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Suppression of T_c in Co-doped $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ superconductor

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ABSTRACT

The most possible sources of suppression of superconducting properties (i.e., zero resistivity $T_c(R=0)$, magnitude of diamagnetism, etc.) in cobalt-doped $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ ($x = 0$, 0.05, 0.1, 0.5) superconductor have been explored. It has been observed from these studies that Co-doping at Cu^{2+} sites in the CuO_2 planes somehow localizes the carriers at Co^{3+} sites and decreases the density of mobile carriers in the superconducting CuO_2 planes. We doped Li at Tl sites in $Cu_{0.5}Tl_{0.5}Ba_2O_{4-\delta}$ charge reservoir to obtain an efficient carriers doping $(Cu_{0.5}Tl_{0.25}Li_{0.25})Ba_2O_{4-\delta}$ charge reservoir layer of $(Cu_{0.5}Tl_{0.25}Li_{0.25})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ compound to replenish the density of mobile carriers in the CuO_2 planes; since alkali metals are known to lose their outermost "s-orbital" electron easily, which can be supplied to the superconducting CuO_2 planes. The increased free carrier density in the superconducting CuO_2 planes enhances the superconducting properties of the final compound.

1. Introduction

Since the discovery of oxide superconductors the exact mechanism of superconductivity at microscopic level has not been completely explored yet. In cuprate high temperature superconductors (HTSCs) $MBa_2Ca_{n-1}Cu_nO_{2n+4-\delta}$ [$n = 2$–5 and M = Y, Tl, Hg, Bi, Cu, (Cu,Tl)] superconductor families, the two major constituents of unit cell are $MBa_2O_{4-\delta}$ a charge reservoir layer and $nCuO_2$ conducting planes [1]. The key structural unit in the oxide superconductors is the conducting CuO_2 planes, which is considered to be responsible for the superconductivity [2–5]. The charge reservoir layer supplies carriers to the conducting $nCuO_2$ planes and these carriers in CuO_2 planes become the source of superconductivity. The carriers in CuO_2 planes can be perturbed by directly doping at the planner sites. Many research groups working in the field of high T_c superconductivity focus their attention on the CuO_2 planes; they substitute different elements such as Co, Ni, Fe and Zn at Cu sites in CuO_2 planes in order to study the response of the superconductor material to a disorder created by these ions in CuO_2 planes [6–14]. In these studies, it was found that doping of such impurities leads to a decrease in the critical temperature "T_c". The proposed reasons for the T_c suppression after the substitution of impurity atoms in CuO_2 planes are magnetic pair breaking, potential scattering of carriers, localization of carriers and suppression of effective pairing interaction [15–17]. The magnetic ions Co, Ni and Fe produce the magnetic pair breaking and localization of carriers due the interaction between the Cooper pairs and the magnetic spins of the impurity atoms [18–20]. The $Cu_{1-x}Tl_xBa_2Ca_2Cu_3O_{10-\delta}$ ($x = 0.4$–0.8) superconductor is a promising material in cuprate family of superconductors due to its low anisotropy and the ability of carrying high critical current [21,22]. In this article, we substituted Co at Cu in CuO_2 planar sites in $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_3O_{10-\delta}$ superconductor to observe the response of these materials to the substitution of magnetic impurity in CuO_2 planes. This material has Tl^{3+}/Cu^{2+} atoms in the charge reservoir layer; therefore, the charge transfer mechanism to CuO_2 planes is different from that of other cuprates, which has only trivalent atoms in their charge reservoir layer [23–25]. Therefore, we expected some different results as compared to the other superconductor materials after Co-doping. However, our experiments on Co-doped $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ superconductors have suggested that the mobile free carriers are most likely to be localized at Co^{3+} sites in CuO_2 planes due to (3+) oxidation state of cobalt [26]. These studies have also suggested that if we replenish the free carriers by some other means to CuO_2 planes, superconduc-

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Table 1
Fraction of different phases present in the $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ ($x = 0$, 0.05, 0.1, 0.5) superconductor.

Sr. no.	Co-concentration	% of 1223	% of 1234	% of 1212	*a*-axis (Å)	*c*-axis (Å)
1	0	95	4	1	3.98	15.02
2	0.05	94	4	2	4.03	15.08
3	0.1	94	5	1	4.04	15.32
4	0.5	95	4	1	3.89	15.88

tivity can be restored in Co-doped $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ material. Therefore, we have also substituted Li at Tl sites in the $(Cu_{0.5}Tl_{0.5})Ba_2O_{4-\delta}$ charge reservoir layer to obtain an efficient carriers doping $(Cu_{0.5}Tl_{0.25}Li_{0.25})Ba_2O_{4-\delta}$ charge reservoir layer of $(Cu_{0.5}Tl_{0.25}Li_{0.25})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ compound. The objective of Li-doping was to supply required number of mobile carriers to CuO_2 planes, which would have been depleted due to the localization of carriers at Co^{3+} in CuO_2 planes; the supply of excess carriers could restore the superconductivity in Co-doped samples [27,28].

2. Experimental

The samples were prepared by solid-state reaction method accomplished in two steps. In the first step $Cu_{0.5}Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ ($x = 0$, 0.05, 0.1, 0.5) and $(Cu_{0.5}Li_{0.25})Ba_2Ca_2Cu_{2.5}Co_{0.5}O_{10-\delta}$ precursor materials were synthesized by using $Ba(NO_3)_2$, $CaCO_3$, Cu(CN), $Co(NO_3)_3$ and LiCl as starting compounds. These compounds were mixed in appropriate ratios and ground for about an hour in an agate mortar and pestle. The ground material was loaded in quartz boat and fired in chamber furnace at 850 °C for 24 h followed by furnace cooling to room temperature. The precursor material was then mixed with Tl_2O_3 and ground for about an hour. Thallium mixed material was then palletized under 3.2 tonnes/cm^2 pressure and the pellets were wrapped in a gold capsule. Pellet containing gold capsule was heat-treated at 850 °C for 10 min and quenched to room temperature to get $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ ($x = 0$, 0.05, 0.1, 0.5) and $(Cu_{0.5}Tl_{0.25}Li_{0.25})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ as final reactants compositions. The resistivity of these samples was measured by four probe method and ac-susceptibility by using SR530 Lock-in Amplifier at a frequency of 270 Hz with $H_{AC} = 5.69$ mOe of primary coil at various temperatures. The bar shaped samples of dimension 2 mm × 10 mm × 2.5 mm were used for resistivity and ac-susceptibility measurements. The structure of material was determined by using X-ray diffraction scan (D/Max IIIC Rigaku) with a CuKα source of wavelength 1.54056 Å and cell parameters were determined with the help computer software program (i.e., Check cell).

3. Results and discussion

The X-ray diffraction (XRD) scans of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ ($x = 0$, 0.05, 0.1, 0.5) {CuTl-1223} samples prepared at 850 °C are shown in Fig. 1. Most of the diffraction lines are indexed according to tetragonal structure following *P4/mmm* space group. The inclusions of derivative phases such as Cu–Tl–Ba–Ca–Cu–Co–$O_{12-\delta}$ {CuTl-1234} and Cu–Tl–Ba–Ca–Cu–Co–$O_{8-\delta}$ {CuTl-1212} were also observed in the XRD scans. The relative percentages of these phases with Co-doping concentrations are given in Table 1, which were calculated by using the following relations [29].

% of CuTl-1223 phase

$$= \frac{\sum I(\text{CuTl-1223})}{\sum I(\text{CuTl-1223}) + \sum I(\text{CuTl-1234}) + \sum I(\text{CuTl-1212})} \times 100$$

% of CuTl-1234 phase

$$= \frac{\sum I(\text{CuTl-1234})}{\sum I(\text{CuTl-1223}) + \sum I(\text{CuTl-1234}) + \sum I(\text{CuTl-1212})} \times 100$$

% of CuTl-1212 phase

$$= \frac{\sum I(\text{CuTl-1212})}{\sum I(\text{CuTl-1223}) + \sum I(\text{CuTl-1234}) + \sum I(\text{CuTl-1212})} \times 100$$

‘*I*’ is the intensity of the phases present in the final compound.

The lengths of ‘*a*’- and ‘*c*’-axes increase with the increase in Co-doping at Cu sites in CuO_2 planes. The dominant phase in all these samples is CuTl-1223, while the other phases in minute quantity are due to the impurities arising during the preparation processes of these samples [10]. Some additional phases such as CuTl-1234 and CuTl-1212 are observed along with the desired CuTl-1223 phase. We have taken XRD spectra with a very slow scan speed of 1°/min to observe even small amount of impurities.

The resistivity measurements of Co-doped $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ ($x = 0$, 0.05, 0.1, 0.5) samples with magnetic ac-susceptibility measurements in inset are shown in Fig. 2. A metallic variation of resistivity from room temperature down to onset of the superconductivity is the salient feature of these samples. The zero resistivity critical temperature $\{T_c(R = 0)\}$ is observed around 114, 104, 84 and 80 K for Co-doping concentration of $x = 0$, 0.05, 0.1, 0.5, respectively. The increased concentration of Co ($x \geq 0.5$) shifts the transition temperature below 77 K. The magnetic ac-susceptibility curves of these samples

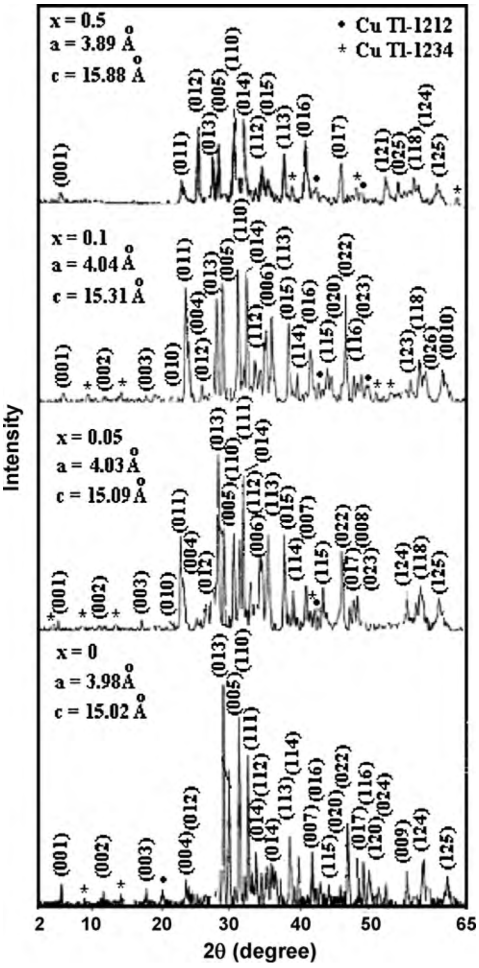

Fig. 1. The X-ray diffraction (XRD) scans of $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ ($x = 0$, 0.05, 0.1, 0.5) superconductors and “*” & “·” denote the CuTl-1234 and CuTl-1212 phases, respectively.

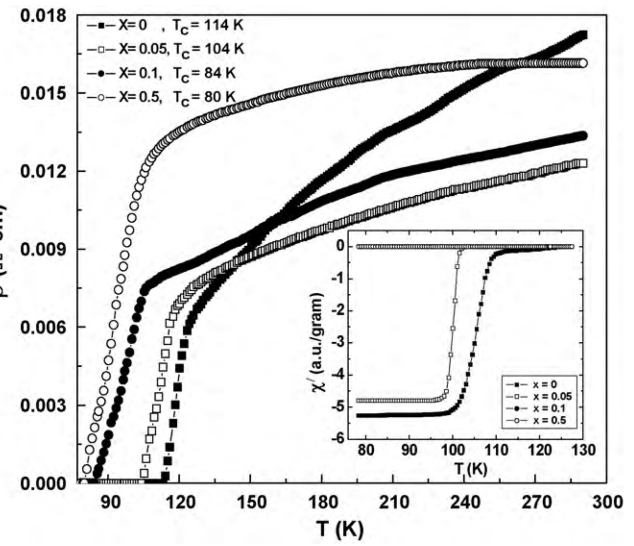

Fig. 2. The resistivity vs temperature measurements of $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ (x = 0, 0.05, 0.1, 0.5) superconductors and (in the inset magnetic ac-susceptibility measurements $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ (x = 0, 0.05, 0.1, 0.5) superconductors).

show that the onset of diamagnetism is around 112, 103 K for x = 0, 0.05 and less than 77 K for x = 0.1, 0.5. The magnitude of the diamagnetism observed from magnetic ac-susceptibility measurements is decreased with the increased concentration of Co in $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ samples. This effect may appear due to the pair breaking caused by the substitution of ferromagnetic impurities. The effects of carrier concentration on the superconductivity of Co-doped $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ samples are explored by Li-doping at Tl sites in $(Cu_{0.5}Tl_{0.25}Li_{0.25})Ba_2O_{4-\delta}$ charge reservoir layers. The doping of Li at Tl sites in the charge reservoir layer has been found to enhance the superconducting properties of the final compound. The density of mobile carriers in CuO_2 planes has been possibly increased by the substitution of Li at Tl sites in the charge reservoir layer required for superconductivity at higher critical temperature. If magnetic pair breaking effect has become dominant, it would be able to suppress T_c in spite of an increase in the density of mobile carriers in CuO_2 planes. But we have observed that the rate of T_c suppression with Co-doping is decreased with the increase of mobile carriers density in CuO_2 planes by the substitution of Li at Tl sites in the charge reservoir layer. This may be due to carrier localization effect or may also be due to hole filling in CuO_2 planes. Hence the different possible sources of suppression of $T_c(R=0)$ with Co-doping are mentioned above but the localization of carriers at Co^{3+} sites in CuO_2 plane seems to be the most promising one or it may also be due to hole filling in CuO_2 planes. It was observed that superconductivity was not improved even after the substitution of Mg and Be at Ca sites in Co-doped samples [30]. One of the possible sources of suppression of superconductivity with Co-doping is the localization of carriers at Co^{3+} sites in CuO_2 planes [27,28]. The substitution of Mg and Be could not remove the localization effects due to the deficiency of carriers in CuO_2 planes [30]. In order to put this root cause to a test, we have deliberately doped Li at $(Cu_{0.5}Tl_{0.5})Ba_2O_{4-\delta}$ charge reservoir layer sites to have $(Cu_{0.5}Tl_{0.25}Li_{0.25})Ba_2O_{4-\delta}$ charge reservoir layer. This has been done to replenish the localized density of carriers in the conducting planes [27,28,31]. Lithium has been chosen for this purpose due to its lowest ionization potential (i.e., ionization energy of Li is 124 kcal/g mol) and smaller size (ionic radius of Li is 0.72 Å). Alkali metals are known to lose their outer most electrons very easily, which can be delivered to the conducting CuO_2 planes. The X-ray diffraction scan (XRD) of Li-doped $(Cu_{0.5}Tl_{0.25}Li_{0.25})Ba_2Ca_2Cu_{2.5}Co_{0.5}O_{10-\delta}$ superconductor sample is shown in Fig. 3. The Li free $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{2.5}Co_{0.5}O_{10-\delta}$ sample has shown $T_c(R=0)$ around 80 K in resistivity measurements, and has shown no sign of diamagnetic signal up to 77 K. The absence of diamagnetism in $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{2.5}Co_{0.5}O_{10-\delta}$ sample is most likely to arise from the localization of carriers at Co^{3+} sites. We have replenished this density of mobile charge carriers by Li-doping at the charge reservoir layer sites by synthesizing $(Cu_{0.5}Tl_{0.25}Li_{0.25})Ba_2Ca_2Cu_{2.5}Co_{0.5}O_{10-\delta}$ superconductor. The samples have shown $T_c(R=0)$ around 96 K and sizable magnitude of diamagnetism in the inset of Fig. 4. These experiments have given irrefutable evidence that the Co^{3+} ions develop localization of the carriers at their sites in the CuO_2 planes and suppress the density of mobile carriers. The decreased density of free carriers in the conducting planes decreases the Fermi velocity $V_F = (\pi\xi_c\Delta/\hbar^2)$, coherence length $\xi_c = (\hbar K_F/m2\Delta)$ and hence their order-parameter $\Delta = (1.176K_BT_c)$. In the previous experiments of Li-doping in Bi-2212 superconductors, it was found that the $T_c(R=0)$ increases with the increase in Li content in the compound [32–39]. The reason of $T_c(R=0)$ enhancement is suggested to be due the optimization of carriers in CuO_2 planes.

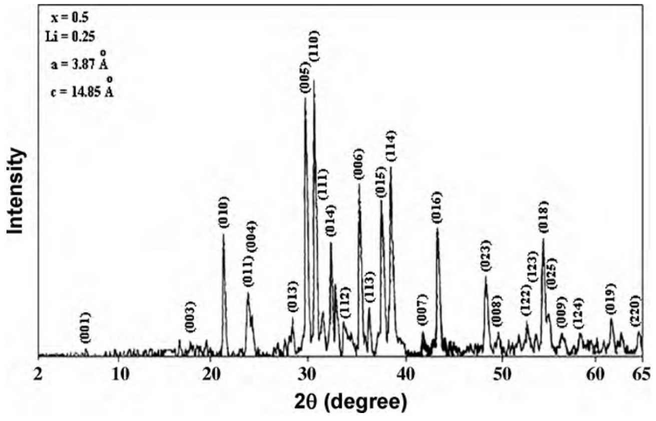

Fig. 3. The X-ray diffraction (XRD) scan of $(Cu_{0.5}Tl_{0.25}Li_{0.25})Ba_2Ca_2Cu_{2.5}Co_{0.5}O_{10-\delta}$ superconductor.

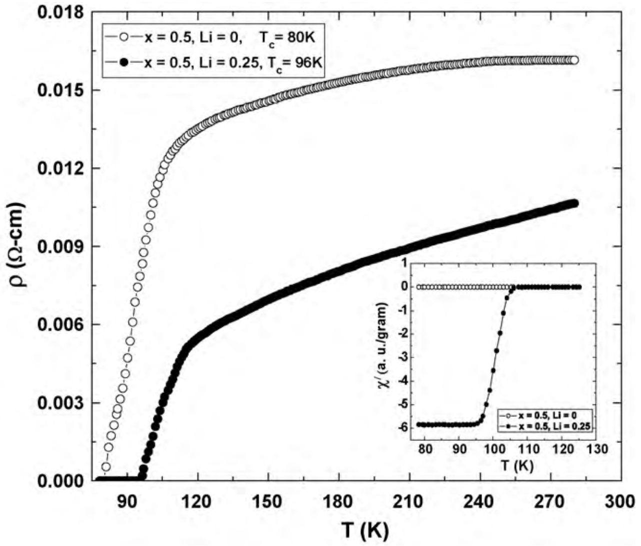

Fig. 4. The resistivity vs temperature measurements of $(Cu_{0.5}Tl_{0.25}Li_{0.25})Ba_2Ca_2Cu_{2.5}Co_{0.5}O_{10-\delta}$ (Li = 0, 0.25) superconductors and (in the inset magnetic ac-susceptibility measurements $(Cu_{0.5}Tl_{0.25}Li_{0.25})Ba_2Ca_2Cu_{2.5}Co_{0.5}O_{10-\delta}$ (Li = 0, 0.25) superconductors).

4. Conclusions

We have successfully synthesized Co-doped $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ ($x = 0$, 0.05, 0.1, 0.5) samples. The zero resistivity critical temperature $\{T_c(R=0)\}$ and magnitude of diamagnetism are found to decrease with increased Co-doping. We can conclude from the preceding results that substitution of magnetic impurities such as Co at the Cu sites in CuO_2 planes is a source of localization of carriers. The localization of these carriers is a source suppression of superconductivity as observed from the lowering of $T_c(0)$ in $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ samples or it may also be due to hole filling in CuO_2 planes. However, the localization of carriers can be compensated if more carriers are supplied to CuO_2 planes. The Li-doping at Tl sites in the $(Cu_{0.5}Tl_{0.25}Li_{0.25})Ba_2O_{4-\delta}$ charge reservoir layer of $(Cu_{0.5}Tl_{0.25}Li_{0.25})Ba_2Ca_2Cu_{2.5}Co_{0.5}O_{10-\delta}$ has been found to enhance the superconducting properties. The substitution of Li has provided a sufficient number of mobile carriers to CuO_2 planes required for superconductivity at higher critical temperature. These observations have shown that the magnetic impurity Co^{3+} has developed localization of the carriers at its sites in the CuO_2 planes or it may also be due to hole filling in CuO_2 planes and hence due to deficiency of mobile carriers, the superconductivity has been suppressed in Co-doped $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_{3-x}Co_xO_{10-\delta}$ superconductor.

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