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Charge localization from local destruction of antiferromagnetic correlation in Zn-doped $YBa_2Cu_3O_{7-\delta}$

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The in-plane normal-state resistivity of Zn-doped $YBa_2Cu_3O_{7-\delta}$ single crystals is measured down to low temperatures by suppressing superconductivity with magnetic fields up to 18 T. Substitution of Cu with Zn in the CuO_2 planes is found to induce carrier localization at low temperatures in ''clean'' samples with $k_F l > 5$, where the mean free path l is larger than the electron wavelength and thus localization is not normally expected. The destruction of the local antiferromagnetic correlation among Cu spins by Zn is discussed to be the possible origin of this unusual charge localization. [S0163-1829(99)50206-3]

After more than ten years of intense research, the mechanism of high-T_c superconductivity remains to be elucidated, as well as the origin of the peculiar normal-state properties of the high-T_c cuprates.[1] It has been repeatedly pointed out that the antiferromagnetic (AF) spin correlation in the CuO_2 planes may play a fundamental role in the normal-state properties and the superconductivity. Recently, there appeared several experiments which show AF correlation is likely to be fundamentally relevant to the high-T_c superconductivity. For example, incommensurate spatial modulation of AF fluctuation has been reported in superconducting $La_{2-x}Sr_xCuO_4$ (LSCO) (Ref. 2) and in $YBa_2Cu_3O_{7-\delta}$ (YBCO),[3] and the incommensurability is found to be closely related to T_c.[2]

The AF correlation in the CuO_2 planes can be disturbed by partially replacing Cu with other atoms. It is known that partial substitution of Cu (which has spin $\frac{1}{2}$) with Zn (which is nonmagnetic) destroys local AF correlation and thereby quite drastically suppresses superconductivity.[4] On the other hand, the effect of Zn substitution on the normal-state charge transport has been believed to be rather modest; it is reported that Zn substitution simply adds some residual scattering[5,6] and leaves the signature of the pseudogap in transport properties unchanged.[7] Therefore, it would be interesting to look for some fundamental change in the normal-state charge transport caused by the destruction of the local AF correlation upon Zn doping. This becomes particularly intriguing in light of the recently observed logarithmic divergence of the normal-state resistivity in $La_{2-x}Sr_xCuO_4$ (LSCO),[8] in which system presence of a dynamical charge stripe order is discussed.[9,10] The charge stripes intervene in the antiferromagnetically correlated domains, so that the stripes are one-dimensional (1D) defects to the AF correlation; similarly, Zn is a pointlike defect to the AF correlation. If the charge stripe in the LSCO system is the source of the unusual increase in resistivity at low temperatures as recently proposed,[11] then Zn doping might also induce similar unusual localization behavior at low temperatures, although the dimensionality of the defect is different.

In this paper, we report measurement of the low-temperature normal-state resistivity along the CuO_2 planes, ρ_{ab}, of Zn-doped YBCO single crystals by suppressing superconductivity with dc magnetic fields up to 18 T. We found that the Zn substitution induces unusual carrier localization at low temperatures in a ''clean'' system with $k_F l > 5$, where l is the mean free path and k_F is the Fermi wave number. This observation strongly suggests that the local destruction of the AF correlation by Zn doping can severely and qualitatively affect the charge transport in cuprates; therefore, the AF correlation in the CuO_2 plane seems to be indispensable to the peculiar metallic normal-state charge transport.

There are two different Cu sites in YBCO, the chain site Cu(1) and the plane site Cu(2); when Zn is doped to YBCO, it is reported that Zn replaces mostly Cu(2) atoms, leaving the carrier density unchanged.[12] NMR measurements have revealed that nonmagnetic Zn induces local magnetic moment by causing a spin ''hole'' in the AF background of the Cu spins.[4] This local moment gives rise to a Curie term in the magnetic susceptibility at low temperatures.[12] In recent years, the effects of Zn doping on the pseudogap phenomena in the underdoped YBCO have been extensively investigated. Essentially, physical probes that are sensitive to the excitations with $\mathbf{q}=(\pi,\pi)$ [$(T_1T)^{-1}$ in NMR, inelastic neutron scattering, etc.] find that Zn substitution diminishes the pseudogap, while the probes that are only sensitive to excitations with $\mathbf{q}=(0,0)$ (magnetic susceptibility, NMR Knight shift, electrical resistivity, etc.) find that pseudogap signatures are intact upon Zn substitution.[7] The two-dimensional (2D) superconductor-insulator (S-I) transition, which occurs at a critical sheet resistance (per CuO_2 plane) of $h/4e^2$ (=6.5 kΩ), has also been studied in Zn-doped YBCO.[6,13] In this transition, Cooper pairs are conjectured to localize in the presence of disorder;[14] Zn in this case is considered to introduce disorder potentials in the usual sense, though the scattering from Zn is maximally large (unitarity limit).[7,15] Specifically, the S-I transition in YBCO takes place at around 400 $\mu\Omega$ cm in single crystals[6] (a larger critical value of 750 $\mu\Omega$ cm is reported for thin films[13]).

The single crystals of $YBa_2Cu_3O_{7-\delta}$ are grown by flux method in pure Y_2O_3 crucibles to avoid inclusion of any impurities other than Zn. The purity of the crucible and the starting powders are 99.9% and 99.95%, respectively. All the crystals measured here are naturally twinned. The oxygen content y ($\equiv 7-\delta$) in the crystals is controlled by annealing

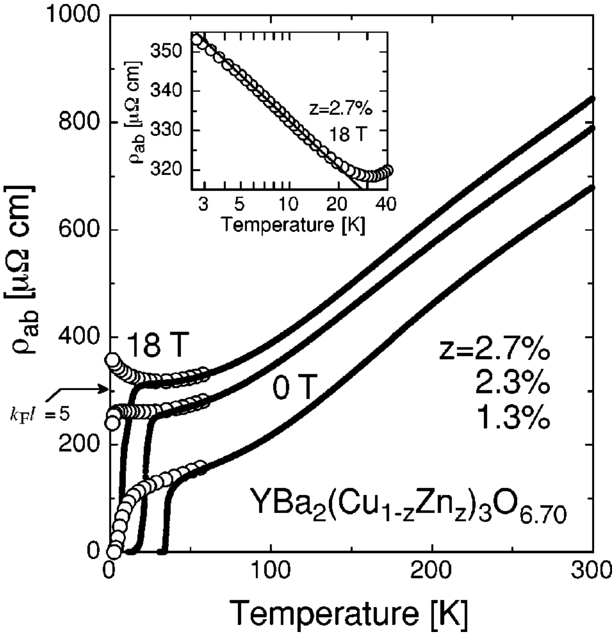

FIG. 1. $\rho_{ab(T)}$ of $y=6.70$ crystals with different z (2.7%, 2.3%, and 1.3%) in 0 T (solid lines) and in 18 T (open circles). Inset: log T plot of $\rho_{ab}(T)$ for the $z=2.7\%$ sample to show the log$(1/T)$ dependence (straight line). The deviation at the lowest temperatures is due to superconducting fluctuations.

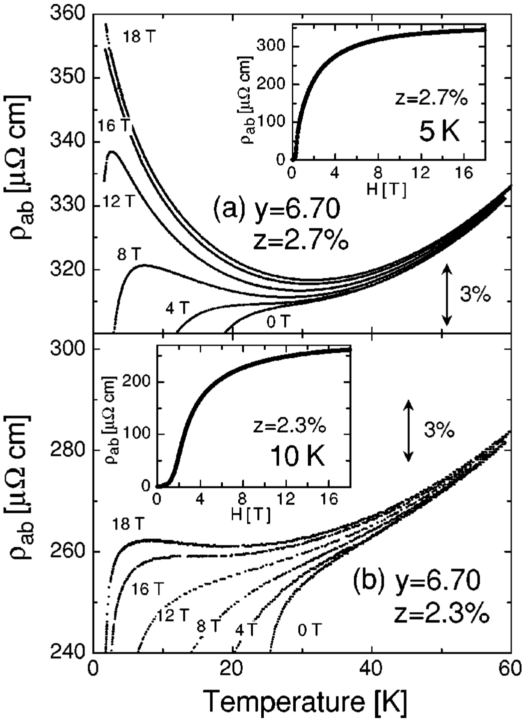

FIG. 2. $\rho_{ab}(T)$ in 0, 4, 8, 12, 16, and 18 T for (a) $y=6.70$, $z=2.7\%$ sample and (b) $y=6.70$, $z=2.3\%$ sample. Insets show the H dependence of ρ_{ab}.

in evacuated and sealed quartz tubes at 500–600 °C for 1–2 days together with sintered blocks and powders, for which the oxygen content is controlled beforehand. The final oxygen content is confirmed by iodine titration method and also by measuring the weight of the sintered blocks. The obtained values from the two techniques are consistent within ± 0.02. To obtain sharp superconducting transition, the quartz tubes are quenched with liquid nitrogen at the end of the annealing. Although it has been discussed that quenching from a high temperature causes oxygen disorder in the CuO chains,[7] the residual resistivities of the quenched samples and slowly-cooled samples are identical for our crystals. Because of the careful control of the purity and the oxygen content, the variation of ρ_{ab} and T_c is less than 5% and 2 K, respectively, and our data are in good agreement with the best data in the literature.[6,7] The measurements of ρ_{ab} are performed with ac four-probe technique under dc magnetic fields up to 18 T applied along the c axis.

The actual concentration of Zn in the $YBa_2(Cu_{1-z}Zn_z)_3O_y$ crystals is measured with inductively-coupled plasma spectrometry (ICP) with an error in z of less than $\pm 0.10\%$. To supplement the ICP result, we compared T_c of fully oxygenated crystals with that of sintered samples where Zn concentrations are known. The results from the two techniques are consistent with each other. The concentration of Zn in the crystals thus determined is roughly half of the content before the crystal growth. The homogeneity of Zn in the crystals is confirmed with electron-probe microanalysis (EPMA).

Figure 1 shows the temperature dependence of ρ_{ab} in 0 and 18 T, for three samples with different Zn concentrations. Note that the three samples have the same oxygen content of 6.70. In zero field, all the samples are metallic $(d\rho/dT \geqslant 0)$ and show superconductivity. We can see in Fig. 1 that the effect of Zn doping in zero field is to reduce T_c and simply add some constant residual resistivity [thereby shifting up the $\rho_{ab}(T)$ curve], as is already reported in the literature. Thus, there is no qualitative difference among the three samples in the zero-field properties. However, under a high magnetic field of 18 T, a drastic difference emerges; the $z=2.7\%$ sample shows a clear upturn in ρ_{ab} at low temperatures, while other samples with smaller z do not show such a clear upturn. We also prepared a slowly cooled sample to check if the observed localization behavior $(d\rho/dT<0)$ in the $z=2.7\%$ sample is due to oxygen disorder; essentially the same localization behavior is observed in the slowly cooled sample, which is supposed to have a smaller amount of oxygen disorder (at the expense of transition width). Thus, for an oxygen content of 6.70, we may conclude that 2.7% of Zn induces carrier localization in YBCO at low temperatures in 18 T, irrespective of the annealing procedure. The inset to Fig. 1 shows a plot of ρ_{ab} vs log T for the 18 T data of the $z=2.7\%$ sample; the temperature dependence of ρ_{ab} is consistent with log$(1/T)$, as in the case of the underdoped LSCO.[8]

Figures 2(a) and 2(b) show the temperature dependence of ρ_{ab} in various fields for the $y=6.70$, $z=2.7\%$ sample and the $y=6.70$, $z=2.3\%$ sample, respectively. The magnetic-field dependence of ρ_{ab} at a representative temperature is shown in the insets. Apparently, 18 T field is sufficient to suppress superconductivity above 5 K in the $z=2.7\%$ sample and above 10 K in the $z=2.3\%$ sample. One may notice in Fig. 2(b) that the 18 T data show a very slight upturn below 15 K, which suggests that this $z=2.3\%$ sample might show a stronger upturn in much higher fields.

By analogy with the metal-insulator crossover observed in the low-temperature normal state of LSCO,[16] one may ex-

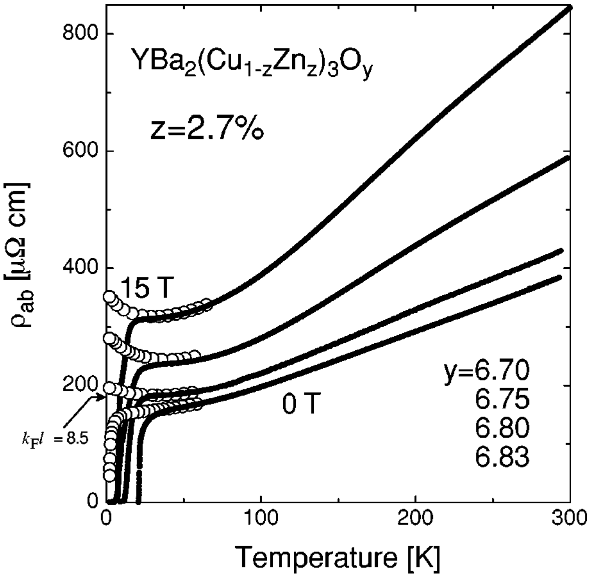

FIG. 3. $\rho_{ab}(T)$ in 0 and 15 T for $z=2.7\%$ samples with $y=6.70$, 6.75, 6.80, and 6.83.

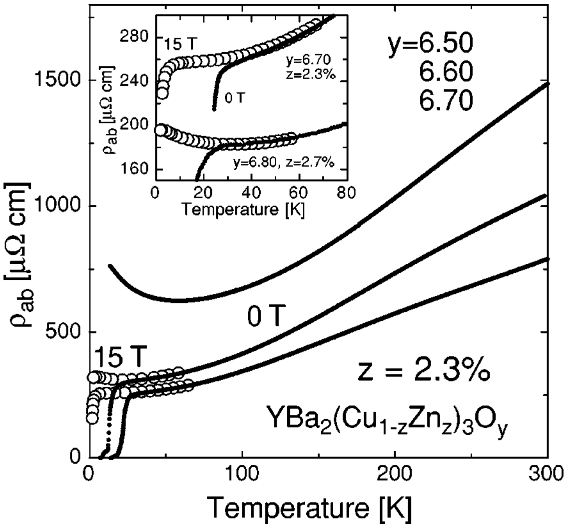

FIG. 4. $\rho_{ab}(T)$ in 0 and 15 T for $z=2.3\%$ samples with $y=6.50$, 6.60, and 6.70. Inset: $\rho_{ab}(T)$ in 0 and 15 T for $y=6.70$, $z=2.3\%$ sample and $y=6.80$, $z=2.7\%$ sample.

pect that increasing oxygen content makes the $z=2.7\%$ sample metallic. This is actually the case, as is shown in Fig. 3, where $\rho_{ab}(T)$ of the $z=2.7\%$ samples with $y=6.70$, 6.75, 6.80, and 6.83 are plotted. When the oxygen content exceeds 6.80, the low-temperature upturn in $\rho_{ab}(T)$ disappears and a metallic behavior is observed in the $y=6.83$ sample in 15 T. It should be noted that the localization behavior is observed here in samples with quite small resistivity. For example, the minimum of $\rho_{ab}(T)$ for the $y=6.80$, $z=2.7\%$ sample is about 180 $\mu\Omega$ cm. This value corresponds to $k_F l \simeq 8.5$, if we use the formula $l = hc_0/\rho k_F e^2$ for 2D electrons[16] (c_0 is the c-axis unit length); therefore, the localization behavior is taking place in quite a clean system where localization is not normally observed.[17] In this sense, the localization behavior observed here is quite unusual.

The inset to Fig. 4 shows a comparison of $\rho_{ab}(T)$ of two samples, $y=6.70$, $z=2.3\%$ (sample M1) and $y=6.80$, $z=2.7\%$ (sample I1), to demonstrate that the onset of the localization behavior is not correlated with residual resistivity ρ_{res}. Sample M1 has apparently higher ρ_{res} than that of sample I1, indicating there is no critical ρ_{res} for the localization behavior to take place. This observation is in contrast to the zero-field S-I transition observed in YBCO and LSCO,[6] which occurs whenever the resistivity exceeds the critical value, as mentioned earlier. It should be noted that all the samples which show the localization behavior in Figs. 1 and 3 have smaller resistivity than the critical value 400 $\mu\Omega$ cm (Ref. 6) and thus are on the superconductor side of the S-I transition; in fact, all those samples are superconducting and do not show any upturn in ρ_{ab} in zero field, which is in accord with the zero-field S-I transition picture. The unusual localization behavior shows up once the superconductivity is suppressed, where charge is no longer carried by Cooper pairs. The result shown in the inset to Fig. 4 indicates that the onset of the unusual localization behavior is determined primarily by the Zn concentration but not by ρ_{res}.

The evolution of $\rho_{ab}(T)$ with changing oxygen content for $z=2.3\%$ is shown in the main panel of Fig. 4. For this Zn concentration, the charge transport has much weaker tendency towards localization; even the $y=6.60$ sample shows only a slight upturn in 15 T. When the oxygen content is further reduced to 6.50, ρ_{res} exceeds the critical value for the S-I transition and $\rho_{ab}(T)$ shows an insulating behavior already in zero field. Comparison between the $z=2.7\%$ samples (Fig. 3) and the $z=2.3\%$ samples (Fig. 4) lead to the conclusion that the tendency toward carrier localization in the high-field normal state becomes much stronger when the Zn concentration is increased from 2.3% to 2.7%. Note that there is no drastic difference between the $z=2.7\%$ samples and the $z=2.3\%$ samples if we only look at the zero-field data.

Now let us discuss the possible origin of the unusual localization behavior. What we observed in Zn-doped YBCO has strong similarities to the unusual localization behavior found in underdoped LSCO:[8,16,18] In both systems, the localization behavior takes place in a region of ‘‘clean’’ charge transport where $k_F l$ is larger than 5; the temperature dependence of the resistivity is consistent with log(1/T) and the size of the increase in resistivity is larger than what is expected from weak localization; the localization behavior shows up at low temperatures only when the superconductivity is suppressed with high magnetic fields. Therefore, it is naturally expected that the localization behaviors observed in the two systems have a common origin. When we look for some common features in the underlying electronic/magnetic structures of the two systems, we hit on the fact that, as mentioned earlier, both systems have some kind of ‘‘defects’’ to the AF correlation in the CuO$_2$ planes; the LSCO system is likely to have dynamical charge stripes which separate AF domains, while the Zn impurity in the YBCO system acts like a pointlike defect in the AF correlation. Note that both the dynamical charge stripes and the Zn-substitution effect are most strongly seen at excitations with wave vectors near $\mathbf{q}=(\pi,\pi)$.

The above considerations lead to the conclusion that the local destruction of the AF correlation in the CuO$_2$ planes might be the origin of the unusual insulating behavior. This conclusion in turn suggests that the dynamical AF correlation in the CuO_2 planes governs the peculiar normal-state charge transport of the cuprates. If so, the charge in cuprates may be carried by a quasiparticle which is closely related to the AF correlation, and a local disturbance of the AF correlation, in the presence of a high magnetic field, may strongly scatter the quasiparticle and bring about the unusual charge localization. The fact that the localization behavior is much stronger in the LSCO system [where there is more than a factor of 2 increase in resistivity at low temperatures in 60 T (Ref. 8)] may mean that the scattering from 1D defects is much stronger than that from pointlike defects.

In summary, we measured the in-plane resistivity of Zn-doped YBCO crystals down to low temperatures by suppressing superconductivity with high magnetic fields. It is found that the Zn substitution induces unusual carrier localization at low temperatures in a ''clean'' system with $k_F l$ as large as 8.5, and the tendency toward carrier localization becomes much stronger when the Zn concentration is increased from 2.3% to 2.7%. Strong similarities are found between the localization behavior observed here and the $\log(1/T)$ behavior observed in underdoped LSCO. Examination of the common characteristics in LSCO and Zn-doped YBCO leads to the conclusion that the local destruction of the AF correlation either by 1D defects (dynamical charge stripes in LSCO) or by pointlike defects (Zn in YBCO) might be the origin of the unusual charge localization.

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