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STUDY OF ZN DOPING IN UNDER DOPED AND OVER DOPED REGIMES CUPRATE PEROVSKITES

KHUSHVANTS SINGH[1] & SUMIT KUMAR GUPTA[2]

[1]Department of Physics, BSA College, Mathura, Uttar Pradesh, India

[2]Department of Physics, Maharishi Arvin Institute of Engineering & Technology, Jaipur, Rajasthan, India

ABSTRACT

The experimental results are presented Zn substitution in copper oxide superconductors has a strong influence on the critical temperature T_c and offers an opportunity to characterize the high-T, superconducting state. Most experimental and theoretical studies have been conducted to determine or explain the T_c depression as a function of Zn content. In this paper we show the results of resistivity measurements on the single crystals of Zn-substituted $YB_{a2}Cu_3O_{7-y}$ (Y123) and $La_{2-x}Sr_xCuO_4$ (La214) with different levels of hole doping. How the Zn-induced residual resistivity varies with hole density and establish the depairing relation between T_c and the two-dimensional resistance which would serve as a constraint for various theoretical models. It is highlighted that Zn probe a remarkable difference in the electronic state between under doped and over doped superconducting regime.

KEYWORDS: Transport Properties, Effect of Crustal Defect, Doping and Substitution

INTRODUCTION

Zn substitution in copper oxide superconductors has a strong influence on the critical temperature T_c and offers an opportunity to characterize the high-T, superconducting state. Since a small concentration of Zn impurities introduced into the CuO_2 plane produces a significant change in the low-energy spin fluctuation as evidenced by the NMR (1,2) and neutron scattering (3,4) experiments.

Compared with the magnetic studies much *less efforts* have been devoted to the study of Zn-substitution effect on the charge dynamics in the doped CuO_2 planes. Most experimental and theoretical studies have been conducted to determine of explain the T_c depression as a function of Zn content. Zn in the CuO_2 plane is itself a non-magnetic impurity with a closed d shell and is expected to be strong potential scatterer for charge carriers.

In this paper we shoe the results of resistivity measurements on the single crystals of Zn of substituted $YB_{a2}Cu_3O_7$-y (Y123) and $La_{2-x}Sr_xCuO_4$ (La214) with different levels of hole doping. We demonstrate how the Zn-induced residual resistivity varies with hole density and establish the depairing relation between T_c and the two-dimensional resistance which would serve as a constraint for various theoretical models. It is highlighted that Zn probe a remarkable difference in the electronic state between under doped and over doped superconducting regime.

CRYSTAL GROWTH AND CHARACTERIZATION

Single crystals of Zn doped Y123 were grown by CuO-BaO self-flux method using an Y_2O_3 crucible to avoid contamination from the crucible. This is crucial for the present experiments. As we are concerned with the Zn doping level

of 1%. The oxygen concentration was controlled by annealing the crystals at 600 ^{0}C for 12 hours in a sealed quartz tube together with Y123 powders which had a prescribed oxygen concentration. Then we cooled them slowly to promote oxygen ordering and to ascertain the same oxygen concentration as that of the powders. The crystals grown and annealed by these processes have high quality and homogeneity which are evidence by a sharp superconducting transition as well as by the lowest normal-state resistivity's among so far reported values, and thus allow for a systematic and quantitative study. We cheeked the sample dependence of ρ_{ab} using several crystals grown and annealed at the same time. The variation of T_c among different crystals was within 1 k and that of ρ_{ab} was within 5%. A rather sharp superconducting transition even for Zn-doped crystals and systematic increase in the normal-state resistivity's with z as shown below provide further evidence for homogeneous distribution of Zn.

In Plane and Out-of-Plane Resistivity

The temperature dependence of the resistivity$\rho_{ab}$is shown in figure. for y=0.37 and 0.07 of Y123 with Zn content ranging up to z = 0.04. The compound with y = 0.037 shows Zn substitution effects typical of underdoped cuprates. The effects of Zn-substitution are two-fold: (i) T_c rapidly reduced and the superconductivity disappears at Zc$\underline{\sim}$ 0.03. (ii) A T-independent component ρ_o (residual resistivity) adds to the T-dependent resistivity. No-tably the y = 0.37 compound becomes insulating when the superconductivity is destroyed and a superconductor-insulator (SI) transition occurs at ρ_0o~ 400μΩ cm (we have got a crystal with Zc$\underline{\sim}$ 0.03 which is incidentally very close to the critical point). This value corresponds to the two-dimensional (2D) resistance ~ 6.8 kΩper cuO_2 plane (not per bilayer) and is close to the universal value $\lambda/4e^2\underline{\sim}$6.5 kΩwhich is predicted to separated superconducting and insulating behavior at T = 0 in 2D (8,9). This also the case with the ≈ 0.15 compound of la 214 as shown in figure 2.

The overdoped superconducting compound shows quite a contrasting behavior. As demonstrated for ≈ = 0.20 of La214, the material remains metallic even after the superconductivity disappears for z > 0.03. The 90K-Y123 (y = 0.07) would behave in the same manner, when more Zn could be introduced, judging from considerably small residual resistivity as compared with the 60K-Y123 (y = 0.37).

A notable fact in Figure 1 is that the Zn doping induces a T-independent term in ρ_{ab} without changing the T –dependent term. This is the case also with fully oxygenated Y123 as shown in the insert of Figure 1 for detwinned single crystals with z= 0 and 0.02 in which Zn does not affect the T-linear term in ρ_a. Particularly, in the case of the oxygen reduced compound, even for the z = 0.04 where superconductivity disappears, the feature inρ_{ab} (T) around a characteristic temperature T^* remains, although the carriers tend to localize below 50 K. The results demonstrate that the Zn doping does not affect the characteristic temperature T^*in ρ_{ab} which indicates the temperature below which the spin gap starts to open (10, 11). Thus we may conclude that Zn doping increases the elastic scattering rate without much influencing the inelastic scattering process.

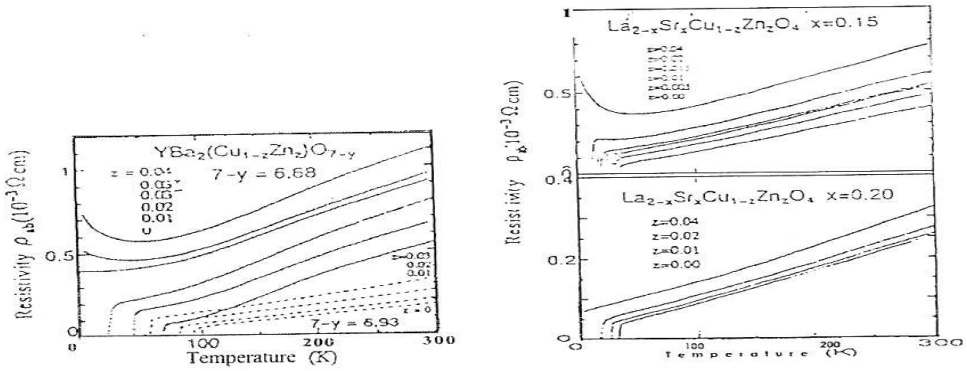

Figure 1: The Temperature Dependence of the In-Plane Resistivity of Zn-Substituted $YBa_2 Cu_3O_{7-y}$ with y = 0.37 (Solid Curves) and y = 0.07 (Dashed Curves)

Figure 2: In-Plane Resistivity for Zn-Substituted $La_{2-\chi}Sr_{\chi} CuO_4$ with = 0.15 and 0.20

We show in Figure 3 the temperature dependence of ρ_c for the oxygen reduced crystals with and without Zn doping. The relationship between the interplane charge transport and the spin gap formation has recently been suggested for underdoped Y123 (12, 13). It is shown that a crossover in ρ (T) from metallic T dependence (at high temperatures) to nonmetallic one take place at temperature near T,in this regard, the semiconducting ρ_c (T) is more sensitive to the opening or otherwise of a pseudogap. It is remarkable that both magnitude and T- dependence of ρ_c do not change with Zn- doping, providing another evidence for the persistence of the spin gap in the Zn doped compounds.

Universal T_c Depression

Figure 4 illustrates how the in-plane resistivity varies with changing oxygen content between 6.68 and 6.93 measured for a Zn-and a Xi-substituted Y123 crystal with z= 0.02 in comparison with the variation of ρ_{ab} for Zn-free Y 123 (dashed curves). One recognizes that the magnitude of the residual resistivity is fairly large in the under doped regime (y< 6.83) and is much reduced for higher oxygen contents. Using the results in Figures 1, 2 and 4, the varies of T_c normalized to the value T_{co} for Zn-free compound are plotted in Figure 5 against 2D residual resistivity for the two systems La214 and Y123 Figure 5 display another aspect of the universal T_c depression in the under doped cuprates. Irrespective of material and doped hole density the data for the under doped cuprates are near a pair-breaking curve which points toward the universal 2D resistance $h/4e^2$ as $T_c \rightarrow 0$. The universal T_c depression is reminiscent of the universal linear relation between T_c and n_s/m^* (the superconducting carrier density divided by the effective mass) discovered by Uemura et al., is most of the known high-1; cuprates in the under doped regime (14).

By Contrast, the results for the over doped cuprates show much steeper T_c-ρ_o characteristics which apparently depend on the material and the dopant concentration. For comparison, a theoretical estimate (15) assuming normal impurities in a d-wave superconductor is mapped on Figure 5. Actually the theoretical curve is near the data for 90 for Y123 but it is not clear if the same theoretical estimate can fit the universal T_c-ρ_o Y123characteristics in the under doped regime.

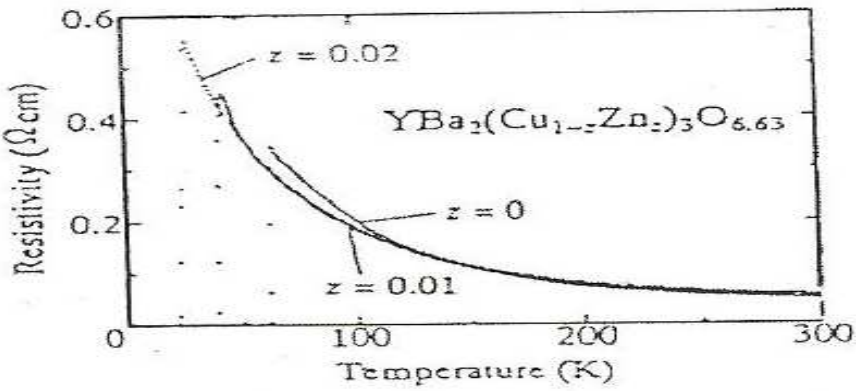

Figure 3: Temperature Dependence of the Out-Of-Plane Resistivity for the Crystals with z =o, 0.01 and 0.02 (y = 0.37)

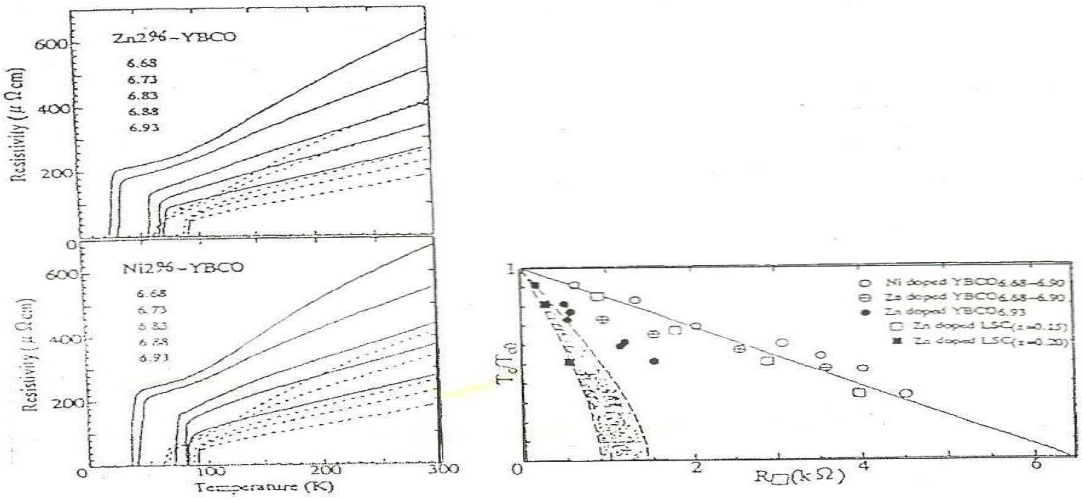

Figure 4: The Temperature Dependence of the In-Plane Resistivity of YB_{a2} $(Cu_{1-z}Zn_z)_3$ O_{7-y} and YB_{a2} $(Cu_{1-z}M_z)_3$ O_{7-y} with z = 0.02 for Various Oxygen Contents between 6.68 and 6.93 (from Top to Bottom). The Data for z = 0 Crystals are Shown by the Dashed Curves

Figure 5: Normalized Critical Temperature T_c/T_{co} Plotted as a Function of the In-Plane Residual Resistivity (Per CuO_2 Plane) due to Zn (Ni) Impurities in Y 123 and La214. Theoretical Estimates by Radtke et al. for a d-Wave Superconductor with Non-Magnetic Impurities are in the Region between the Dashed Curves. The Solid Curve is a Guide for the Eyes

DISCUSSIONS

A possible explanation for this contrasting behavior would be that the over doped material is a three-dimensional (3D) superconductor. Certainly, it is a general trend that the magnitude of the anisotropic resistivity (ρ_c/ρ_{ab}) is decreased with increase of dopant concentration (5, 16). However, the recent c-axis optical and transport study has demonstrated that the normal-state charge dynamics in the over doped regime is not strictly 3D and that a truly 3D state is realized in the over doped non-superconducting region (17). An alternative explanation in the non-universal behavior in the over doped regime may result from a phase separated state, that is, a normal metallic phase may coexist in the over doped superconducting compound. Imagine that a normal fluid in which the electrons are not easily localized due to Zn impurities forms a parallel circuit with super fluid which readily lose superconductivity and become insulating for Zn-substitution at z = 0.02.

Then, the superconductivity would disappear at resistance appreciably smaller than $h/4e^2$ and the material would keep metallicity. There is no direct evidence for such phase separation in the over doped cuprates.

Turning to the Zn-substitution effect in the normal state, the residual resistivity determined from the nearly parallel shift of the ρ (T) curves and/or from the zero temperature intercept of the T-linear part of $\rho$is plotted as a function of Zn content in Figure 6. In every case ρo increase linearly with z but the rate $d\rho_o/dz$ for the underdoped materials is about 4 times faster than that for the over doped ones. Following the analysis made by Chien et. al. (6), the resistivity arising from s-wave impurity scattering in two dimensions is $\rho o=4\ (h/€^2)n_i/n)\ \sin^2 \delta_{0,}$

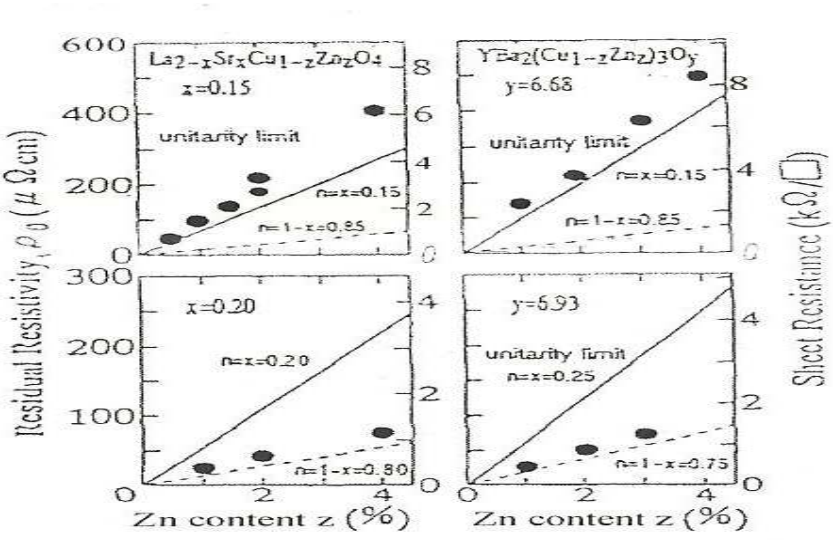

Figure 6: Variation of the Residual Resistivity with Zn Content in $YBa_2Cu_3O_{7-y}$ = 0.37 and 0.07 and in ($La_2 - _xsr_xcuo_4$). With x = 0.15 and 0.20. The Soild (Dashed) Line Indicate the Unitarity Limit with Carrier Density n=x (n=1-$_x$)

Where *ni* 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* doped hole density per Cu (2). For Y 123 we assume that *n* = $_x$~ 0.23 for y = 0.07 and *n* = $_x$~ 0.14 for y = 0.37 (21, 22) and that Zn atoms are substituted only on the plane Cu (2) sites such that *ni* = 3/2 z (1,2).

It turns out that the experimental values of ρo for the under doped materials are close to (or even larger than) the unitarity limit with n = *x*. The carrier density in the under doped regime should thus be identified with the density of doped hole number. The residual resistivity for the over doped materials becomes substantially smaller than the unitarity limit (n = *x*) but is near the uniarity limit with n = 1 - *x* Then, the decrease ρo is either from decrease in δ_0 orfrom increase in *n* toward the overdoping limit where *n* is identified with 1 – *x* as in the usual Fermi liquid. The result shown in Figure 7.

Finally, we discuss the Zn-doping effect on the transport properties in relation with the magnetic properties investigated for Zn doped Y 123 and Y 124 and Y 124. NMR studies have presented quite significant results of the Zn- doping effect on spin dynamics. For Y123 and Y124 which are in the under doped regime, $(T_1T)^{-1}$ of Cu_2 proportional to the susceptibility €" (Q w ~O) with Q = (π, π) representing antiferro magnetic (AF) wave-vector shows a Curie-Weiss like T dependence at high temperatures. As temperature goes down. $(T_1T)^{-1}$ is reduced relative to the Curie Weiss behavior below a temperature indicating spin gap formation and as a result form a peak well above T_c Upon Zn doping the peak of

$(T_1T)^{-1}$ is washed out and $(T_1T)^{-1}$ continues to increase ith lowering temperature (24). This result seems in to indicate that the spin gap at Q = (π, π) disappears. The sensitiveness of $(T_1T)^{-1}$ to Zn doping is in sharp contrast to the *T* dependences of ρ_{ab} *(T)* and p, (T). In particular p, (T) which is expected to have a more intimate connection with the spin gap, shows no significant change with Zn doping. This fact may indicate that the spin gap at q = Q has no direct connection to the charge transport.

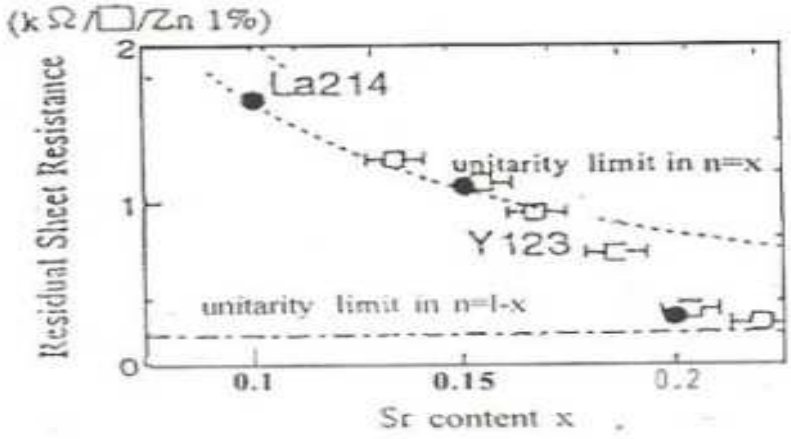

Figure 7: Variation of the Residual 2D Resistivity (per 1%Zn) with Doped Hole Density in Y123 and La214

As evidenced by the recent NMR experiment (27), a Zn impurity effects the Cu (2) sites only in its vicinity and it does not change the global feature of the spin pseudo gap. We may conclude that the charge transport has a connection with the global feature in the spin fluctuations in the momentum space, not nith a particular spin gap, say, at q = Q (27).

The effect of Zn doping does not change the main feature in the high temperature region. A difference becomes apparent only in the low temperature region, where a curie term in X_m due to local magnetic develops progressively with Z. The induced moment is estimated to be ~ 0.8 $\mu_ß$ per Zn. A surprising fact is that the Curie term, and evidence for the localized magnetic moments induced by non-magnetic An, manifests itself in X, (T) even for Z < 0.04, whereas the in-plane resistivity is metallic over the entire temperature range above Tc without showing any indication for carrier localization. This is quite anomalous in view of the NMR evidence that Cu (3d) and O (2p) holes forms a strongly hybridized sate that is the decoupling into localized Cu (3d) spins and O (2p) charge carriers does not take place in the CuO_2 plane. Formation of localized moments in the metallic state is a challenging problem for both Fermi liquid and non-Fermi liquid theories. In the Fermi liquid picture where the same electron carries both charge and spin, it seems hard to suppose that the strong impurities, in the unitarity scattering limit, produce localized magnetic moments in the metallic state. Actually, in the highly doped material 90K-Y123, Zn induces a reduced magnetic moment and the magnetic moment is much more reduced in the over doped

$La2\text{-}xSr_xCuO_4$ with x = 0.30

CONCLUSIONS

We have presented the experimental results of Zn-substitution effects on the in-plane resistivity of single crystals high T_c cuprates with various hole densities. It is found that the doped Zn acts as a strong scatterer for the in-plane charge transport, but the principal feature in the *T* dependences of the transport coefficients, associated possible with the opening of a spin pseudo gap, is not affected by Zn doping, From these results we conclude that the charge transport has relevance

to local spin fluctuations not restricted a q = Q in the momentum space, It is also characteristic of the under doped spin gap regime that Zn induces a fairly large local magnetic moments (~0.8$\mu_{ß}$) even when charge carriers are not localized.

Zn probes a critical change in the electronic state upon traversing between the under doped and the over doped superconducting regime, and reveals a universal T_c depression and a subsequent. Superconductor insulator transition in the under doped regime. The results suggest a first order electronic phase transition or/ and an abrupt 2D-3D dimensional crossover occurring between the under doped and the over doped regime.

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