Transformation of in-plane $\rho(T)$ in $YBa_2Cu_3O_{7-\delta}$ at fixed oxygen content

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This paper reveals the origin of variation in the magnitude and temperature dependence of the normal state resistivity frequently observed in different YBCO single crystal or thin film samples with the same $T_c$. We investigated temperature dependence of resistivity in $YBa_2Cu_3O_{7-\delta}$ thin films with $7-\delta=6.95$ and 6.90, which were subjected to annealing in argon at 400-420 K ($120-140^\circ$C). Before annealing these films exhibited a non-linear $\rho_0(T)$, with a flattening below 230 K, similar to $\rho_0(T)$ and $\rho_{ab}(T)$ observed in untwinned and twinned YBCO crystals, respectively. For all films the annealing causes an increase of resistivity and a transformation of $\rho_{ab}(T)$ from a non-linear dependence towards a more linear one (less flattening). In films with 7-\delta = 6.90 the increase of resistivity is also associated with an increase in $T_c$. We proposed the model that provides an explanation of these phenomena in terms of thermally activated redistribution of residual O(5) oxygens in the chain-layer of YBCO. Good agreement between the experimental data for $\rho_{ab}(t,T)$, where t is the annealing time, and numerical calculations was obtained.

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1. INTRODUCTION

Right after discovery of high temperature superconductivity, it has been observed that temperature dependence of the normal state resistivity $\rho(T)$ is linear in many samples over a wide range of temperatures. Phillips explained this result (believed to be universal for all optimally doped HTSC) using two-carrier percolation model. Later, the observation of a linear $\rho(T)$ has been also used to support the argument that non-Fermi liquid states are responsible for the normal state properties of HTSC.

The analysis of the experimental data for $\rho(T)$ obtained on untwinned and twinned YBCO single crystals and YBCO thin films revealed however that the pure linear temperature dependence of resistivity is not the characteristic of $\rho(T)$ in HTSC. In fact, it has been found that $\rho_0(T)$ measured along chain direction in untwinned crystals of YBCO exhibits a non-linear behavior with a flattening at temperatures below 220K [see Fig.1(a)]. On the other hand, $\rho_a(T)$ measured in a direction perpendicular to the chains, increases linearly with an increasing temperature over a wide temperature range. The magnitude of $\rho_a(T)$ is about 2-2.5 times larger than that of $\rho_0(T)$ at corresponding temperatures. An exception is the result of Welp et al., who observed a linear temperature dependence of both $\rho_a$ and $\rho_0$. However, the absolute values of these resistivities are about 2 times higher than those reported elsewhere for untwinned YBCO crystals.

The in-plane resistivity $\rho_{ab}(T)$ measured in twinned YBCO crystals of $T_c = 93K$, $\Delta T_c = 0.2K$, and of low resistivity $\rho_{ab}(300K) = 130\mu\Omega cm$ displays non-linear temperature dependence similar to that observed in the most of untwinned YBCO crystals along the chain direction [see Fig.1(b)]. Surprisingly, other YBCO twinned crystals of the same $T_c$ and $\Delta T_c$, and higher room temperature resistivity $\rho_{ab}(300K) = 160\mu\Omega cm$ are characterized by $\rho_{ab}$ with a linear dependence on temperature [Fig.1(b)]. This value of resistivity is close to that of a non-linear $\rho_0(T)$ measured at 300 K along the chain direction in some untwinned YBCO crystals.

Temperature dependence of $\rho_a$, measured in c-axis oriented epitaxial YBCO thin films shows features similar to those observed in twinned YBCO crystals. Optimally doped films with resistivity as low as 150$\mu\Omega cm$ at 300K have non-linear $\rho_{ab}(T)$ with a flattening at temperatures below 230K (see Fig.2). However, it appears that linear temperature dependence of $\rho_{ab}(T)$ characterizes films with higher resistivity (above approximately 250$\mu\Omega cm$ at 300 K). Underdoped films with $T_c \approx 84 - 85K$ exhibit $\rho_{ab}(T)$ similar to that observed in optimally doped films (see Fig.2).

Two different theoretical approaches were used so far to explain the observed non-linear $\rho(T)$. The first one is based on classical electron-phonon Bloch-Gr"uneisen (BG) theory of metallic conductivity. It predicts a non-linear $\rho(T)$ for temperatures less than characteristic temperature $T^* = \eta \Theta_D$ (where $\Theta_D$ is the Debye temperature and $\eta \sim 1 - 2$), and a linear $\rho(T)$ for temperatures above $T^*$. The other one employs a thermal Frenkel disorder of long range-ordered chains of oxygen, a process during which an oxygen atom jumps from its O(1) chain site to a nearby O(5) interstitial (inter-chain) vacant site.

Poppe et al. attempted to explain the non-linear $\rho(T)$ observed in YBCO thin films by assuming that the resistivity is the sum of the temperature independent residual resistivity $\rho_r$, due to impurities or defects, and the Bloch-Gr"uneisen resistivity $\rho_{BG}(T)$. They found that BG theory can be applied only if one assumes very high values of Debye temperature within the range of 650-950K.
FIG. 1: (a) Temperature dependence of resistivities, $\rho_b(T)$ and $\rho_a(T)$, measured in untwinned YBCO crystals along the chain direction (b-direction) and along the a-direction perpendicular to the chains, respectively. Note a flattening of $\rho_b(T)$ below 220 K and a linearity of $\rho_a(T)$ over a wide range of temperatures for all three samples.\textsuperscript{4,5,6} (b) Temperature dependence of the in-plane resistivity $\rho_{ab}(T)$ for twinned YBCO crystals. Note that in this case $\rho_{ab}(T)$ exhibits either a non-linear temperature dependence (with flattening) below 220 K or a linear dependence in the sample with higher resistivity.\textsuperscript{8}

Martin et al.\textsuperscript{12} were able to obtain a good theoretical fit to a non-linear $\rho(T)$ measured in YBCO(124) films at temperatures up to 600K, using BG theory and an effective transport Debye temperature $\Theta_D^* = \frac{2k_F}{G}\eta\Theta_D$ of 500K. They argued that $\Theta_D^*$ can be substantially smaller than $T^* = \eta\Theta_D$ if the Fermi vector $k_F$ is sufficiently smaller than the reciprocal lattice vector G.

Goldschmidt et al.\textsuperscript{11} interpreted the non-linear temperature dependence of resistivity in fully oxygenated polycrystalline YBCO in terms of a thermally activated process of Frenkel pair formation, during which the chain oxygen jumps from O(1) site to a nearby O(5) site. According to Goldschmidt et al. this process accounts for an excess electron scattering with an increasing temperature. Each Frenkel pair creates an intrinsic defect resistivity $\rho_d$ which represents an excess resistivity connected in series to the perfect chain resistivity. The upturn in resistivity that occurs at temperatures above 200K, is explained as due to an increase of Frenkel pair density with an increasing temperature. The excess resistivity is defined as: $\rho_{\text{excess}} = \rho_{\text{chain}} - (\rho_a + aT) = \rho_d C_d(T)$, where $C_d(T)$ is the temperature dependent fractional defect pair occupancy. Temperature dependence of $C_d$ was taken as that for a two level system with the energy barrier $E_d = \exp(-E_d/kT)$, where $E_d$ is the energy barrier between the final O(5) and initial O(1) oxygen sites. Consequently $\rho_{\text{excess}} \simeq \rho_d \exp(-E_d/kT)$. Goldschmidt et al. obtained good theoretical fit to $\rho(T)$ data measured for a granular YBCO taking the activation energy $E_d \simeq 120\text{meV}$ and defect resistivity $\rho_d \simeq 2\text{m}\Omega\text{cm}$. Measurements performed on untwinned YBCO crystals by Gagnon et al.\textsuperscript{6} revealed however that temperature dependence of the excess resistivity above 300K is linear, not at all exponential.

FIG. 2: Temperature dependence of the in-plane resistivity $\rho_{ab}$ measured in c-axis oriented epitaxial YBCO thin films. The data indicate that films with low resistivity exhibit a deviation from the linear temperature dependence at temperatures below approximately 230K.\textsuperscript{9,10}

The experimental results obtained on untwinned and twinned YBCO crystals, and YBCO thin films could be summarized as follows: Although $\rho(T)$ for the most of untwinned crystals show a non-linear $\rho_b(T)$ and a linear $\rho_a(T)$, with the magnitude of $\rho_a(T)$ being higher than that of $\rho_b(T)$, these two forms of $\rho(T)$ have been also randomly observed in twinned YBCO crystals and thin films. Surprisingly, the absolute values of resistivity of
twinned YBCO crystals and films are often lower than those measured in some untwinned YBCO crystals along the b-direction. Also, there is some correlation between the magnitude of resistivity and a specific form of \( \rho(T) \) (non-linear or linear) in twinned YBCO crystals and in YBCO films. In these cases the resistivities of a linear \( \rho(T) \) are in general higher than the corresponding ones for a non-linear \( \rho(T) \). In untwinned YBCO crystals, a non-linear and a linear \( \rho(T) \) has been attributed to the chain and plane conductivities, respectively. Therefore, the random appearance of a non-linear and linear \( \rho(T) \) in twinned YBCO crystals and films is surprising and should be investigated.

In this paper we provide alternative explanation of the presence of a non-linear and a linear dependence of resistivity \( \rho_{ab} \) on temperature in YBCO. Our studies were focused on YBCO c-axis oriented thin films, both optimally doped \((7 - \delta \approx 6.95, T_c \approx 90 - 91K)\) and slightly underdoped \((7 - \delta \approx 6.90, T_c \approx 85K)\), with a non-linear \( \rho_{ab}(T) \). We investigated the effect of oxygen ordering (redistribution) on \( \rho_{ab}(T) \) in those films. The applied experimental procedure included systematic annealing of the samples in argon at 400-420K \((120 - 140^\circ \text{C})\), followed by the measurements of \( \rho_{ab}(T) \) at temperatures between \( T_c \) and 300K.

The results revealed a transformation of \( \rho_{ab}(T) \) from a non-linear temperature dependence towards a more linear one (less flattening). This transformation is accompanied by an increase in the absolute value of resistivity. In underdoped samples, annealing causes also an increase in \( T_c \). We present possible explanation of this phenomenon, which is based on thermally activated redistribution of oxygen in the chain-layer of YBCO.

2. EXPERIMENTAL PROCEDURE

We investigated c-axis oriented YBCO thin films, which were deposited on different substrates: \( \text{SrTiO}_3, \text{LaAlO}_3 \) and sapphire (with \( \text{CeO}_2 \) buffer layer) using the standard off-axis rf magnetron and laser ablation techniques. The standard photolithography was used to produce \( 60 \mu \text{m} \) wide and 6.4 mm long thin film bridges with four measurement probes. The distance between the voltage probes was 0.4 mm. Silver contacts were sputter-deposited using rf-magnetron technique. The samples were annealed in argon at temperatures 120 – 140°C several times for a total time between 8 to 24 hours. The measurements of \( \rho_{ab}(T) \) (in the a-b planes of the films) at temperatures between \( T_c \) and 300K, were conducted after each 2-5 hour annealing period. During these measurements a dc current of 10\( \mu \text{A} \) was applied to the sample in the form of short pulses (duration less than 200ms) in order to reduce Joule’s heating. Current reversal was used to eliminate the effects of thermal emf in the leads. The voltage was measured using Keithley 2182 nanovoltmeter connected to Keithley 236 current source, with the nanovoltmeter working as a triggering unit for the current source. The nanovoltmeter operated in a “Delta” mode which allows one to perform very fast multiple measurements of the voltage from two voltage measurements for two opposite directions of the current. Temperature was monitored by a carbon-glass thermometer and stabilized (using the inductance-less heater and a temperature controller) within \( \pm 10mK \).

3. EXPERIMENTAL RESULTS

The temperature dependence of resistivity \( \rho_{ab}(T) \) measured for slightly underdoped YBCO films with \( T_c(R=0)= 85-86K \), is shown in Figures 3(a) and 4(a) as a function of annealing time in argon at 120 – 140°C. The \( \rho_{ab}(T) \) for unannealed samples exhibits a flattening [an upward deviation from a linear temperature dependence; see straight solid line in Figures 3(a) and 4(a)] at temperatures between \( T_c \) and approximately 220-230K. The annealing causes an increase of \( T_c \) [see Figures 3(b) and 4(b)], an increase of resistivity, and a transformation from a non-linear temperature dependence of resistivity towards the linear one. The increase of \( T_c \) depends on the sample i.e. it is about 1K after 8 hour annealing at 120°C for film 12, but only 0.5K after 24 hour annealing at 140°C for film 13. The temperature dependence of resistivity \( \rho_{ab}(T) \) measured for optimally doped YBCO film with \( T_c(R=0) \) about 90K is shown in Fig 5(a) as a function of annealing time in argon at 120°C. The annealing of this sample leads to an increase of the magnitude of \( \rho_{ab}(T) \) and a reduction of the flattening between \( T_c \) and 230-240K. However, no change of \( T_c \) was observed in this case [see Fig 5(b)]. Temperature dependence of \( \rho_{ab}(T) \) at temperatures above approximately 220K is linear (see solid lines in Figures 3 and 4). Deviation from the linear dependence at temperatures below 220K is shown in Fig 6 as a function of temperature at different annealing times. It is defined in the following form: \( \rho_{ab}(T) - \rho_{linear}(T)/\rho_{ab}(T1) \), where \( T1 \) is the temperature at which a maximum upward deviation of \( \rho_{ab}(T) \) from \( \rho_{linear}(T) \) occurs. This deviation initially decreases with an increasing annealing time, however it reaches saturation at longer annealing times. The magnitude of resistivity \( \rho_{ab}(T) \) of all samples studied was found to increase gradually with an annealing time [see Figures 3, 4 and 5].

4. DISCUSSION

According to Jorgensen annealing of YBCO at 120 – 140°C causes redistribution of oxygen in the chain-layer, without a change in the overall concentration of oxygen in the sample. Low temperature annealing was performed previously by Jorgensen et al and Shaked et al. They carried out room temperature annealing of sintered samples of underdoped YBCO (with \( 7 - \delta = 6.41 \) and 6.25 ) which were quenched first from a temperature of...
FIG. 3: (a) Temperature dependence of the in-plane resistivity $\rho_{ab}$ measured in a c-axis oriented slightly underdoped YBCO thin film 12 with $T_c = 85.4K$, before and after annealing for 2 and 8 hours in argon at 120°C. Note a systematic increase of resistivity and a reduction of a deviation from a linear temperature dependence with an increasing annealing time. (b) Dependence of resistivity on temperature close to $T_c$ before and after annealings. Note a systematic increase of $T_c$ with an increasing annealing time.

FIG. 4: (a) Temperature dependence of the in-plane resistivity $\rho_{ab}$ measured in a c-axis oriented slightly underdoped YBCO thin film 13 with $T_c = 85.4K$, before and after annealing for 15 and 20 hours in argon at 140°C. Note a systematic increase of resistivity and a reduction of a deviation from a linear temperature dependence with an increasing annealing time. (b) Dependence of resistivity on temperature close to $T_c$ before and after annealings. Note a systematic increase of $T_c$ with an increasing annealing time.

100 - 150°C down to a liquid helium temperature. Room temperature annealing led to a dramatic change of the transport and structural properties of this compound i.e., an increase of $T_c$ and a decrease of lattice parameters with annealing time. This behavior was explained in terms of a local ordering of oxygen atoms at room temperature, which causes formation of oxygen supercells (alternating partial and full chains) in the chain layer. Veal et al. reported the observation of time-dependent behavior at room temperature in the superconducting and structural properties of $Y Ba_2Cu_3O_x$ single crystals, with stoichiometries controlled within the range $6.3 < x < 6.6$ by quenching the samples from 520°C down to a liquid nitrogen temperature. They proposed two explanations of this behavior. One interpretation was that oxygens, quenched into O(5) sites, move to neighboring O(1) chain sites thus enhancing the order in the O(5) oxygen vacancy array in the chain layer. The other one suggested that the ordering associated with low temperature annealing likely involves the formation of alternating full and empty (oxygen free) chains in the chain layer. Andersen et al. investigated ordering kinetics of ortho-superstructures in
FIG. 5: (a) Temperature dependence of the in-plane resistivity $\rho_{ab}$ measured in a c-axis oriented optimally doped YBCO thin film with $T_c = 90.4K$ before and after annealing for 9 hours in argon at 120°C. The resistivity increases with an increasing annealing time. Deviation from a linear temperature dependence at temperatures below 230-240K decreases with an increasing annealing time (see Fig. 6 for more details). (b) Dependence of resistivity on temperature close to $T_c$ measured before and after annealing. $T_c$ was found to be independent of the annealing time.

FIG. 6: Upward deviation of $\rho_{ab}(T)$ (measured for films 11, 12 and 13) from a linear temperature dependence of resistivity $\rho(T)$ defined as $\Delta\rho(T)/\rho(T_1) = [\rho_{ab}(T) - \rho(T)]/\rho_{ab}(T_1)$, where $T_1$ is the temperature at which the maximum upward deviation of $\rho_{ab}(T)$ from a linear $\rho(T)$ occurs. Note the saturation of $\Delta\rho(T)/\rho(T_1)$ after annealing times of 6 hours for film 11, and 20 hours for film 13, respectively.

The chain layers of YBCO single crystals as a function of temperature up to about 150°C and oxygen doping level over a range between 6.3 and 6.8. They provided the structural phase diagram of the superstructure ordering in YBCO (see Fig. 5 in Ref. 17). According to this diagram, our samples with oxygen content of about 6.90 and 6.95 should have the ortho-I structure with no sign of a superstructure formation between room temperature and 150°C.

Our interpretation of the data is based on the assumption that the redistribution of oxygen during annealing at temperatures of 120 – 140°C takes place in the chain layers of YBCO. It is known that even optimally doped $RBa_2Cu_3O_x$ (RBCO) (including YBCO) contains a few percent of interchain oxygen O(5) in the chain layers. Therefore, we also assume that O(5) oxygens form links (bridges) between partially occupied chains of oxygen O(1). These links could be formed by oxygen O(5) during cooling of the films from a high deposition temperature (usually around 650 – 750°C). This allows the transport current in the chain layer to zigzag between the chains via O(5) sites, thus reducing the overall resistivity in the chain layer. For a twinned sample, the overall resistivity
\( \rho_{ab} \) is an average of \( \rho_a \) and \( \rho_b \) in the a- and b-direction, respectively, i.e., \( \rho_{ab} = (\rho_a + \rho_b)/2 \). It is also equal to the plane- and the chain-layer-resistivities connected in parallel according to the formula,

\[
\frac{1}{\rho_{ab}} = \frac{1}{\rho_{\text{plane}}} + \frac{1}{\rho_{\text{chain-layer}}} \quad (1)
\]

or

\[
\rho_{ab} = \frac{\rho_{\text{plane}}}{1 + \frac{\rho_{\text{plane}}}{\rho_{\text{chain-layer}}}} \quad (2)
\]

According to the experimental results obtained on untwinned YBCO crystals, temperature dependence of resistivity in the planes is linear i.e.,

\[
\rho_{\text{plane}}(T) = a_1 + a_2 T \quad (3)
\]

where \( a_1 \) and \( a_2 \) are constants. On the other hand, according to references, temperature dependence of the chain resistivity in untwinned YBCO crystals contains both linear and quadratic components. The latest reference, revealed however that on average the chain resistivity of four untwinned YBCO crystals follows almost pure \( T^2 \) dependence over a temperature range between 130 and 300 K, i.e.,

\[
\rho_{\text{chain}}(T) = b_1 + b_2 T^2 \quad (4)
\]

where \( b_1 \) and \( b_2 \) are constants. In equations Eq 3 and Eq 4, resistivity and temperature are in units of \( \mu \Omega \cdot \text{cm} \) and K, respectively.

\( T^2 \) dependence of resistivity was observed in quasi-1D organic conductors (see for example Weger et al.). Abrikosov and Ryzhkin developed a theory of the temperature dependence of resistivity in quasi-1D metals. According to this theory in quasi-1D metals with a long mean-free-path, electron-phonon scattering produces \( T^2 \) dependence of resistivity. Such model may be applied to the chains of YBCO, since in pure YBCO the mean-free-path should be much larger than the lattice constant. So far however no calculations of \( \rho_{\text{chain}}(T) \) have been performed using the relevant phonon frequencies for YBCO.

In an underdoped YBCO, and even in an optimally doped YBCO, the chains are not fully occupied. As mentioned above, in the presence of interchain O(5), the transport current could therefore zigzag between the chains in the chain layer via O(5) bridges along a path of the lowest resistance (see Fig 7). In this case electrical transport would still be quasi-1D and the resistivity should be proportional to \( T^2 \). One also could expect that O(5) bridges lower the overall resistance in the chain layer. The activation energy for the motion of O(5) oxygen between chains in the b-direction is very small and during annealing at relatively low temperatures of 120 – 140°C oxygen O(5) should be highly mobile in this direction. High mobility of O(5) during annealing could change the electrical properties of the chain layer. O(5) can fill some O(1) vacancies in the chains, which leads to a higher \( T_c \). On the other hand, the motion of O(5) between the chains in the chain direction could interrupt the paths of the lowest resistance, causing an overall increase of resistivity in the chain layer. One could make a counter-argument that even in the absence of interchain O(5) oxygen, annealing could cause more disorder in the chains, resulting in an increase of resistivity. This however should be accompanied by a decrease in \( T_c \).

We considered the following model that could explain an increase of resistivity with the annealing time in twinned YBCO thin films. In the presence of oxygen O(5) the electrical transport in the chain layer is determined by the resistance of the chain fragments and the interchain bridges through O(5) sites. The resistance of the interchain bridges \( R \) between any two neighboring chains could be considered as that of average N "effective" bridges (through which the current flows) connected in parallel i.e. \( R = R_o/N \), where \( R_o \) is the resistance of a single effective bridge. During annealing at 120 – 140°C in argon, due to high mobility of oxygen O(5) the number of effective bridges could be reduced. We assumed that this process is completely random and the number of effective bridge disintegrations that occurs, is proportional to the number of effective bridges present i.e.,

\[
\rho_{ab}(T) = \rho_{ab}(0) + \frac{1}{N} \sum_{i=1}^{N} \rho_i(T)
\]

where \( N \) is the number of effective bridges, and \( \rho_i(T) \) is the resistance of each effective bridge.
\[
\frac{dN}{dt} = -\alpha N
\]  
(5)

where \(\alpha\) is a decay constant. Dependence of \(N\) on time is then:

\[
N = N_0 \exp(-\alpha t)
\]  
(6)

where \(t\) is the annealing time and \(N_0\) is the number of effective bridges at \(t=0\). The resistance \(R\) then increase with time \(t\) according to the expression:

\[
R = \frac{R_0}{N} = \frac{(R_0/N_0) \exp(\alpha t)}{N}
\]  
(7)

The total average resistance offered to the current by any two chains could be written as the sum of the resistance due to chain fragments (which we consider time independent) and that due to the time dependent O(5) bridges [Eq.(4)]. Therefore, the overall resistivity in the chain layer as a function of annealing time \(t\) and temperature \(T\) can be expressed in the following form,

\[
\rho_{\text{chain-layer}}(t, T) = \frac{\rho_0(T)}{1 + C \exp(\alpha t)}
\]  
(8)

where \(\rho_0(T) = b_1 + b_2 T^2\) is the total chain layer resistivity at temperature \(T\) and annealing time \(t=0\) (which includes contributions due to both chain fragments and O(5) bridges), and \(C\) is the normalization constant which ensures that at \(t=0\) the total chain layer resistivity equals \(\rho_0(T)\).

Then the overall resistivity \(\rho_{ab}(t, T)\), including the contribution due to the planes, could be written as

\[
\rho_{ab}(t, T) = \frac{\rho_{\text{plane}}(T) + \rho_{\text{chain-layer}}(t, T)}{1 + \frac{\rho_{\text{plane}}(T)}{\rho_{\text{chain-layer}}(t, T)}}
\]  
(9)

We assumed that \(\rho_{\text{plane}}(T)\) in this equation depends on temperature according to Eq.8 and not on the annealing time, so all changes in resistivity due to annealing occur in the chain layers.

We performed computer calculations of \(\rho_{ab}(t, T)\) and compared them with the data obtained for twinned c-axis oriented YBCO films. The fits to the experimental data are shown in Figures 8 and 9. The fitting parameters are \(a_1, a_2, \alpha\), and \(C\). Constants \(a_1\) and \(a_2\) in the equation \(\rho_{\text{plane}}(T) = a_1 + a_2 T\) are shown in Table I for films 11, 12, and 13 and for untwinned YBCO crystals. The slopes of \(\rho_{\text{plane}}\) curves in Figures 8 and 9, which are represented by constants \(a_2\), were assumed to follow roughly the slope of the experimental data for \(\rho_{ab}(t, T)\) at temperatures above 250K. Constants \(b_1\) and \(b_2\) for \(\rho_{\text{chain}}(T)\) (see Eq.11) were calculated using Eq.2, Eq.3, and the experimental data for \(\rho_{ab}(t = 0, T)\) in unannealed \((t=0)\) YBCO films. Good agreement between the experimental data and the model was obtained for both optimally doped and slightly underdoped YBCO films.

According to Eq.9 the total resistivity \(\rho_{ab}(t, T)\) approaches that of the planes (which increases linearly with an increasing temperature) if the chain layer resistivity is very high (i.e., it exhibits a semi-insulating or insulating behavior). This could happen after a long annealing time \(t\) if the decay constant \(\alpha\) (which describes the process of disintegration of O(5) bridges between O(1) chains), is independent of time. On the other hand, if the chain layer resistivity is small (i.e., a metallic-like), then the chain layer contributes substantially to the total resistivity. In this case, one could expect a deviation of \(\rho_{ab}(t, T)\) from a linear temperature dependence towards a quadratic-like one, due to a \(T^2\) dependence of \(\rho_{\text{chain}}(T)\). Eq.9 shows that the change of resistivity from a small towards a large one in the chain layer (which corresponds to a transition from a metal-like to an insulating behavior), causes an increase of the total resistivity \(\rho_{ab}(T)\) and a transformation of its temperature dependence from a non-linear towards a linear one. This could explain why so many YBCO single crystal samples (or YBCO thin film samples) with the same \(T_c\) have very different magnitudes and temperature dependencies of resistivity.

| Material          | \(a_1\) | \(a_2\) | \(\alpha\) | \(C\) | Ref |
|-------------------|---------|---------|-----------|------|-----|
| YBCO film 11      | 0       | 1.58    | 0.066     | 10   | this work |
| YBCO film 12      | 5.5     | 1.12    | 0.442     | 17   | this work |
| YBCO film 13      | 5.5     | 0.92    | 0.042     | 2    | this work |
| YBCO untwinned crystal 1 | 16.5  | 1.00    |           | 4    |     |
| YBCO untwinned crystals 1-4 | 0      | 1.00    |           | 4    |     |
| YBCO untwinned crystal | -10.5 | 0.79    |           | 4    |     |
| YBCO untwinned crystal | -10.3 | 0.76    |           | 4    |     |

5. CONCLUSIONS

Our investigation of the temperature dependence of resistivity of twinned YBCO thin films as a function of annealing time at 120 – 140°C led us to conclude that the resistive properties of the chain layer (located between two CuO2 double-planes) could have a dramatic effect on the magnitude and the temperature dependence of the total resistivity \(\rho_{ab}(T)\). We suggest that the resistive properties of the chain layer could be modified by the residual amount of the interchain oxygen O(5) which allows a current to flow between the chains along a path of
FIG. 8: Experimental data and theoretical fitting curves obtained between 120 and 300K, using the model described in the text, for $\rho_{ab}(t,T)$ measured in slightly underdoped films 12 and 13. The assumed resistivity of the planes $\rho_{\text{planes}}(T)$ in Eq. (9) is represented by the solid line marked $\rho_{\text{plane}}$ at temperatures between 120 and 300K.

Recent measurements of the angular dependence of resistivity in the ab-planes close to the superconducting transition temperature revealed larger than expected changes of resistivity in some YBCO thin films, when the angle between the direction of an external magnetic field and the ab-planes was changed from 0 to 90 degrees. These large changes suggest large anisotropy (and strong pinning of magnetic flux between the CuO$_2$ planes), which could be caused by an insulating state of the chain layers in these samples. On the other hand, the changes of chain-layer resistivity may be also responsible for different temperature dependencies of c-axis resistivity observed in different YBCO single crystals. Surprisingly, the c-axis resistivity of YBCO crystals studied so far exhibits either a $1/T$ dependence or a linear temperature dependence.

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