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ScienceDirect

Materials Chemistry and Physics 105 (2007) 298–302

MATERIALS CHEMISTRY AND PHYSICS

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Suppression of anti-ferromagnetism by enhanced solubility of Ni in $Cu_{1-x}Tl_xBa_2Ca_2Cu_{3-y}Ni_yO_{10-\delta}$ ($y = 0, 0.5, 1.0, 1.5$) superconductor

Nawazish A. Khan*, Najmul Hassan

Materials Science Laboratory, Department of Physics, Quaid-i-Azam University, Islamabad, Pakistan

Received 27 November 2006; received in revised form 25 April 2007; accepted 27 April 2007

Abstract

Enhanced solubility of Ni in CuO_2 planes of $Cu_{1-x}Tl_xBa_2Ca_2Cu_3O_{10-\delta}$ has been observed. The main objective of ferromagnetic Ni substitution in $Cu_{1-x}Tl_xBa_2Ca_2Cu_3O_{10-\delta}$ superconductor at Cu sites was to reduce any possible anti-ferromagnetic order existing in the inner CuO_2 planes (IP); this anti-ferromagnetism is suggested to be suppressing the zero resistivity critical temperature [T_c ($R = 0$)]. If the anti-ferromagnetic order has some role in bringing about superconductivity at a particular temperature, the doping of ferromagnetic Ni would destroy it and hence the superconductivity. Our studies have shown that the doping of 50% ferromagnetic Ni at Cu sites in CuO_2 planes does not destroy the superconductivity; most likely reasons for the enhanced superconductivity have also been discussed. The increased doping of Ni beyond 50% destroys superconductivity and the final material becomes perfect insulator. These studies have suggested that Ni possibly breaks the anti-ferromagnetism existing in the inner CuO_2 planes, and the critical temperature is not suppressed very much. The post-annealing experiments demonstrated that the magnitude of diamagnetism is enhanced when carriers are optimum in CuO_2 planes. These experiments have contradicted the previous notion of non-uniform doping of inner (IP) and outer planes (OP) as a source of suppression of [T_c ($R = 0$)] of final compound. These experiments have also manifested that the superconductivity and ferromagnetism can co-exist.

PACS: 74.76.−w; 74.76.Bz; 74.72.−h; 74.72.−Jt

Keywords: Ni doped $Cu_{1-x}Tl_xBa_2Ca_2Cu_{3-y}Ni_yO_{10-\delta}$ ($y = 0, 0.5, 1.0, 1.5$) superconductors; Magnetic materials; Fermi surface; Annealing

1. Introduction

Among the unresolved issues in high T_c cuprates, the most important is the role of anti-ferromagnetism (AF) in the mechanism of superconductivity (SC) in CuO_2 planes, constituting SC-AF-SC alternative layered structure [1,2]. In the unit cell of $Cu_{1-x}Tl_xBa_2Ca_2Cu_3O_{10-\delta}$ the two outer-pyramidal CuO_2 planes (OP) have five fold oxygen coordination and an inner CuO_2 (IP) with four fold oxygen coordination [3]. Nuclear magnetic resonance experiments have shown that OP are over-doped and the IP is under-doped with carriers [4–6]. Outer-pyramidal CuO_2 planes have higher carrier density because of their presence in the vicinity of $Cu_{1-x}Tl_xBa_2O_{4-\delta}$ charge reservoir layer. The other possible reason could be the formation of anti-ferromagnetic order in the Cu atoms in the inner CuO_2 plane, which can lower the energy of the carriers tied to it [1,2]. The outer planes has a superconductivity around 108 K whereas the inner plane has around 60 K. The question arises whether there is any role of anti-ferromagnetic order suggested to be present in the IP in reducing the T_c ($R = 0$); the inner planes are suggested to attain this anti-ferromagnetic state due to the deficiency of carriers. In order to settle these outstanding issues we have doped the CuO_2 planes of $Cu_{1-x}Tl_xBa_2Ca_2Cu_3O_{10-\delta}$ with ferromagnetic Ni which is expected to break the anti-ferromagnetism existing in the inner-plane. The lower T_c ($R = 0$) of IP would increase if anti-ferromagnetism is broken. In the previous studies Ni^{2+} ion has been found to suppress T_c ($R = 0$) in all the families of high temperature superconductors. It was suggested in these studies that the localized magnetic moment of Ni^{2+} promotes the pair breaking effects which decreases T_c ($R = 0$) [7]. In some other studies, the variation of density of states near the Fermi level is suggested as one of possible reasons for the decrease of T_c ($R = 0$) in the Ni doped compounds [1]. Contrary to all the previous studies [7–16], we have observed marginal suppression of T_c ($R = 0$) by doping Ni at the CuO_2 planar sites. These studies have suggested that ferromagnetic

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* Corresponding author. Tel.: +92 51 2875906.
E-mail address: nawazishalik@yahoo.com (N.A. Khan).

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doi:10.1016/j.matchemphys.2007.04.064

Ni changes the anti-ferromagnetism present in the inner CuO_2 plane and makes the distribution of carriers homogeneous in inner and outer planes; therefore, the critical temperature is not suppressed very much as observed in the previous studies. The most possible source of enhanced superconductivity in Ni doped $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{1.5}Ni_{1.5}O_{10-\delta}$ system has also been discussed in this article.

2. Experimental

The samples were prepared by solid-state reaction method accomplished in two stages. At the first stage $Cu_{0.5}Ba_2Ca_2Cu_{3-y}Ni_yO_{10-\delta}$ ($y = 0$, 0.5, 1.0, 1.5) precursor material was synthesized using $Ba(NO_3)_2$, $Ca(NO_3)_2$, Cu(CN) and $Ni(NO_3)_2$ as starting compounds. These compounds were mixed in an appropriate ratio in a quartz mortar and pestle. Thoroughly mixed material was fired in air in a quartz boat at 850 °C for 24 h followed by furnace cooling to room temperature. The precursor material was then ground for about an hour and mixed with Tl_2O_3 to give $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Ni_yO_{10-\delta}$ ($y = 0$, 0.5, 1.0, 1.5) as a final reactants composition. Thallium mixed material was then pelletized under 3.2 tonnes cm^{-2} and pellets were wrapped in a gold capsule. Pellet containing gold capsule was heated at 850 °C for 10 min and quenched to room temperature after the heat treatment. The resistivity of the samples was measured by four-probe method and the diamagnetism by ac-susceptibility measurements at lock-in frequency of 270 Hz. The superconductor phase was identified by X-ray diffraction scans (XRD). The post annealing of the samples was carried out in a tubular furnace in flowing N_2 or O_2 atmosphere.

3. Results and discussion

In Fig. 1 are shown the X-ray diffraction scans of one of the representative $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{1.5}Ni_{1.5}O_{10-\delta}$ sample prepared at 850 °C. Most of the diffraction lines could be indexed according to tetragonal structure following *P*4/*mmm* space group; the Ni substituted impurities $Cu_{0.5}Tl_{0.5}Ba_2CaCuNiO_{10-\delta}$ and $Cu_{0.5}Tl_{0.5}Ba_2Ca_3Cu_2Ni_2O_{10-\delta}$ are also marked in the diffraction scans. The lengths of *a*- and *c*-axes decrease with increased Ni doping in $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Ni_yO_{10-\delta}$ ($y = 0.5$, 1.0, 1.5). The variation of axes length with Ni concentration is given in the Table 1.

The resistivity measurements of Ni doped $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Ni_yO_{10-\delta}$ superconductor sample are shown in Fig. 2. Metallic variation of resistivity from room temperature down to onset of superconductivity is a typical feature of these samples. These samples have shown T_c ($R = 0$) around 95.4, 94.4, 89 and 90 K for Ni doping concentration of y = 0, 0.5, 1.0 and 1.5, respectively. The higher doping concentration of Ni in CuO_2 planes ($y \geq 2.0$) makes $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Ni_yO_{10-\delta}$ samples highly resistive and their resistivity by four-probe method could not be measured. These observations also lead to a conjecture that the superconductivity is destroyed if number of Ni atoms per CuO_2 plane is more than half the number of Cu atoms. The ac-magnetic susceptibility of these samples is shown in Fig. 3. The magnitude of the diamagnetism is decreased with the increased Ni concentration in $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Ni_yO_{10-\delta}$

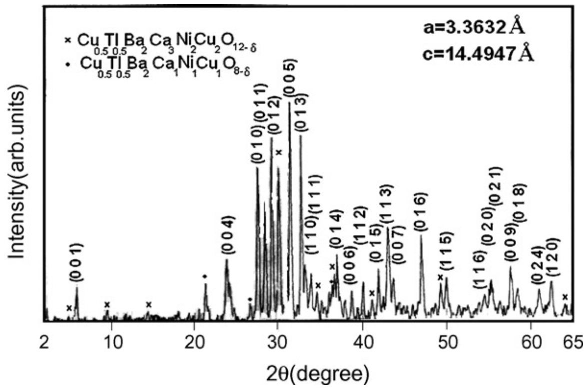

Fig. 1. X-ray diffraction pattern of superconductor sample of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{1.5}Ni_{1.5}O_{10-\delta}$.

Table 1
Variation of axes length with Ni content

Ni concentration	*a*-Axis length (Å)	*c*-Axis length (Å)
0.0	3.8294	14.9245
0.5	3.6978	14.7934
1.0	3.4996	14.5929
1.5	3.3632	14.4947

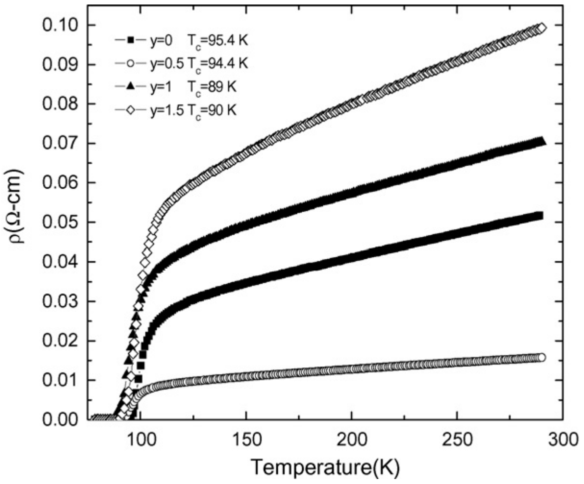

Fig. 2. Resistivity measurements vs. temperature of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Ni_yO_{10-\delta}$ ($y = 0$, 0.5, 1.0, 1.5) superconductor samples.

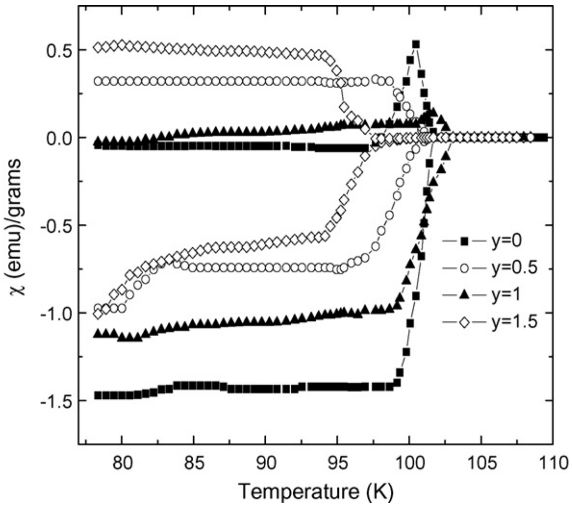

Fig. 3. ac-Susceptibility measurements vs. temperature of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Ni_yO_{10-\delta}$ ($y = 0$, 0.5, 1.0, 1.5) superconductor samples.