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Normal state pseudo-gap in c-axis oriented $NdBa_2Cu_3O_x$ thin films

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Abstract. The pseudo-gap temperatures (T_g) in the normal state for c-axis oriented $NdBa_2Cu_3O_x$ thin films are investigated by dc four-probe measurements. With reduction of the oxygen content, T_c decreases, and T_g increases accordingly. The T_g–T_c relation suggests a strong correlation between superconductivity and the normal state pseudo-gap. An obvious jump was observed on the plot of T_g against T_c, which corresponds to transition from the 90 K to 60 K superconducting phase. The under-doped electronic diagram of $NdBa_2Cu_3O_x$ is constructed. This under-doped diagram qualitatively agrees with the mean field phase diagram.

1. Introduction

The universal electronic phase diagram of cuprate superconductors highlights the uniqueness of high-T_c cuprates compared to any other known compounds [1]. One of the most interesting features of the phase diagram is the existence of the normal state pseudo-gap in the under-doped region. In the over-doped region, the exponent α of $\rho_{ab}(T) \propto T^\alpha$ changes from $\alpha = 1$ to 2, suggesting a Fermi liquid behaviour. In the under-doped region, $\rho_{ab}(T)$ exhibits a linear behaviour at high temperature, and deviates from the linear dependence at low temperature. There is strong evidence for the normal state pseudo-gap in the under-doped region. The pseudo-gap was observed from magnetic susceptibility [2], specific heat [3], angle-resolved photoemission (ARPES) [4], tunnelling [5] and some kinetic measurements [6]. The temperature below which $\rho_{ab}(T)$ apparently deviates from the T-linear behaviour was thought to correspond the opening of the pseudo-gap. It is becoming clear that the superconducting gap emerges from the normal state spin gap. The d-wave nature of the order parameter holds for both the superconducting gap and the pseudogap [7]. The unusual normal state transport properties of the high-T_c cuprates have been suspected of giving clues to the basic mechanism responsible for superconductivity. T_g increases with the reduction of the doping level of an under-doped $YBa_2Cu_3O_{7-\delta}$ [8] and $YBa_2Cu_4O_8$ [9], which qualitatively agrees well with the mean field phase diagram proposed by Nagaosa and Lee [10]. Francois *et al* studied the effects of Pr doping and oxygen deficiency on T_g [11]. They concluded that T_g was completely determined by the carrier density. Liu and Guan [12] studied the effect of Pr doping on T_g in $RBa_2Cu_3O_{7-\delta}$ by using bulk samples. They found that Pr doping not only reduced the carrier density, but also simultaneously suppressed T_g. In fact, the details of T_g are not very clear at the moment. So study on different systems is helpful to understand the basic

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mechanism of T_g. $NdBa_2Cu_3O_{7-\delta}$ has the same crystal structure as $YBa_2Cu_3O_{7-\delta}$. It has the highest T_c of all the $RBa_2Cu_3O_{7-\delta}$ family. A parabolic relation between T_c and c-axis lattice parameter has been reported [13] This relation reflects the superconducting diagram of high-T_c superconductors. To our best knowledge, the relation between the pseudo-gap temperature and the oxygen deficiency for this system has not been investigated until now. In this paper, we present an under-doped electronic diagram of the $NdBa_2Cu_3O_{7-\delta}$ superconductor.

2. Experiment

c-axis oriented $NdBa_2Cu_3O_{7-\delta}$ thin films were used for this study. The thin films were deposited on (100) $SrTiO_3$ (STO) substrates by an off-axis rf sputtering technique [14]. Under the optimized deposition conditions, thin films with $T_{c0} \sim 90$ K are obtained reproducibly [13]. We used two methods to obtain different oxygen depletions. First, thin films with different oxygen deficiencies were obtained by *in situ* growing. Different oxygen deficiencies of the grown films were obtained by slightly changing the substrate temperature, the ratio of P_{Ar}/P_{O_2} and the annealing process. Second, one grown thin film with $T_{c0} = 90$ K was treated by subsequent post vacuum annealing. The chosen film was stuck on the heater by silver paste, and put it into the evacuated chamber ($\sim 10^{-2}$ mbar). The heater was heated to 100–350 °C and held for 10–20 minutes, and then cooled down to room temperature. After this vacuum treatment, some oxygen diffused out of the film. The oxygen depletion depends on the temperature and time of the post vacuum annealing. Typically, the film thickness is $\sim$ 1500 Å.

The quality of the thin film was examined by $\theta/2\theta$ and ω scanning performed on the Siemens D5000 x-ray diffractometer. The temperature dependence of resistance was measured by a standard DC four-probe method using a closed-cycled cryogenerator with a measuring current of 0.1 mA. A Pt resistance thermometer was used to measure the temperature of the sample. The cooling rate was well controlled to be 1–2 K $\min^{-1}$ by a computer program.

3. Results and discussion

XRD patterns indicate that all obtained thin films are well c-axis oriented. The rocking curves of the (005) peaks of the thin films are 0.2–0.3°, indicating good crystallinity of thin films. The c-axis lattice parameter shows strong correlation to the oxygen depletion [13]. Oxygen depletion of the thin film results in the reduction of T_c, but the transition width (ΔT_c(90%–10%)) is always very small, usually smaller than 2 K. That again suggests good crystallinity of the depleted film. Thus, we assume that the resistance behaviours only depend on the oxygen content of the thin films.

For an optimal c-axis oriented $NdBa_2Cu_3O_{7-\delta}$ thin film, the resistance behaves with a linear character above $\sim$100 K. The ratio of the resistance at room temperature (R_{300}) to the resistance at 100 K (R_{100}) is about 3. In the T-linear region, $\rho_{ab}(T) = (4\pi/\omega_{pD}^2)\tau^{-1}$, where τ^{-1} is the scattering rate and $\tau^{-1} = 2\pi\lambda T$; $\omega_{pD}^2 \sim n/m^*$, n is the density of the carrier and m^* the effective mass. The T-linear dependence of resistivity was also observed in La–Sr–Cu–O and Y–Ba–Cu–O. At the optimal doping level, the in-plane resistivity $\rho_{ab}(T)$ is linear in temperature over a wide temperature range from just above T_c to 1000 K for $La_{1.825}Sr_{0.175}CuO_{4-\delta}$ [15] and to 600 K for $YBa_2Cu_3O_{7-\delta}$ [16]. For an oxygen-depleted sample $\rho_{ab}(T)$ deviates from T-linear behaviour below a temperature well above T_c. Takenaka *et al* [16] investigated the anisotropic resistivity of a detwinned $YBa_2Cu_3O_{7-\delta}$ crystal. The out of plane resistivity ρ_c shows a crossover from high-T metallic to low-T semiconducting

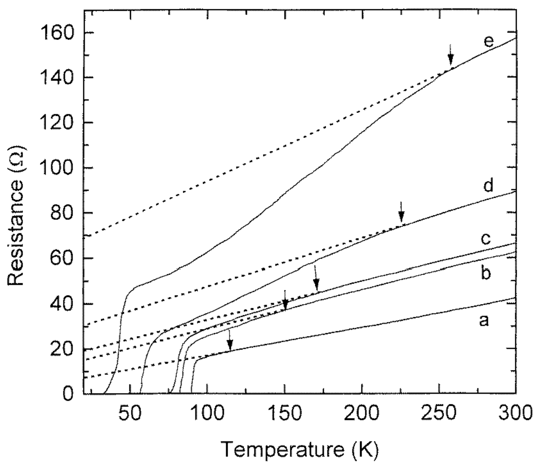

Figure 1. Several curves of the resistance against the temperature of the subsequent post vacuum annealed *c*-axis oriented $NdBa_2Cu_3O_x$ thin films. The dotted line guides the linear dependence of the resistance. The arrow indicates the pseudo-gap temperature. Symbol a indicates the as-deposited film with $T_c = 90$ K; symbols b, c, d and e indicate the order of post vacuum annealing.

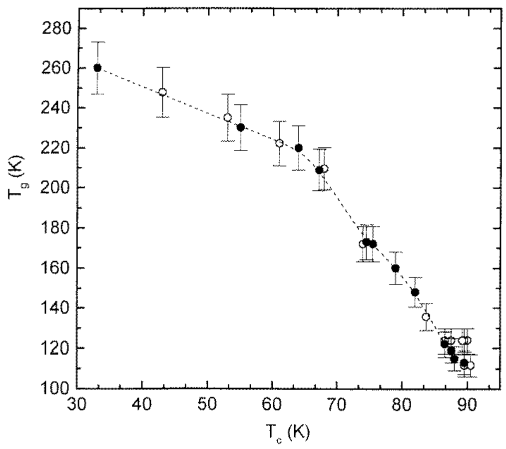

Figure 2. The plot of T_g against T_c from different thin films denoted by the closed symbols and from the same film treated by subsequent post vacuum annealing denoted by the open symbols. The dotted line guides the eyes.

behaviour, while the in-plane resistivity ρ_{ab} deviates from T-linear dependence. The crossover in ρ_c is linked with the onset of nonlinearity in ρ_{ab}. They suggested that the crossover temperature be associated with the pseudo-gap temperature T_g. Recently, the effect of oxygen depletion and Pr doping on T_g in $YBa_2Cu_3O_{7-\delta}$ have been studied by Francois *et al* [11] and Liu and Guan [12]. In this work, the temperature at which the resistivity starts to deviate from T-linear dependence defined T_g, the opening of the pseudo-gap. Figure 1 shows the curves of the resistance against the temperature of the post vacuum annealed thin films. At high temperature, the resistance decreases with temperature linearly. Below a certain temperature, the resistance starts to deviate from linear variation. The temperature at which the resistivity starts to deviate from T-linear dependence corresponds to the opening of the pseudo-gap. It is

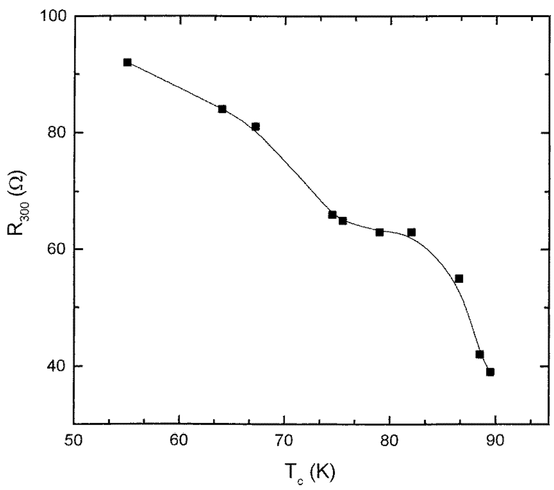

Figure 3. The variation of the room temperature resistance with the superconducting transition temperature for the same film treated by subsequent post vacuum annealing.

found that T_c decreases, but T_g increases accordingly, with the reduction of oxygen content. In figure 2, we plot the T_g against T_c from different thin films denoted by the closed symbols and from the same film treated by subsequent post vacuum annealing denoted by the open symbols. The deviation of T_g was set to be $\pm 5\%$ of the T_g value. It is found that two sets of data agree very well. From figure 2, we can find a jump at about 70 K. That must be related to the transition from 90 K to 60 K superconducting phases. This transition indicated on the T_g–T_c plot seems also to exist in the system of $SmBa_2Cu_3O_{7-\delta}$ [12], but has never been mentioned in the literature, to our best knowledge. As is well known, 90 K and 60 K superconducting phases generally exist in the R123 system. For different rare earth elements, the 90 K and 60 K superconducting phases are formed in different regions of oxygen content due to the size effect [17]. For $YBa_2Cu_3O_{7-\delta}$ and $NdBa_2Cu_3O_{7-\delta}$, the phase transition from 90 K to 60 K superconducting phase occurs at about $\delta = 0.35$ and 0.25, respectively [18, 19]. It was thought that the 60 K superconducting phase was derived from the 90 K phase by oxygen ordering. Oxygen ordering results in hole localization, and thus reduces the density of carrier. Lower density of carrier corresponds to higher pseudo-gap temperatures (T_g). Thus the jump in the T_g–T_c plot clearly indicates the phase transition from 90 K to 60 K superconducting phase. The room temperature resistance of the thin film is plotted against T_c in figure 3. The transition from 90 K to 60 K superconducting phase was also clearly indicated. Based on the equation of $\rho_{ab}(T) = (4\pi/\omega_{pD}^2)\tau^{-1}$, we have $\rho_{ab}(T) \propto 1/n$, n is the density of the carrier. The lower the density of the carrier, the higher the resistivity is. The 60 K superconducting phase has higher resistivity than the 90 K phase. Thus figure 2 and 3 reflect the same physical nature. The T_g–T_c relation suggests a strong correlation between superconductivity and the normal state pseudo-gap.

The oxygen content was considered to be a key factor in the superconducting temperature of high-T_c cuprates. For a bulk sample, the influence of the oxygen content on superconductivity was extensively studied. A nearly linear relation between the c-axis parameter and the oxygen content was reported in both $YBa_2Cu_3O_x$ and $NdBa_2Cu_3O_x$ [18, 19]. For thin films, however, it is very hard to determine the oxygen content precisely. Thus, little work has been reported on the oxygen content in thin films. The linear relation between

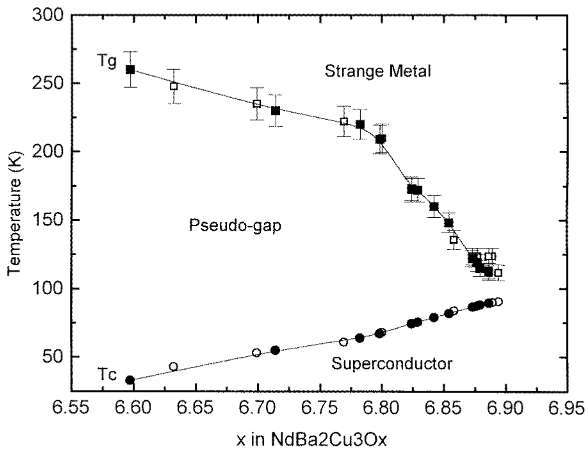

Figure 4. The under-doped electronic diagram of $NdBa_2Cu_3O_x$ on the scale of the oxygen content x. The closed symbols represent the data from different thin films, and the open symbols from the same film treated by subsequent post vacuum annealing. The lines guide the eyes.

c-axis lattice parameter and the oxygen content for bulk samples was used to scale the oxygen content for $NdBa_2Cu_3O_x$ thin films [13]. A parabolic relation between T_c and c-axis lattice parameter has been obtained. This relation reflects the general superconducting diagram of high-T_c superconductors. In this work, the reported T_c–δ relation [19] was used to deduce the oxygen contents of our samples. Then the under-doped electronic diagram of $NdBa_2Cu_3O_x$ was constructed on the scale of the oxygen content x (see figure 4), which qualitatively agrees well with the mean field phase diagram.

4. Conclusion

The pseudo-gap temperatures (T_g) in the normal state for c-axis oriented $NdBa_2Cu_3O_x$ thin films are investigated by dc four-probe measurements. With reduction of the oxygen content, T_g increases and T_c decreases. An obvious jump was observed on the plot of T_g against T_c, which corresponds to transition from the 90 K to 60 K superconducting phase. The T_g-T_c relation suggests a strong correlation between superconductivity and the normal state pseudo-gap. The under-doped electronic diagram of $NdBa_2Cu_3O_x$ has been constructed.

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