T dependence for a large range of R_d values, if one omits considering the downturn associated with the proximity of the superconducting transition. Upward deviations with respect to this common behavior begin to occur at low T for $R_d \approx 3.8 \text{ k}\Omega/\square$, that is, $R_{2D}^* \approx 5 \text{ k}\Omega/\square$. This is even more prominent for the largest defect content in Fig. 1b which varies faster than $\ln T$, and might therefore correspond to the onset of strong localization.

It is interesting at this stage to compare these results with that obtained for underdoped Zn substituted samples [7]. In that case refined analyses, as done here, are impossible in the high T range as the substituted sample has to be compared to a pure sample which is physically distinct. Systematic errors due to differences in the sample geometries and hole contents can then be hardly avoided.

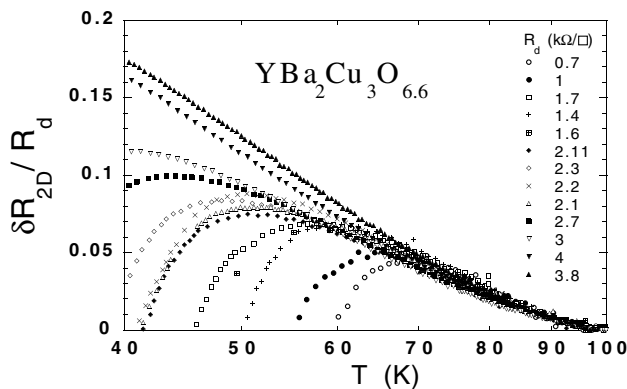


FIG. 4. The low T increase of R_{2D} is normalized to the defect contribution to the residual resistivity R_d in YBCO_{6.6}. Except for the onset of superconductivity, the data are found to collapse onto a unique curve as long as $R_d \lesssim 4$ k Ω/\square .

However, superconductivity could be nearly suppressed with a 18 T field in YBCO_{6.7} with 2.7% Zn [7] which allowed one to evidence a $\ln T$ contribution to the resistivity (not to the conductivity) below 30 K. This would be compatible with a Kondo contribution with a low T_K value. A rough estimate for $\delta R_{2D}/R_d$ yields a value for this $\ln T$ contribution about a factor of 2 smaller than that obtained here. This order of magnitude agreement is quite satisfactory [24], as we are comparing here the influences of different microscopic defects.

In conclusion, we have evidenced that the low T resistivity behaves quite differently in overdoped and underdoped systems. In the former case we have shown the relevance of weak localization theory, as $\Delta\sigma$ has a $\ln T$ variation independent of defect content and in rough quantitative agreement with the exponent of $R_{2D}(T)$. A negative transverse magnetoresistance has been evidenced at low T , but further experiments are needed to determine the eventual contribution of interactions to the $\ln T$ term and the low T variation of τ_i .

We have shown that the large δR_{2D} measured in the underdoped case can be interpreted in terms of a Kondo scattering on the moment induced by the “spinless” defects. This implies *per se* that the scattering by such defects should saturate at low T [25] and should become unitary only below T_K . Therefore, pure elastic scattering by spinless impurities is expected only for $T \ll T_K$. As large T_K values, exceeding 200 K, have been evidenced by NMR only for the overdoped case [6], elastic scattering is expected at low T for these systems. This is then coherent with the occurrence of weak localization contributions in the Tl2201 compound, if the NMR observations for Li impurities do apply as well for irradiation defects.

It is worth noting here that $-\ln T$ contributions to R_{2D} were also observed for the “pure” underdoped LSCO [26] and Bi(SrLa)CO 2201 [27]. They have been associated with a “peculiar” charge localization leading to a metal insulator transition (MIT) with decreasing hole content. It is usually assumed that some disorder is responsible for the

anomalously low value of the optimum T_c in these compounds and that this disorder might be at the origin of the $-\ln T$ term. Our data lead us to suggest that this unknown disorder might result primarily in a scattering on magnetic perturbations induced in the planes, as for spinless impurities. In both cases, one can wonder whether the scattering saturates at low T or whether strong localization proceeds before this saturation can be detected. In other words, the MIT might result from the conjunction of uncontrolled disorder and of the enhanced sensitivity of the underdoped regime to this disorder. This would explain why the MIT is strongly dependent on the cuprate family.

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