Anomalous suppression of T_c in an overdoped region of $TlBa_2Ca_2Cu_3O_{9-\delta}$

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Abstract

Anomalous suppression of T_c has been found in an overdoped region of $TlBa_2Ca_2Cu_3O_{9-\delta}$ superconductors. A heavily overdoped sample ($T_c \simeq 100$ K) was synthesized under high pressure. The T_c was measured with decreasing the oxygen content until the sample becomes optimally doped state with a $T_c \simeq 131$ K. A dip of T_c was observed before the T_c reaches to an optimal value i.e. in an overdoped region. This behavior is quite strange because T_c is known to change monotonically in an overdoped region. Some possible explanations for this anomalous behavior will be presented.

Key words: $TlBa_2Ca_2Cu_3O_{9-\delta}$ (Tl-1223); overdoped state; suppression of superconductivity; stripe order; multi-layerd superconductor; Tl valence change

1. Introduction

$TlBa_2Ca_2Cu_3O_{9-\delta}$ (Tl-1223) have been found to have a comparable T_c ($\simeq 133.5$ K) with $HgBa_2Ca_2Cu_3O_{9-\delta}$ which is known to have the highest T_c in superconductors [1][2]. Samples are synthesized under high pressure. An intrinsic T_c is brought out by suppression of disorder due to Tl substitution for Ba- and Ca-site which inherently occurs in samples prepared by a conventional method. An as-synthesized sample is located in a heavily overdoped region with a $T_c \simeq 100$ K. The T_c is optimized by post-annealing in reducing atmosphere. When we were studying the annealing effect of T_c in detail, we noticed that the T_c did not monotonically increase from 100 K to an optimal value. In this paper, we will show this strange behavior of T_c and some possible candidates to explain it.

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2. Experimental

A sample was prepared under high pressure. Details of the sample preparation are written in elsewhere [1][3]. In order to reduce oxygen content, the sample was post-annealed for 6 h or 12 h (hereafter refer to as sample(6h) or sample(12h), respectively) in a N_2 flow (500 ml/min) with a tube furnace (26 mm in diameter). The annealing temperature (T_a) was increased step-by-step from 200 to 700 °C. Therefore, the oxygen content of the sample was gradually and monotonically reduced by the annealing. T_c was measured after the each annealing followed by annealing at higher temperature than previous one. The magnetic susceptibility was measured with decreasing temperature in a magnetic field of 20 Oe (field cooled (FC)) with a SQUID magnetometer (Quantum Design, MPMS).

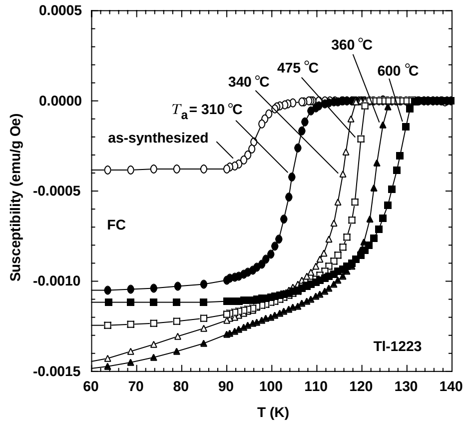

Fig. 1. Temperature dependence of susceptibility for the sample(12h) as a function of annealing temperature T_a.

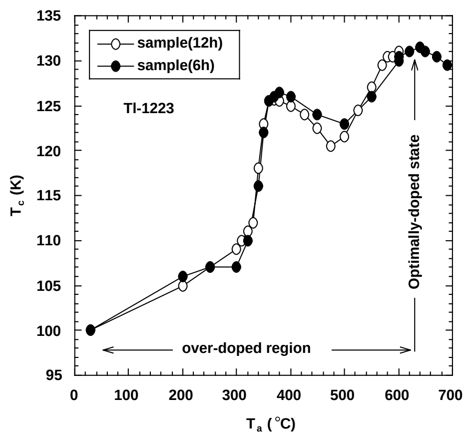

Fig. 2. Annealing temperature T_a dependence of T_c for the sample(6h) and sample(12h).

3. Results and discussion

Figure 1 indicates the selected temperature dependence of susceptibility for the sample(12h) as a function of T_a. Superconducting transitions were sharp enough to determine onset T_c uniquely. For example, the T_c of the samples of T_a = 310, 340, 360, 475 and 600 °C shown in Figure 1 were 110.0, 118.0, 125.5, 120.5 and 131.0 K, respectively. Note that the T_c of sample (T_a= 475 °C) is lower than those of sample (T_a= 360 and 600 °C). Figure 2 shows the T_a dependence of the T_c for the sample(6h) and sample(12h). One can see two T_c peaks i.e. a dip of T_c in Figure 2. The T_c increases with T_a up to about 370 °C. Then, the T_c surprisingly decreases with increasing T_a up to about 480 °C and increases again with T_a. Finally, the sample reaches or passes the optimally doped state with T_c =131.5 K. The steep increase of T_c around $T_a \simeq 320$ °C will correspond to rapid decrease of oxygen content (hole density) that was shown by thermogravimetric analysis [2].

The result strongly indicates that there exists suppression of superconductivity in an overdoped region of Tl-1223. As far as we know Tl-1223 (tetragonal unit cell) has no structural phase transition, the anomaly does not correspond to it. This phenomenon can not be explained by the conventional framework because T_c is known to change monotonically in an overdoped region. Similar behavior was found in an overdoped La-214 sample, which is explained by stripe order [4]. In case of La-214, however, the suppression of T_c is enhanced by Zn doping to Cu site which pins stripe order. It would be difficult to think that such stripe order suppresses the T_c in Tl-1223 because CuO_2 planes of the non-doped Tl-1223 must not have enough disorder to pin it. Reentrant of hole density accompanied with Tl valence change (from Tl^{3+} to $Tl^{(3-\delta)+}$) i.e. hole transfer from Tl-O planes to CuO_2 ones, which was observed in Tl-rich (Cu,Tl)-1223 by X-ray photo emission spectroscopy (XPS) [2][5], is a possible explanation for the anomaly of Tl-1223. The dip of T_c could be also explained by considering inhomogeneous carrier distribution between crystallography different CuO_2 planes in Tl-1223 [6]. Each CuO_2 plane must have a bell-shaped hole density dependence of T_c. When the hole density changes over the two peaks, a dip of T_c would be observed. At the present moment, we can not declare the mechanism of this anomaly. Further experiments such as doping effect or microscopic measurements must be performed to understand it. We also try to find the anomaly in the other system. If it is an essential phenomenon in high-T_c cuprates, the result will give an important information for the high-T_c superconducting mechanism.

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