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Metal-insulator transition induced by oxygen in nanoscale Bi:2201 thin films and in bulk Y:123 superconductiong materials

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The bulk underdoped $YBa_2(Cu_{0.96}\ Fe_{0.04})_3O_y$ superconducting system has been synthesized and investigated. The effect of oxygen content and the thermal treatment (fast quenching of underdoped samples from 250 K to 4.5K) on the electrical resistance were studied. Bi:2201 nanoscale thin films were deposited onto $SiTiO_3$ and MgO substrates by using DC magnetron sputtering. The effect of different oxygen pressure (f_{O2}) in the sputtering gas and the nature of the substrate on the electrical resistivity of thin films are presented. For both systems, in the lower temperature range, the underdoped samples show a ln(1/T) behavior of electrical resistance. The origin of this behavior is discussed.

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1. Introduction

The central feature of high temperature superconductors (HTS) is that they are doped insulators obtained by chemically adding charge carriers to a insulating antiferromagnetic (AF) state of CuO_2 planes [1]. It has been well established that already small concentrations of hole doping in CuO_2 planes reduces rapidly the AF ordering temperature. There is a strong tendency for an AF to expel holes, which lead to phase separation into a hole rich and hole -poor phase [2]. In $YBa_2Cu_3O_{7-y}$ the decrease of hole concentration by increasing oxygen deficiency from y=0 to y=1 lead to the increase of apical oxygen distance to $Cu(2)O_2$ plane [3].

For the heavily underdoped single crystal $YBa_2Cu_3O_y$ (y=6.32 and 6.3), a hysteresis was observed below 20 K in the magnetoresistance curve as a function of magnetic field. This result was explained by assuming a developed array of charged stripes and by the fact that the magnetic field induces a topological ordering in the stripes [4].

The Fe ions substitute mainly in Cu(1) chains and are grouped together to form clusters. For low concentration of Fe (3-5at% Fe) the cluster shape must be linear, and the distance between the linear clusters is large [5].

By varying the Bi content in the $Bi_{2+X}Sr_{2-X}CuO_{6+\delta}$ films prepared by the RF magnetron sputtering method, the electrical resistance changed from superconductor to insulator [6,7]. Bi:2201 epitaxially thin films grown by RF and DC sputtering on a $SiTiO_3$ substrate presents a clear nonmetallic upturn near the superconducting transition in ρ(T) [9-11].The Bi:2201 superconductor is found to have a large residual resistivity. This behavior of Bi:2201 films is similar to the superconductor-insulator change observed in films of other materials [12-14].

By controlling the oxygen concentration δ (by successive annealing treatments of $Bi_2Sr_{1.6}La_{0.4}CuO_{6+\delta}$) the same film is changed from overdoped to strongly underdoped state [15]. As a result the electrical resistivity is changed from T-linear behavior (optimal doping) to insulating behavior.

In order to obtain underdoped samples, we used a partial substitution of Cu by x = 0.04 Fe in $YBa_2\ (Cu_{0.96}\ Fe_{0.04})_3Oy$ bulk samples (with y=6.55; 6.75 and 7) and low concentration of oxygen in the sputtering gas in the synthesis of c-axis oriented epitaxial Bi:2201 thin films deposited onto $SrTiO_3$ and MgO substrates We present a systematic evolution of electric resistance as a function of temperature for underdoped samples and the results were analysed in relation with the influence of Fe clusters and oxygen ordering.

2. Experimental

A series of $YBa_2\ (Cu_{0.96}\ Fe_{0.04})_3O_y$ samples (y=6.55;6.75; 7) were prepared by using solid state reaction method for $CuO, Y_2O_3, BaCO_3$ and Fe_2O_3,[16].

The oxygen content of the samples was changed by the thermal treatment of samples at 500 °C in a controlled air pression atmosphere. The standard iodometric titration, shows in our x=0.04 Fe doped Y:123 samples the values y=6.55, 6.75 and 7 for the oxygen content

X-ray diffraction measurements show that all samples are single phase of Y:123 system with a tetragonal structure.

Electrical resistance was measured by using the standard four-point method. Gold wires were attached with silver paste on the samples.The electrical contact resistivity was typically less than $2.10^{-4}\ \Omega cm^2$. The current

intensity passed through sample was between 1 mA and 10 mA.

First, the electrical resistance measurements as a function of temperature were performed by a slowly raising of the temperature from 5 K to room temperature, followed by the slowly lowering to 5 K. After these measurements, the samples were fast quenched from 250 K to liquid helium temperature and the electrical resistance was measured by a slowly raising of temperature up to 300 K. Thermal fast quenching of the samples was performed by the quenching in liquid nitrogen followed by insertion into the cryostat, which was precooled, to helium temperature.

Bi:2201 thin films were deposited onto heated single crystal $SrTiO_3$ and (100) MgO substrates by using an inverted cylindrical DC magnetron for the sputtering [11]. An off-stoichiometric target with a nominal composition Bi:Sr:Cu = 2.0:1.95:1.05 was home made by a solid state reaction method. The sputtering gas was a mixture of oxygen and argon with the following ratio f_{O2}/f_{Ar} =0.5/0.5; 0.45/0.55 ;0.4/0.6, and.0.35/0.65. The deposition time was 1 h, leading to nanoscale thin films with thickness of approximately 90 nm. Energy dispersive X-ray analysis (EDX) and X-ray photoelectron spectroscopy (XPS) shows that the compositions for the deposited films are Bi:Sr:Cu=2.00:1.92:1.01.

The films are chemically patterned and equipped with silver sputtered contacts pads. The temperature dependence of the in-plane resistivity is measured by using a standard four probe dc method. The XRD $2\theta/\theta$ – scanning patterns showed the presence of peaks associated exclusively with (00l) planes, and confirmed that the films had c-axis orientation of Bi:2201.

3. Results

Fig. 1 shows the influence of two distinct T1 and T2 thermal cycle on the electrical resistance as a function of temperature for $YBa_2(Cu_{0.96}Fe_{0.04})_3O_y$ sample (y=6.75).

In the first thermal cycle (T_1), the electrical resistance was measured by slow cooling (resistance down in Fig. 1) followed by slow heating (resistance up in Fig. 1). For all samples the R(T) dependence in thermal cycle T_1 is reversible. For y=6.75 sample the first derivative of electrical resistance versus temperature has a structure of two peaks centred on the T_{c1}=35.5 K and T_{c2}=18 K. For samples with y=7 and 6.55 only a single peak located at mean field critical transition temperature T_c=68.4 K and T_c = 19K is present. The decrease of Tc with decreasing oxygen content in our samples is in relation with the disorder in Cu(1)O chains, as a result of interstitial oxygen attracted by Fe clusters from apical positions. This lead to the decrease of of the carrier density in $Cu(2)O_2$ planes and the introduction of correlated Cu(2) magnetic moments. As a result, the samples contain micro and nano-regions, which are magnetically correlated. The amount of sample showing magnetic character depends not only on the average oxygen content but also on how the oxygen is distributed about the chain sites [17]. The two values of T_c suggest that in y=6.75 sample coexist two different phases, as a result of oxygen migration near the cluster positions. In these regions the oxygen content, and the ordering of oxygen in these micro-and nano domains may be different.

In the thermal cycle T_2 (resistance quenched in Fig. 1) the electrical resistance was measured by slow heating after a fast quenching from T=250 K to 5 K. Some irreversibilities are present in the R(T) curves (resistance quenched in Fig. 1) after this thermal cycle. The R(T) data for y=6.55 sample was nearly the same as for T_1 cycle, but T_c decreased with 1K. For y=7 sample, the electrical resistance of normal state increases and T_c decrease with 4K. For y=6.75 the low peak centred on 17 K disappear and the transition is characterized by a single peak centred at T_{c2}=35.5 K. The disappearance of low resistive transition after the thermal quenching from 250 K to 5 K, may be related to the oxygen distribution around the Fe cluster positions near this temperature.

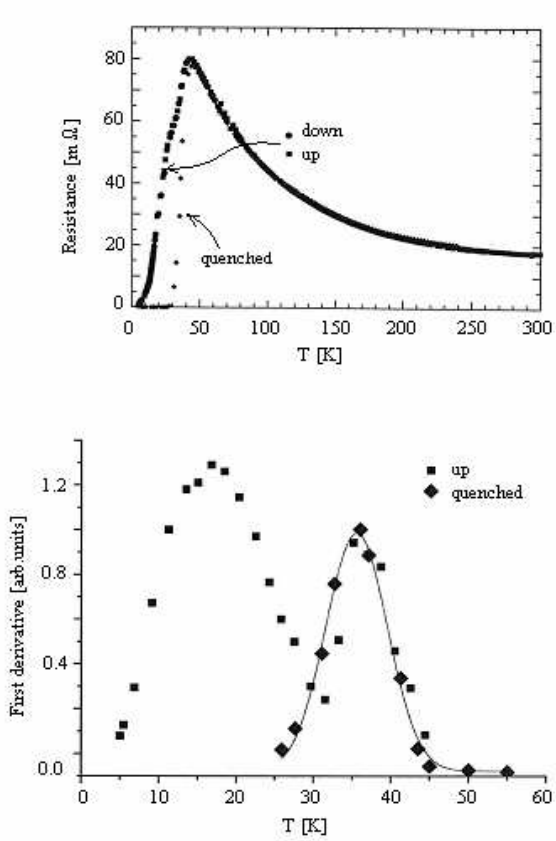

Fig. 1. Electrical resistance and first derivative of electrical resistance as a function of temperature for underdoped $YBa_2(Cu_{0.96}Fe_{0.04})_3O_y$ sample with oxygen content y = 6.75 for the thermal cycles T1 (*resistance down and resistance up* in the figure) and T2 (*resistance quenched in the figure).*

Fig. 2 displays ρ-T characteristics for Bi:2201 thin films deposited on $SrTiO_3$ substrates, prepared by using sputtering gas with an oxygen fraction in the range $0.3 \le f_{O2} \le 0.42$.

The ρ-T characteristics were changed drastically for slightly different f_{O2}, and the resistivity at room temperature increases monotonically with decreasing f_{O2}.

By decreasing the concentration of oxygen in sputtering gas, the critical transition temperature decreases and for $f_{O2} = 0.30$ the superconductivity is lost. This result suggest that the oxygen acts as a hole – providing layer to the Cu-O plane and determines the superconducting properties. Insulating behavior occurs starting at the temperature T_M, were ρ(T) is minimum (represented by arrows in Fig. 3). The gradually increases of T_M by decreasing f_{O2}, agree with the decrease of the carrier concentration.

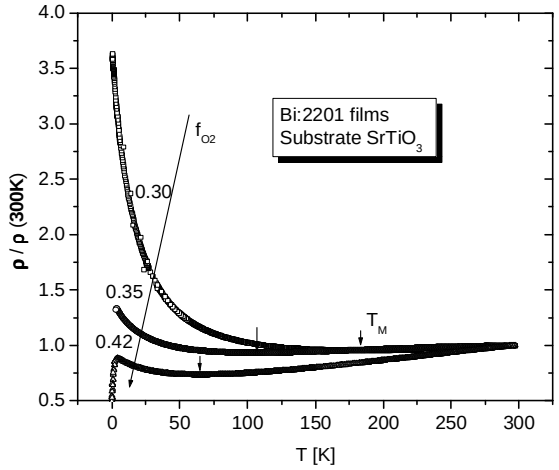

Fig. 2. The temperature dependence of the normalized electrical resistivity of Bi:2201 films obtained for different oxygen fraction f_{O2} in the sputtering gas. The minimum in $\rho(T) / \rho(300\,K)$ is indicated by vertical arrows.

Similar behaviors were also found in $Bi_2Sr_{1,6}La_{0.4}CuO_y$ thin films and single crystal, by changing the oxygen content by successive annealing treatments [15] and by increasing the magnetic field [18], respectively.

4. Discussion

We turn attention to the precise temperature dependence of the normal state resistivity for our samples.

A variety of functional forms do not fit the normal state resisitance, including thermal activation ($\ln\rho \sim -1/T$), various types of variable range hopping(VRH)conduction ($\ln\rho \sim T^{-\beta}$with $\beta = 1/2,1/3, 1/4$) and power law dependence (($\ln\rho \sim \ln T$).The data for samples by y=6.75 and y=6.55 exhibit a ln(1/T) dependence [$\rho \sim \ln T$] over most of temperature range (above T_c up to 70 K), as shown in Fig. 3. The slight downward deviation from ln(1/T) dependence at the lowest temperature results from the proximity of the superconducting transition. The origin of the logarithmic divergence of electrical resistance at lower temperature, in principle may lie in the Kondo effect, localization, interaction effect or pinning of the dynamically fluctuating striped phase. We shall take up the question of its origin later in the discussion. The transition from superconductor to insulator is attributed to same kind of localization.

Our previous studies on Bi:2201 thin films [19] show that in the low temperature region, the temperature dependence of log ρ vs. $T^{1/(n+1)}$ does not give straight lines for n=1,2,3; which suggest that the VRH (variable range hopping) model does not explain the localization in our samples.

A lg (1/T) behavior of ρ(T) was previously observed in underdoped superconducting LSCO [20] and BSLCO [21] both in a 60 T magnetic field , and in BSLCO thin films with various oxygen concentrations, respectively [15].

This behavior is observed only for the state close to the metal – insulator transition (MI) and for $T \leq 20$ K.

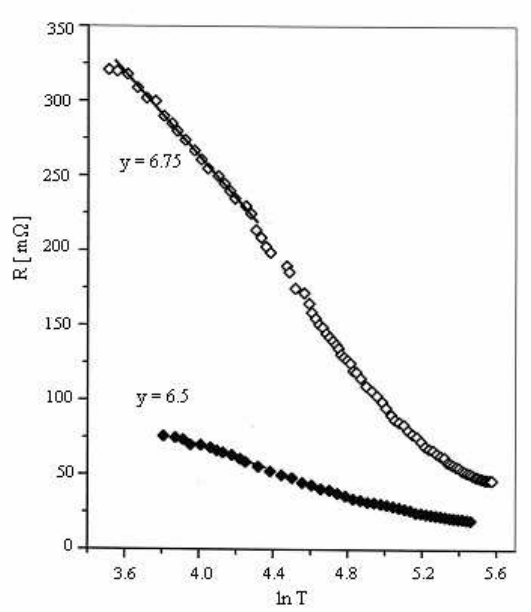

Fig. 3. The electrical resistance -plotted versus lnT , for y=6.55 and y=6.75 samples after the quenching process in T_2 cycle .

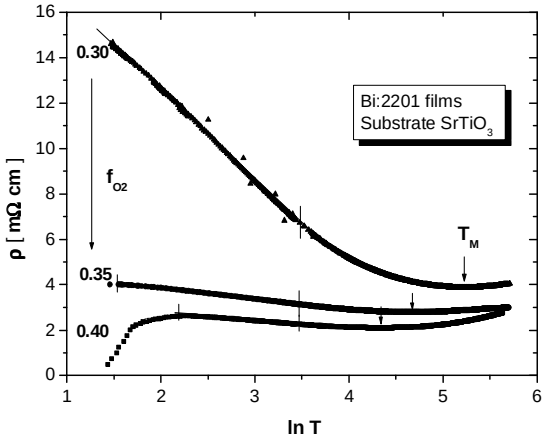

Fig. 4. Electrical resistivity versus lnT in lower temperature region. The vertical lines shows the temperature range for linear dependence.

To check the validity of this observation in our Bi:2201 films, in Fig. 4 ρ_{ab} was replotted vs. lnT for films with f_{O2} in the (0.3 – 0.4) range. The plot gave almost a straight line, for all underdoped samples, below the temperature T=34 K.

In the Emery model [22], the local pre-formed pairs has the gap equal to the spin-gap which is transferred to the holes in the stripe by hopping of electron pairs perpendicular to the stripe. The concept of stripes suggest the existence of AFM spin structure modulation in form of antiphase domains of AFM ordered spins separated by narrow extended stripes of holes.

It is interesting to analyse our resistance data in the normal state within a model considering 1D stripes formation in the $Cu(2)O_2$ plane [22,23]. An argument for this analysis is that inelastic neutron scattering experiments on Y:123 show evidence for the existence of rather dynamic stripes, and the 1D feature should not be limited to the Cu(1)O chain direction only [24]. This model is based to the assumption that the dominant scattering mechanism is of magnetic origin. The temperature dependence of electrical resistivity is described with the inelastic length L_Φ, controlled by the magnetic correlation length ξ_m. In this model, the occurrence of stripe fragmentation (or pinning) is related to the scattering processes with $L_\Phi \sim \xi_m$ which destroy the fragile regime of stripped CuO_2 planes. By using a $L_\Phi \sim 1/T^\alpha$ for the temperature dependence of inelastic length, an ln(1/T) dependence of electrical resistivity was predicted for low temperatures.

This result agrees with a model considering 1D stripes in the $Cu(2)O_2$ planes of underdoped bulk Y:123 samples and Bi:2201 nanoscale thin films.

5. Conclusions

The underdoped bulk $YBa_2 (Cu_{0.96} Fe_{0.04})_3O_y$ and Bi:2201 nanoscale thin films were obtained by the control of oxygen content.

For all samples of Y:123 underdoped system, electrical resistance as function of temperature shows insulating behaviour below room temperature, in the normal state.

The presence of two resistive transitions before the fast quenching of sample with y=6.75 sample is more probably produced by the two different ordering of apical oxygen in chains. The disappearance of low resistive transition after the thermal quenching from 250 K to 5 K, may be related to the oxygen distribution around the Fe cluster positions near this temperature.

For Bi:2201 films, by decreasing the oxygen fraction, insulating behavior occurs starting at the temperature T_M, were $\rho(T)$ is minimum. Above the temperature T_M electrical resistivity shows a "metallic" behavior.

A (lnT) dependence of electrical resistivity was evidenced below 70 K for Y:123 samples and below 34 K for underdoped Bi:2201 thin films. This result agrees with a model by 1D stripes in the $Cu(2)O_2$ planes.

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