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Normal and superconductive properties of Zn-substituted single-crystal $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$

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Zn-impurity-induced anisotropy reduction of superconductivity is obtained from a resistive transition analysis of single-crystal $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$. The out-of-plane resistivity (ρ_c) increases with Zn doping, while the carrier concentration remains almost unchanged. The results are not consistent with the simple Lawrence-Doniach model. Instead, an anisotropic three-dimensional metal model seems to be promising. A Zeeman-contribution-subtracted magnetoresistance analysis indicates that a Zn impurity does not cause any magnetic pair breaking.

The effect of impurities on high-T_c superconductivity is considerably different from that on conventional superconductivity.[1] Especially, Zn substitution for Cu caused a lot of discussion about the T_c reduction mechanism.[2,3] In this paper, we report on the normal and superconducting transport properties on Zn-doped $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$ single crystals. We focus our attention on the dimensionality of the one-electron state in $YBa_2Cu_3O_{7-\delta}$ (YBCO), as well as the magnetic vs nonmagnetic issue of Zn impurities.[4,5]

How impurities cause mixing of one-electron states depends on their dimensionality. For instance, if the one-electron state is strictly two dimensional, confined in the CuO_2 plane, the impurity causes mixing of only in-plane states. Consequently, ρ_c remains essentially unchanged upon impurity doping. A similar effect is also expected in the superconducting state.

To obtain information about the anisotropy in the superconducting state, we estimate the in-plane and out-of-plane coherence lengths from a resistive transition analysis of single-crystal $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$. For this analysis we use a superconducting-fluctuation-renormalized theory including a ψ^4 term,[6] which has successfully been applied to these Zn-impurity-containing systems.[8] We discuss the dimensionality of the one-electron state in YBCO based on the obtained coherence lengths and change of ρ_c with x. Then, using the parameters obtained, the analysis is extended to magnetoresistance in the Gaussian fluctuation region to determine the type of scattering induced by Zn doping.

We have grown a series of Zn-substituted single-crystal $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$ with x ranging from 0 to 0.03. Well-decarbonated ceramic powder with composition ratio [Y]:[Ba]:[Cu+Zn] = 1:4:9–11 is heated to 1050 °C, then cooled below 900 °C at a rate of 3 °C/h in air. Samples are annealed in flowing oxygen at 500 °C for 100 h, then cooled to room temperature at a rate of 3 °C/min. The bulk superconductivity was checked with a dc superconducting quantum interference device. Crystal quality was checked with a polarized light microscope and an x-ray precession camera. Even for a large doping of x = 0.03, the c axes uniformly align in one direction. (110), (1$\bar{1}$0) twinning is usually observed and the measured quantities in the a-b plane are averaged over the a and b direction. After measurement of transport properties, the crystals were analyzed by electron-probe-microanalysis technique (Table I). The Zn distribution in the a-b plane varies 0.001 Zn per Cu atom. The contamination by the crucible material (Pt) is 0.002 – 0.006 Pt per Cu atom.

Figure 1 shows the in-plane resistivity (ρ_{ab}) for various levels of Zn doping. The additional resistivity is roughly proportional to x, which shows that Zn acts as a simple impurity.[9] The small change in the average $d\rho/dT$ with x indicates that Zn only slightly affects the carrier concentration and the inelastic scattering processes.

We cannot obtain a reliable value of coherence lengths from conventional H_{c_2} measurements, due to the broadening of the resistive transition under a magnetic field. Recently, Ikeda, Ohmi, and Tsuneto[6] proposed a phenomenological theory which succeeded in describing the resistive transition, and offering a reliable estimation for the coherence lengths. The theory is valid over a wide range of magnetic field and temperature, the Ginzburg-Landau-fluctuation regime, as long as flux motion is not important. Here, we use their theory to obtain the coherence lengths. Figures 2(a)–2(c) show the resistive tran-

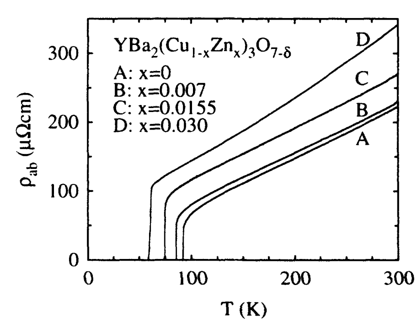

FIG. 1. Temperature dependence of the in-plane resistivity of single-crystal $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$. The samples A, B, C, and D correspond to those in Table I.

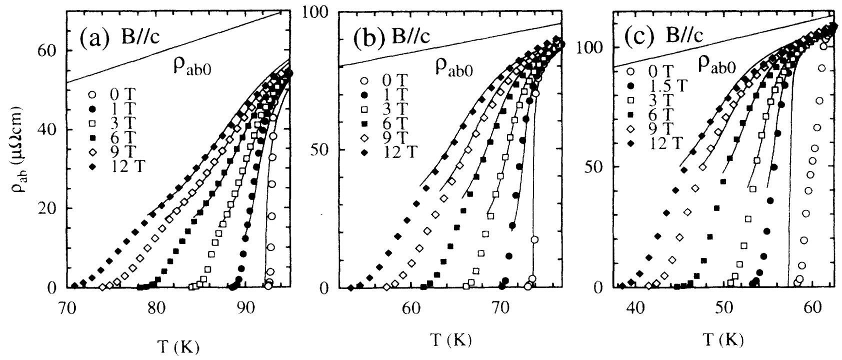

FIG. 2. Temperature dependence of the in-plane resistivity of single-crystal $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$ samples (a) A, (b) C, and (c) D. The magnetic-field direction is perpendicular to the CuO_2 planes. The solid curves are the theoretical fits. The fitting parameters are listed in Table I. In (c), the deviation between the calculated curve and the measured data for zero magnetic field may be due to a filamentary higher-T_c region in the sample. The in-plane resistivity without superconducting fluctuation ρ_{ab_0} is assumed to be a linear extrapolation of the resistivity curve above T_c without magnetic field.

sition of single-crystal $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$ with applied field in the c direction and current flow in the a-b plane. In the analysis, we assume ρ_{ab_0}, the in-plane resistivity without superconductive fluctuation, to be a linear extrapolation of the resistivity curve at higher temperatures. The fluctuational contributions from Landau levels up to the 1000th level are taken into account. Five quantities are obtained from the analysis: the mean-field critical temperature T_{c0}, the in-plane and c-axis coherence lengths ξ_{ab0} and ξ_{c0}, the specific-heat jump ΔC, and the C factor. The C factor is a scaling factor which adjusts resistivity of a real sample to that of an ideal sample.[10] In a real sample, current still does not flow uniformly on a submacroscopic scale, because of possible imperfections. We fixed the distance between the conduction layers to c, the unit-cell dimension, of Zn-doped YBCO.[7] In the analysis, we first optimize ξ_c and the C factor to roughly reproduce the resistivity data without magnetic field near the critical temperature. After that, we optimize the other parameters to reproduce the resistivity data under magnetic field above the "knee" temperature. Then we repeat this process until iteratively a satisfactory fit is obtained. The obtained parameters are listed in Table I. Slightly larger ΔC values are obtained in our analysis compared to reported values for Zn-doped YBCO ceramic.[11,12] If we use the reported values, the fluctuational conductivity decreases and the fitting considerably deviates for samples C and D. For all Zn-doped crystals the theory fits very well. This indicates that there is a certain temperature range below T_{c_0}, where Zn impurities do not cause any flux pinning and the superconducting fluctuation is the relevant factor for generating the resistivity.

TABLE I. Parameters obtained from our analysis of resistive transition data of $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$ crystals under several magnetic fields.

Sample	A	B	C	D
x (%)	0.0	0.7 ± 0.1	1.55 ± 0.1	3.0 ± 0.1
T_{c_0} (K)	92.3	84.0	73.8	57.5
ξ_{ab_0} (Å)	9.0	10.0	13.0	14.5
ξ_{c_0} (Å)	1.0	1.2	1.8	2.2
ΔC (mJ K^{-1} cm^{-3})	45	41	36	20.5
C factor	0.58	1.3	1.5	1.8

In Fig. 3, ξ_{ab0} and ξ_{c0} are plotted against Zn content x. As x increases, both ξ_{ab0} and ξ_{c0} increase but ξ_{c0} increases faster than ξ_{ab0}. A considerable reduction of anisotropy of ξ is observed. This reduction of anisotropy can also be concluded from the magnetoresistance measurements in the Gaussian fluctuation region.[13] In general, as T_c is reduced an isotropical increase of the coherence length is expected by the relation $\xi \propto \hbar v_F/\Delta$. However, a change in anisotropy cannot be explained by a T_c reduction. According to the Lawrence-Doniach (LD) model,[14] if the in-plane effective mass (m^*_{ab}) and the Fermi energy remain unchanged on Zn doping,[15] an increase in ξ_c means an increase of the tunneling between conduction layers, and it should consequently reduce ρ_c. In Fig. 4 the c-axis normal-state resistivity ρ_c for $x = 0$, 0.01, and 0.03 obtained by Montgomery measurements[16,17] are shown. No ρ_c reduction by Zn doping is observed. On the other hand, in fully oxidized crystals, both ρ_{ab} and ρ_c are metallic, i.e., $d\rho/dT > 0$,

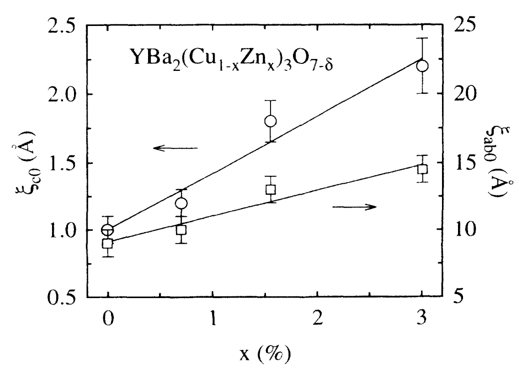

FIG. 3. Variation of ξ_{ab0} (squares) and ξ_{c0} (circles) with Zn content x in single-crystal $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$. The anisotropy ratio ξ_{ab0}/ξ_{c0} reduces 30% when x increases from 0 to 0.03.

and ρ_c is still just below the extended Mott-Ioffe-Regel limit,[18] so an anisotropic three-dimensional (3D) metal model can be applied. If we assume that Zn impurities scatter carriers isotropically, the in-plane and c-axis Fermi velocity ratio can be obtained from the square root of the resistivity increase ratio. $v_{Fc}/v_{Fab} \approx 0.16$. In the clean limit, $\xi_c/\xi_{ab} = (v_{Fc}/v_{Fab})(\Delta_{ab}/\Delta_c)$. Using the obtained coherence lengths and Fermi velocity ratio, $\Delta_{ab} \approx 0.7\Delta_c$ for $x = 0$ and $\Delta_{ab} \approx \Delta_c$ for $x = 0.03$. Note that, a clean limit assumption for the c direction and the $\Delta_{ab} < \Delta_c$ condition for $x = 0$ are controversial issues. However, an anisotropic 3D metal model qualitatively explains the anisotropy reduction in superconductivity by gap averaging, and the ρ_c increase by elastic scattering due to Zn impurities.

Next, we consider the excess pair-breaking process caused by Zn impurities. If the Zn-induced magnetic moment is the main origin of the strong reduction of the transition temperature, it also destroys fluctuation Cooper pairs at temperatures above T_c. This can be detected in the magnetoresistance, through the pair dephasing rate τ_ϕ^{-1} in the orbital contribution of the Maki-Thompson (MTO) process. To see whether τ_ϕ^{-1} contains an excess pair-breaking rate contribution in the Zn-substituted samples, we analyze the Zeeman-contribution-subtracted conductivity data $(-\Delta\sigma_{\perp ab} + \Delta\sigma_{\parallel ab})$ using the following two models.

(a) The elastic scattering model: $\tau_\phi^{-1} = \tau_{\phi_0}^{-1}$. Zn doping mainly causes elastic scattering, and it only reduces the MTO contribution through the reduction of the carrier mean free path, and it does not cause excess depairing.

(b) The inelastic scattering model: $\tau_\phi^{-1} = \tau_{\phi_0}^{-1} + \tau_{\mathrm{Zn}}^{-1}$. Zn doping causes inelastic (magnetic) scattering. It reduces the carrier mean free path and causes excess depairing.

In both cases, the same transport scattering rate $\tau_{\mathrm{tr}}^{-1} = \tau_{\mathrm{tr}_0}^{-1} + \tau_{\mathrm{Zn}}^{-1}$ is used. We assume $\tau_{\phi_0}^{-1} = \tau_{\mathrm{tr}_0}^{-1}$ as before.[20] In sample D, the mean free path $\ell (= v_F\tau_{\mathrm{tr}})$ around T_c can be estimated about 50 Å. It is larger than the obtained in-plane coherence length ξ_{ab0}, which is about 14.5 Å, so a clean-limit analysis is still valid for sample D. In a clean-limit analysis, the fluctuation contribution of the MTO process in the magnetoconductivity $(-\Delta\sigma_{\mathrm{MTO}})$ decreases if $\ell\tau_\phi$ is reduced. So, $-\Delta\sigma_{\mathrm{MTO}}$ is thought to be smaller in the inelastic scattering model (b) than in the elastic model (a). In the fitting, we use the same parameters as obtained from the resistive transition analysis, that is, $\xi_{ab0} = 14.5$ Å, $\xi_{c0} = 2.2$ Å, and $C = 1.8$. We assume the Fermi velocity and the inelastic scattering rate to be the same as in the undoped samples,[19,20] that is, $v_F \sim 2 \times 10^7$ cm/s, $\hbar\tau_{\mathrm{tr}_0}^{-1} = 1.3k_BT$. Figure 5 shows the result of the analysis. It is evident that the elastic scattering model gives a better fit to the data than the inelastic scattering model. This analysis indicates that the rapid T_c supression by Zn doping is not due to magnetic pair breaking.[5]

There is an assumption, $\Delta(r) \simeq$ const, for Anderson's theory[21] of impurity effects on superconductivity. In high-T_c superconductors, it is questionable whether this condition holds, since recent experiments suggest that the superconducting gap is anisotropic and consistent with d-wave superconductivity.[22,23] In the case of gapless superconductivity, even a nonmagnetic impurity is destructive to the superconductivity.

In conclusion, from an analysis of the resistive transition and magnetoresistance of Zn-substituted single-crystal $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$, a reduction in anisotropy in the superconducting state (ξ_{ab}/ξ_c) is observed. On the other hand, an anisotropy reduction in the normal state is not observed from resistivity measurements. The simple LD model cannot describe this situation. An anisotropic 3D metal model qualitatively explains the results. This implies that high-T_c models which assume strict two dimensionality cannot be applied to the YBCO system; therefore, these models are not universally applied to all high-T_c superconductors.

A Zeeman-contribution-subtracted magnetoresistance analysis of Zn-doped single-crystal YBCO shows that Zn impurities primarily act as elastic scattering centers. Zn doping only reduces the carrier mean free path and does not cause excess depairing. These analyses show that the

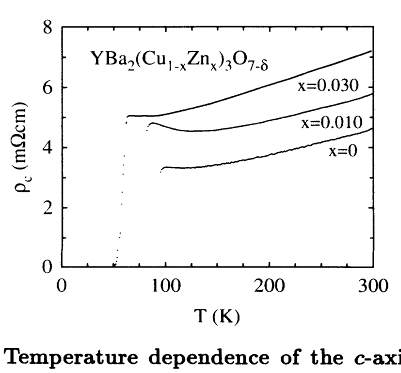

FIG. 4. Temperature dependence of the c-axis resistivity for $x = 0$, 0.01, and 0.03 $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$ crystals. The measurements shown here are taken on a different set of samples than those listed in Table I.

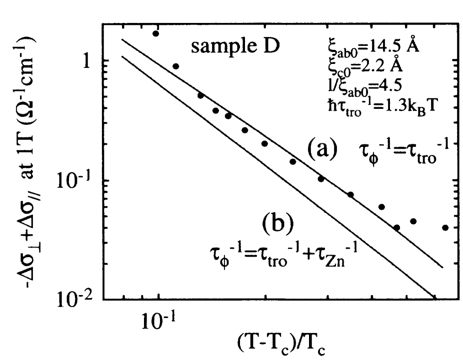

FIG. 5. Zeeman-contribution-subtracted magnetoconductivity data $(-\Delta\sigma_{\perp ab} + \Delta\sigma_{\parallel ab})$ for sample D (dots) against reduced temperature $(T - T_c)/T_c$. $\perp$ and $\parallel$ stand for, respectively, the applied magnetic field perpendicular and parallel to the a-b plane. Two theoretical fits are shown (solid lines) that assume the scattering caused by Zn doping to be (a) elastic and (b) inelastic.

T_c suppression in this system cannot be explained by a depairing effect caused by induced magnetic moments.

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