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# Studies of fluctuation induce conductivity of Mg doped $Cu_{0.5}Tl_{0.5}Ba_2(Ca_{2-y}Mg_y)(Cu_{0.5}Zn_{2.5})O_{10-\delta}$ (y=0, 0.5, 1.0) superconductors

Nawazish A. Khan\*, Asifa Mumtaz

Materials Science Laboratory, Department of Physics, Quaid-i-Azam University, Islamabad 45320, Pakistan

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#### ABSTRACT

The electrical resistivity  $\rho(T)$  versus temperature data of as-prepared and  $O_2$ -annealed  $Cu_{0.5}Tl_{0.5}Ba_2$  ( $Ca_{2-y}Mg_y$ )( $Cu_{0.5}Zn_{2.5}$ ) $O_{10-\delta}$  (y=0, 0.5, 1.0) samples is analyzed for fluctuations induced conductivity analysis in the temperature regime well above the critical temperature. The analysis has been carried out employing Aslamazov and Larkin (AL) and Lawrence and Doniach (LD) models. The coherence length, inter-planar coupling, exponent and dimensionality of fluctuations are determined from this analysis. It has been observed in as-prepared and oxygen post-annealed samples that the crossover temperature associated with two distinct exponents and the excess conductivity data fits very well with the two-dimensional (2D) and three-dimensional (3D) AL equations. The cross-over temperature  $T_0$  is shifted to lower temperatures with enhanced Mg doping. The width of 3D AL region is reduced with increased Mg doping, however, some improvement is observed after post-annealing in the oxygen atmosphere. Moreover, a decrease in the coherence length along the c-axis is observed with the doping of Mg in the final compound. We have elucidated from these analyses that higher thallium contents in the final compound promote a decrease in the density of mobile carriers in  $CuO_2$  planes, which suppresses the coherence length along the c-axis and superconductivity parameters.

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#### 1. Introduction

Oxide high- $T_c$  superconductors (HTSC) are anisotropic and anisotropy in their physical properties arises due to ease in the motion of the carriers in the two dimensional conducting CuO<sub>2</sub> planes [1–5] whereas this motion is impeded by the movement of the carriers normal to these planes. This motion is restricted by the thickness of the MBa<sub>2</sub>O<sub>4- $\delta$ </sub> (M=Y, Bi, Hg, Tl, CuTl, etc) charge reservoir layers and the carriers have to tunnel every time to overcome the thickness of these barriers and fluctuations in the order parameter of the carrier are produced due to this impeded motion. It is widely accepted that higher critical temperature in the oxide superconductors arises due to the formation of the Cooper-pairs well above the critical temperature. These Cooperpairs are simultaneously formed and broken as we approach  $T_c$ , consequently resulting into a representative critical temperature which is much higher than conventional HTSC's. The formation of these Cooper pairs may give rise to excess conductivity of the carriers. There are two main sources of excess conductivity (a) Cooper-pairs formed above the critical temperature arising from the fluctuation induced conductivity given by Aslamazov-Larkin (AL) contribution [6], which is explained by Lawrence-Doniach (LD) model [7] and it predicts a crossover from three dimensional (3D) electronic state of the system to a two dimensional (2D) one, with increasing temperature. In layered oxide superconductors, AL term dominates close to  $T_{\rm c}$ . (b) The effect of superconducting fluctuations on the conductivity of normal electrons [8] given by Maki–Thompson (MT) contribution. The MT term depends largely on the phase-relaxation time  $t_{\varphi}$  and becomes important in 2D fluctuations regime with a moderate pair-breaking [9]. The investigation of intrinsic properties of a material via the fluctuation-induced conductivity (FIC) is acceptable experimental method using the normal state properties of materials well above the critical temperature  $T_{\rm c}$ . The excess conductivity is given by

$$\Delta \sigma = [\rho_{N}(T) - \rho(T)]/[\rho_{N}(T)\rho(T)] \tag{1}$$

where  $\sigma(T) = [(1/\rho(T)]$  is the actually measured conductivity and  $\rho_{\rm N}(T) = \alpha + \beta T$  the extrapolated normal state resistivity ( $\alpha$  is an intercept and  $\beta$  a slope of straight line).

Through the analysis of experimentally observed data for FIC analysis, we can achieve the reliable values of  $\xi_{\rm c}(T)$ ,  $t_{\varphi}$ , and dimensionality of conductivity. Due to a very short Ginzburg–Landau coherence length  $\xi_{\rm GL}(0)$  of oxide HTSC's, it is advisable to look into the FIC analysis of  $\Delta\sigma(T)$  at temperatures well above the  $T_{\rm C}$ .

Soon after the discovery of high temperature superconductivity in oxides, the excess conductivities analyses for  $YBa_2Cu_3O_{7-\delta}$  superconductors have been carried out on bulk and thin film

<sup>\*</sup> Corresponding author.

E-mail address: nawazishalik@yahoo.com (N.A. Khan).

samples using various fitting methods. Most of these fitting schemes have been carried out on the polycrystalline bulk samples which showed three-dimensional thermal fluctuations [10–14], whereas *c*-axis-oriented thin films have shown 2D–3D crossover [15]. Mixed results have been reported in the analysis of single crystal samples [16,17]. However, two dimensional fluctuations have been observed in Bi-2212 and Bi-2223 samples prone to their higher anisotropy [18–22].

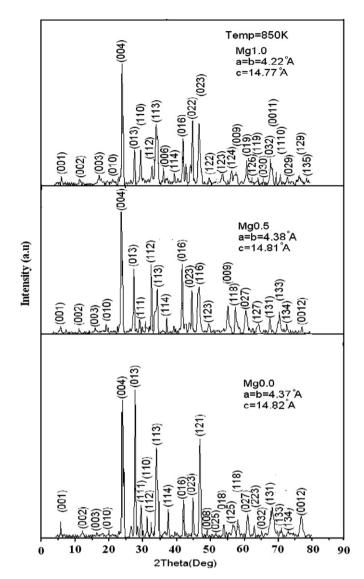
We have recently synthesized  $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Zn_yO_{10-\delta}$  (y=0, 0. 5, 1.0, 1.5, 1.5, 2.0, 2.5) samples with enhanced superconductivity at normal pressure. Contrary to the previous observations on Zn doped samples, we have observed enhanced superconductivity equipped with higher critical temperatures with increased Zn doping. In order to investigate the root causes of enhanced superconductivity in these compounds, we have chosen  $Cu_{0.5}Tl_{0.5}Ba_2(Ca_{2-y}Mg_y)(Cu_{0.5}Zn_{2.5})O_{10-\delta}$  (y=0, 0.5, 1.0) and investigated these samples for fluctuation induced conductivity (FIC) through excess conductivity analysis. The main objectives of Mg doping were to investigate the effect of enhanced inter-plane coupling on the conductivity and hence on the excess conductivity of the carriers.

#### 2. Experimental

Samples were synthesized by solid-state reaction method, accomplished in two stages. At the first stage  $Cu_{0.5}Ba_2(Ca_{2-y}Mg_y)$  $(Cu_{0.5}Zn_{2.5})O_{10-\delta}$  (y=0, 0.5, 1.0) precursor materials were prepared using Cu(CN), Ba(NO<sub>3</sub>)<sub>2</sub>, Ca(NO<sub>3</sub>)<sub>2</sub>, MgO, ZnO as starting compounds. These compounds were mixed in appropriate ratios in an agate mortar and pestle. Thoroughly mixed materials were fired twice in air in a quartz boat at 850 °C for 24 h by furnace cooling to room temperature. The precursor material was then grounded for about an hour and mixed with Tl<sub>2</sub>O<sub>3</sub> (99%, Merck) to give  $Cu_{0.5}Tl_{0.5}Ba_2(Ca_{2-y}Mg_y)(Cu_{0.5}Zn_{2.5})O_{10-\delta}$  (y=0, 0.5, 1.0) as final reactant compositions. Thallium mixed material was then palletized under 3.4 tons/cm<sup>2</sup> pressures and wrapped in a gold capsule. Pellet enclosed in gold capsule was heat treated at 850 °C for 10 min, followed by quenching to room temperature after the heat treatment. The resistivity of the samples was measured by four probe method. The rectangular bar shaped samples of dimensions 2 mm × 10 mm were used for resistivity measurements. The contacts were made by silver paste and constant current of 1 mA was passed through the sample during resistivity measurements in four probe method. The superconductor phase was identified by XRD measurements, using  $CuK_{\alpha}$  radiations. The self doping of the carriers was done by carrying out postannealing of the samples in a tubular furnace in the flowing O<sub>2</sub> atmosphere at 500 °C for 5 h.

#### 3. Results and discussion

The X-ray diffraction scans of  $\text{Cu}_{0.5}\text{Tl}_{0.5}\text{Ba}_2(\text{Ca}_{2-y}\text{Mg}_y)$  ( $\text{Cu}_{0.5}\text{Zn}_{2.5}$ ) $\text{O}_{10-\delta}$  (y=0, 0.5, 1.0) superconductor, 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. Mg un-doped samples have a & c-axes lengths, around 4.37 and 14.82 Å, respectively. The cell parameters of Zn doped  $\text{Cu}_{0.5}\text{Tl}_{0.5}\text{Ba}_2\text{Ca}_{2-y}(\text{Cu}_{0.5}\text{Zn}_{2.5})\text{O}_{10-\delta}$  samples are larger than the one, we observed in the identical batch of samples synthesized at 860 °C. Moreover, the length of c-axis in  $\text{Cu}_{0.5}\text{Tl}_{0.5}\text{Ba}_2(\text{Ca}_{2-y}\text{Mg}_y)(\text{Cu}_{0.5}\text{Zn}_{2.5})\text{O}_{10-\delta}$  is much larger than the samples prepared at 860 °C. In previous studies, we have observed higher Tl contents in 850 °C synthesized samples [23] and are owed to the higher Tl contents in the final compound in 850 °C



**Fig. 1.** X-ray diffraction pattern of  $Cu_{0.5}Tl_{0.5}Ba_2(Ca_{2-y}Mg_y)(Cu_{0.5}Zn_{2.5})O_{10-\delta}(y=0,0.5,1.0)$  superconductor samples.

synthesized samples in the present studies also [23]. However, the cell parameters decrease with increasing Mg-doping. Mg-doped Cu $_{0.5}$ Tl $_{0.5}$ Ba $_{2}$ (Ca $_{2-y}$ Mg $_{y}$ )(Cu $_{0.5}$ Zn $_{2.5}$ )O $_{10-\delta}$  samples have shown cell parameters around 4.38 and 14.81 Å for y=0.5 and 4.22 and 14.77 Å for y=1.0.

The resistivity versus temperature measurements of asprepared  $Cu_{0.5}Tl_{0.5}Ba_2(Ca_{2-y}Mg_y)(Cu_{0.5}Zn_{2.5})O_{10-\delta}$  (y=0, 0.5, 1.0) samples are shown in Fig. 2(a). For all these samples, the variation of resistivity as a function of temperature is metallic from room temperature down to the onset of superconductivity. These samples have shown onset of the superconductivity around 110, 114 and 106 K and the  $T_c(R=0)$  at 92.4, 96.5 and 91 K in the samples with Mg doping of y=0, 0.5 and 1.0, respectively. These samples have shown onset of diamagnetism around 133, 118.4 and 108.4 K, respectively, shown in Fig. 3(a). The magnitude of diamagnetism systematically decreases in the samples with y=1.0. Oxygen post-annealed samples have shown onset of resistivity around 104.2, 104.1 and 107.4 K and the  $T_c(R=0)$ around 87, 89.21 and 94.44 K for Mg doping of y=0, 0.5 and 1.0, respectively, as shown in Fig. 2(b). The onset of diamagnetism in these samples is observed around 122, 114.9 and 120.8 K, respectively, as shown in Fig. 3(b). The magnitude of diamagnetism is decreased for y=0.5 sample.

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