Universal Superconductor-Insulator Transition and T_c Depression in Zn-Substituted High-T_c Cuprates in the Underdoped Regime

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The experimental results are presented on the in-plane resistivity for Zn-substituted single crystals of $YBa_2Cu_3O_{7-y}$ and $La_{2-x}Sr_xCuO_4$ with various hole densities. The primary effect of Zn is to produce a large residual resistivity (ρ_0) as a potential scatterer in the unitarity limit. In the underdoped regime, due also to low carrier density in the CuO_2 plane, only a few percent Zn is sufficient for ρ_0 to reach the critical value near the universal two-dimensional resistance $h/4e^2$ and to induce a superconductor-insulator transition. By contrast, the universal behavior is not seen in the highly doped regime, suggestive of a radical change in the electronic state.

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Zn substitution in copper oxide superconductors has a strong influence on the critical temperature T_c and thus offers an opportunity to characterize the high-T_c superconducting state. Many experimental efforts have been conducted to explain the T_c depression in relation with a "normal" impurity in the d-wave pairing state [1,2]. Since a small concentration of Zn impurities introduced into the CuO_2 plane produces a significant change in the low-energy spin fluctuations as evidenced by the NMR [1,3] and neutron scattering [4,5] experiments, it is argued that the Zn substitution provides a "smoking gun" for the spin-fluctuation-mediated pairing mechanism in high-T_c cuprates [6].

Zn in the CuO_2 plane is itself a nonmagnetic impurity with a closed d shell and is expected to be a strong potential scatterer for charge carriers. Recently, Chien, Wang, and Ong [7] have investigated the Zn-substitution effect on the normal-state charge transport in $YBa_2Cu_3O_7$ and estimated a large scattering cross section of the Zn impurity. As the scattering cross section of Zn impurity is related with the parameters characterizing the electronic state of the high-T_c cuprates, Zn is expected to be an effective probe for detailed study of the evolution of the electronic state with doping.

We present the results of resistivity measurements on the single crystals of Zn-substituted $YBa_2Cu_3O_{7-y}$ (Y123) and $La_{2-x}Sr_xCuO_4$ (La214) over a wide range of Zn content and doped hole density. We demonstrate how the Zn-induced residual resistivity varies with hole density and show that the superconductor-insulator transition in two dimensions is readily induced due to very large residual resistivity in the underdoped regime. It is highlighted that Zn probes a remarkable difference in the electronic state between underdoped and highly doped superconducting regime.

Single crystals of $La_{2-x}Sr_xCu_{1-z}Zn_zO_4$ were grown using the traveling-solvent-floating-zone (TSFZ) method [8]. In the present work the crystals with $x = 0.10$ ($T_c = 27$ K), 0.15 ($T_c = 37$ K), 0.20 ($T_c = 32$ K), and $x = 0.30$ ($T_c = 0$ K) were prepared with Zn content ranging up to 0.04. Zn-substituted single crystals of $YBa_2(Cu_{1-z}Zn_z)_3O_{7-y}$ were grown by the CuO-BaO self-flux method up to $z = 0.04$. The oxygen content was adjusted to have a value between 6.63 and 6.93 by annealing the crystals in the same way as described by Ito, Takenaka, and Uchida [9]. For the oxygen-reduced compound ($y \geq 0.32$), $z = 0.04$, the upper limit of homogeneous Zn substitution in single crystals is enough to destroy superconductivity, but it is insufficient for the oxygenated compound ($y < 0.32$).

The temperature dependence of the resistivity ρ_{ab} is shown in Fig. 1 for $y = 0.37$ and 0.07 of Y123 with Zn content ranging up to $z = 0.04$. Major results on the underdoped compound ($y = 0.37$) are as follows: (i) T_c is rapidly reduced and the compound becomes insulating after the superconductivity disappears at $z_c \simeq 0.03$. (ii) A large T-independent component ρ_0 (residual resistivity) adds to the T-dependent resistivity. Notably a superconductor-insulator (SI) transition occurs at $\rho_0 \sim 400\ \mu\Omega$ cm. This value corresponds to the two-dimensional (2D) resistance $\rho_0^{2D} \sim 6.8\ \mathrm{k}\Omega/\square$ per CuO_2 plane (not per bilayer) and is near the universal value $h/4e^2 \simeq 6.5\ \mathrm{k}\Omega/\square$ [10,11].

Although it is difficult to introduce sufficient Zn into Y123 single crystals to destroy superconductivity for oxygen content higher than 6.7, we can reasonably conclude from the extrapolation of the T_c-z and ρ_0-z characteristics to higher z (see the result in Fig. 3 shown later) that the SI transition will take place at the same critical resistivity for the oxygen content lower than 6.8.

The results for La214 indicate that $x = 0.10$ and 0.15 are in the same regime as shown in the middle panel of Fig. 1 for $x = 0.15$ [12]. The SI transition occurs in both compositions near the universal 2D resistance which corresponds to the resistivity $\sim 370\ \mu\Omega$ cm in the La214. This value is nearly the same as that in Y123, suggesting that each CuO_2 plane in the bilayer of Y123 is independent

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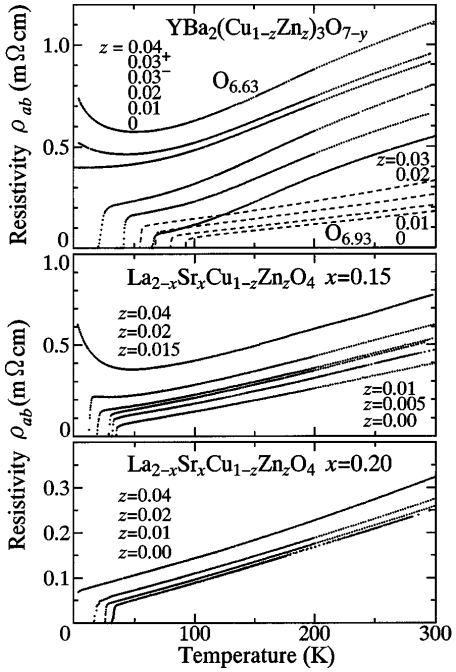

FIG. 1. The temperature dependence of the in-plane resistivity of Zn-substituted $YBa_2Cu_3O_{7-y}$ with $y = 0.37$ (solid curves) and $y = 0.07$ (dashed curves) shown in the upper panel, and the results for $La_{2-x}Sr_xCuO_4$ with $x = 0.15$ and 0.20 are shown in the middle and lower panels, respectively. $z = 0.03^+$ and 0.03^- for $y = 0.37$ of Y123 are different crystals taken from the same batch.

because of nearly equal average spacing between CuO_2 planes.

The highly doped superconducting compounds show a contrasting behavior. As demonstrated for $x = 0.20$ of La214, the material remains metallic even after the superconductivity disappears at $z = 0.04$ where the residual resistivity is by a factor of 4 smaller than the critical value observed for $x = 0.10$ and 0.15. The 90 K Y123 ($y = 0.07$) would behave in the same manner, when more Zn (perhaps $\sim 8\%$ may be necessary for the destruction of superconductivity) could be introduced, as the linear extrapolation of the ρ_0-z curve (see Fig. 4) to $z = 0.08$ reaches only $1/3$ of the critical value.

Figure 2 illustrates how the in-plane resistivity varies with changing doped hole density for fixed z. One recognizes that the magnitude of the residual resistivity is fairly large in the underdoped regime ($x = 0.10$ and 0.15 for La214 and oxygen content 6.68 and 6.73 for Y123) and is rapidly reduced for higher hole density. Using the results in Figs. 1 and 2, the values of T_c normalized to the value T_{c0} for the Zn-free compound are plotted in Fig. 3 against ρ_0^{2D} for the two systems with various z and $x(y)$. Figure 3 displays another aspect of the universal T_c depression in the underdoped cuprates. Irrespective of the doped hole density, the data for the underdoped cuprates merge into a single pair-breaking curve which points toward the universal 2D resistance $h/4e^2$ as $T_c \rightarrow 0$.

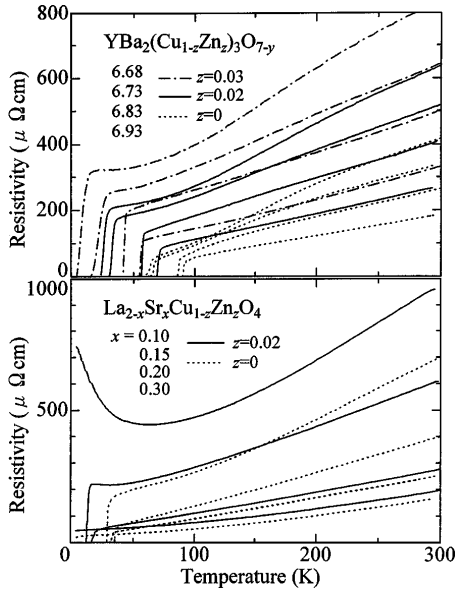

FIG. 2. The temperature dependence of the in-plane resistivity of $YBa_2(Cu_{1-z}Zn_z)_3O_{7-y}$ with $z = 0.02$ and 0.03 for various oxygen contents between 6.68 and 6.93 (from top to bottom). The data for $z = 0$ crystals are shown by the dashed curves. The lower panel shows the result for $La_{2-x}Sr_xCu_{1-z}Zn_zO_4$ with $z = 0.02$ and x ranging from $x = 0.10$ to $x = 0.30$ (from top to bottom).

By contrast, the highly doped cuprates ($x = 0.20$ for La214 and oxygen contents 6.93, 6.88, and 6.83 for Y123) show quite distinct T_c-ρ_0^{2D} curves which are strongly dependent on the doped hole density. As the hole density increases, the T_c degradation speeds up as a function of ρ_0^{2D} and the T_c-ρ_0^{2D} curve appears to end up at ρ_0^{2D} considerably lower than $h/4e^2$.

A possible explanation for nonuniversal behavior would be that the highly doped material is a three-dimensional (3D) superconductor. Certainly, it is a general trend

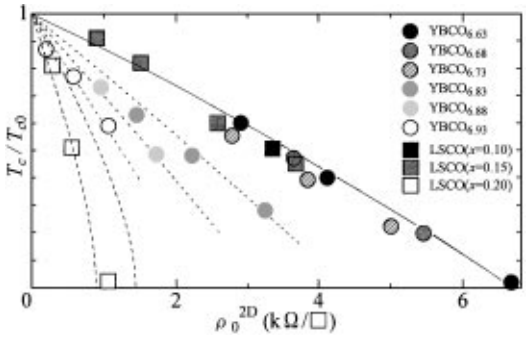

FIG. 3. Normalized critical temperature T_c/T_{c0} plotted as a function of the in-plane 2D residual resistance (per CuO_2 plane). The solid curve is a line onto which all the data in the underdoped regime merge. The data for the same Sr composition or oxygen content in the highly doped regime are connected by a dotted line. Theoretical estimates by Radtke *et al.* [13] for a d-wave superconductor with nonmagnetic impurities are in the region between the dashed curves.

that the magnitude of the anisotropic resistivity (ρ_c/ρ_{ab}) decreases with increase of dopant concentration [8,14]. However, the recent c-axis optical and transport study has demonstrated that a truly 3D state is realized in the overdoped nonsuperconducting region [15].

An alternative explanation, though highly speculative, is that the nonuniversal behavior in the highly doped regime may result from some inherent inhomogeneity such as phase separation or strong fluctuations of the amplitude of the order parameter, that is, an appreciable density of normal carriers may coexist in the highly doped superconducting compound. The density of the normal carriers increases as the nonsuperconducting overdoped regime is approached. Suppose that a normal fluid (e.g., overdoped domains) in which the electrons are not easily localized due to Zn impurities forms a parallel circuit with superfluid (e.g., underdoped domains) which readily lose superconductivity and become insulating for Zn substitution at $z = z_c$. Then, the superconductivity would disappear at resistivity appreciably smaller than the critical value (~400 $\mu\Omega$ cm) observed in the underdoped regime and the material would keep metallicity due to much more conductive normal fluid. In this model the apparent critical resistivity or the slope of the T_c-ρ_0^{2D} curve would depend on how much the normal metallic phase is mixed in. The result in Fig. 3 for highly doped compounds shows a trend just expected from this model.

There is no direct evidence for such phase separation in the highly doped cuprates, but some indications are seen in the recent optical and muon spin rotation (μSR) experiments. The optical conductivity spectra of 90 K Y123 [16,17] and highly doped La214 ($x \gtrsim 0.18$) [18] show a strongly gapless feature with a Drude-like conductivity remaining in the spectrum even at temperatures well below T_c. This "residual" conductivity reduces the missing spectral weight due to the opening of a superconducting gap and is linked to the saturation and a subsequent increase in the magnetic penetration depth observed by μSR on the highly doped cuprates [19,20].

Coming back to the Zn-substitution effect in the normal state, the residual resistivity ρ_0 (or the corresponding ρ_0^{2D}) determined from the nearly parallel shift of the $\rho(T)$ curves and/or from the zero-temperature intercept of the T-linear part of ρ is plotted as a function of Zn content in Fig. 4 for two representative compositions in each system. Following the analysis made by Chien, Wang, and Ong [7], the resistivity arising from s-wave impurity scattering in 2D is

$$\rho_0 = 4(\hbar/e^2)\,(n_i/n)\sin^2\delta_0\,, \qquad (1)$$

where n_i is the impurity concentration and δ_0 is the s-wave phase shift. The straight line in each column is the value in the unitarity limit ($\delta_0 = \pi/2$) with the carrier density $n = x$, the doped hole density per planar Cu. For Y123 we assume that $x \sim 0.23$ for $y = 0.07$ and $x \sim 0.14$ for $y = 0.37$ [21] and that Zn atoms are substituted only on the plane Cu sites such that $n_i = \frac{3}{2}z$ [1,3].

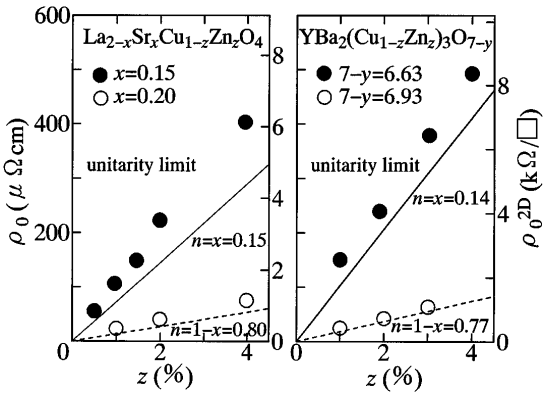

FIG. 4. Variation of the residual resistivity ρ_0 (and ρ_0^{2D} right-hand scale) with Zn content in $YBa_2Cu_3O_{7-y}$ with $y = 0.37$ and 0.07, and in $La_{2-x}Sr_xCuO_4$ with $x = 0.15$ and 0.20. The solid (dashed) line indicates the unitarity limit with the carrier density $n = x$ ($n = 1 - x$).

It turns out that the experimental values of ρ_0 for the underdoped materials are close to (or even larger than) the unitarity limit with $n = x$. The carrier density in the underdoped regime should thus be identified with the density of doped hole number. Here we should check the assumption that Zn is a potential scatterer. Zn is itself a nonmagnetic impurity, but it induces a localized magnetic moment in the CuO_2 plane. The magnitude of the induced moment is as large as $\sim 0.8\mu_B$ (μ_B is the Bohr magneton) for the underdoped cuprates, and is reduced in the highly doped regime, $\sim 0.2\mu_B$ in 90 K Y123 [22]. Borkowski and Hirschfeld [23] have shown that a scattering rate Γ_S from the induced magnetic moments in the 90 K Y123 is much smaller than the potential scattering rate Γ_N. This should be the case also with the 60 K Y123. Since the carrier density or the density of states $N(0)$ should be smaller in 60 K Y123, and $\Gamma_N \sim N(0)^{-1}$ whereas $\Gamma_S \sim N(0)$, Γ_N still overweighs Γ_S with the decreased $N(0)$ compensating the increased magnetic moment.

The residual resistivity for the highly doped materials is remarkably smaller than the unitarity limit ($n = x$) but is near the unitarity limit with $n = 1 - x$. Then, the decreased ρ_0 is ascribable either to a decrease in δ_0 or to an increase in n toward $1 - x$. The fact that the value of ρ_0 for overdoped ($x = 0.30$) La214 is near the value in the unitarity limit with $n = 1 - x$ which is expected in a Fermi liquid state gives evidence for the increase in n while δ_0 remains to be $\pi/2$. Using the data in Fig. 2, we plot in Fig. 5 the variation of ρ_0^{2D} for 1% Zn with doped hole density (x) per planar Cu atom. Even if we admit ambiguity in estimating x in Y123 [21], it is evident that a rapid increase in carrier density or a rapid crossover from $n = x$ to $n = 1 - x$ is taking place when x exceeds ~0.17. This result is indicative of a radical change in the electronic state of the CuO_2 plane with increase of doping.

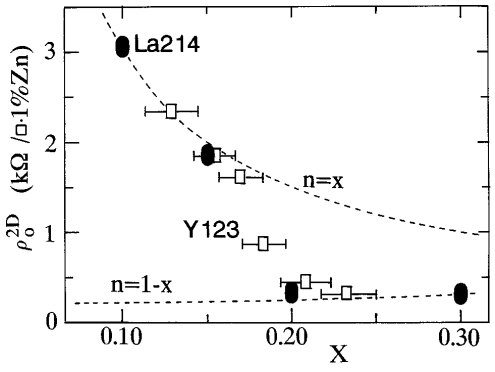

FIG. 5. Variation of the 2D residual resistance for 1% Zn with doped hole density in Y123 and La214. Two dashed curves show the unitarity limit with $n = x$ and $n = 1 - x$ calculated using Eq. (1). Wide error bars for the data of Y123 indicate uncertainty in the estimate of the doped hole density x from the oxygen content $7 - y$.

The crossover region just corresponds to what we call a highly doped region.

We have demonstrated that Zn probes a critical change in the electronic state with increasing doped hole density. In the underdoped regime, a large residual resistivity produced by Zn triggers a universal SI transition in 2D, while nonuniversal behaviors are seen in the highly doped region.

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