# **Doping dependence of magnetoresistivity in polycrystalline Y(Ca)BCO**

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Many important characteristics ( $T_c$ ,  $J_c$ ,  $H_{irr}$  ect.) of cuprate superconductors depend on doping level in three main regions: underdoped, optimally doped and overdoped. In this study magneto-resistive transition in Y(Ca)BCO polycrystalline samples with different level of doping is examined. The upper critical field and irreversibility lines were determined. Their behavior is explained by analogy with domelike shape of doping dependence of the superconducting transition temperature.

(Received September 25, 2012; accepted February 20, 2013)

Keywords: Overdoping, Magnetoresistance, Irreversibility and Hc2(T) lines

## 1. Introduction

High temperature superconductors (HTS) are Mott insulators in the extreme limit of low carrier concentration per CuO<sub>2</sub> plane ( $p \le 0.05$ ). Not only superconductivity but many important superconducting characteristics (critical parameters – T<sub>c</sub>, J<sub>c</sub>, H<sub>c2</sub>, irreversibility line) crucially depend on the doping level.

The upper critical magnetic field  $(H_{c2})$  is important characteristic from fundamental and practical point of view. According to Ginsburg-Landau theory it is connected with the coherence length,  $\xi$ , (H<sub>c2</sub>(0) =  $\Phi_0/2\pi\xi^2$ ) which can be calculated if  $H_{c2}(0)$  is estimated. For that purpose the Werthamer-Helfand-Hohenberg (WHH) formula:  $H_{c2}(0) = -0.69 T_c (-dH_{c2}/dT)_{Tc}$  is often used [1]. However, it must be point out that WHH theory concerns the conventional superconductors with s-pairing and should be used with caution when is applied for the analysis of the experimental data of HTS samples. The large thermal and quantum fluctuations near T<sub>c</sub> (increasing further in magnetic field) are the other problem for  $H_{c2}$ determination from transport and magnetization measurements [2]. In order to overcome the need of extremely high magnetic fields, the resistive transitions studies in cuprates are limited to samples with suppressed  $T_c$  and  $H_{c2}(0)$ , as it is in the underdoped or overdoped samples [3]. The optimal doping is correlated with the maximization of T<sub>c</sub> and it is achieved at p=0.16 carriers per CuO<sub>2</sub> plane. T<sub>c</sub> falls to zero at p=0.05 and p=0.27 in the underdoped and overdoped sides respectively [4]. Underdoped samples are easily obtained by variation of oxygen content, while for overdoped ones additional carriers should be supplied (with chemical substitution for example).

In this study we investigate polycrystalline, Casubsituted, overdoped samples with different level of overdoping. The influence of doping level on the irreversibility line,  $H_{c2}(T)$  line and coherence length is examined using the experimental results of magnetoresistance.

### 2. Experimental

The polycrystalline samples with different Ca  $(Y_{0.975}Ca_{0.025}Ba_2Cu_3O_{7-\delta})$ substitution and  $Y_{0.8}Ca_{0.2}Ba_{2}Cu_{3}O_{7-\delta}$ ) are obtained and investigated. They are prepared by solid state reaction method from high purity Y<sub>2</sub>O<sub>3</sub>, BaCO<sub>3</sub>, CuO and CaCO<sub>3</sub> powders. The obtained mixture was grinded. The grinding and heating steps were repeated three times. The first step was calcinations at 925 °C in flowing oxygen for 23 hours. During the second step samples were heated to 930 °C for 23 hours, followed by slow cooling (2 °C/min) and additional annealing for 2 hours at 450 °C in flowing oxygen. Tablets were pressed before the third synthesis in oxygen at 950 °C for 23 hours, annealed at 450 °C for 48 hours and finally slow cooled to room temperature.

The crystal structure of the specimens was examined by X-ray Powder Diffraction System STOE with  $Cu_{k\alpha}$ radiation at room temperature.

The magnetoresistive measurements were performed on Quantum Design PPMS; the voltages were detected with an error of several nanovolts. The applied magnetic field was perpendicular to the current direction. The resistivity vs. temperature measurements were done at different magnetic fields ranging from 0.1 T to 6.9 T. The magnetic field values for samples have been determined by previous investigations of irreversibility line performed by AC magnetic susceptibility measurements [5].

#### 3. Results and discussion

X-ray analysis shows that both samples are single phase with a minor BaCuO<sub>2</sub> impurity phase for sample with x=0.20. The critical temperatures determined from  $\rho(T)$  measurements at zero magnetic field are 90.6 K and 80.4 K for the samples with x=0.025 and x=0.20, respectively. The T<sub>cmax</sub> for both samples are accepted to be 92.6 K for Y<sub>0.975</sub>Ca<sub>0.025</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>7-\delta</sub> and 86.6 K for Y<sub>0.8</sub>Ca<sub>0.2</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>7-\delta</sub> [5]. The carrier concentration in the CuO<sub>2</sub> plane, p, was estimated from the relation T<sub>c</sub>/T<sub>cmax</sub>=1-82.6 (p-0.16)<sup>2</sup> [4] and the obtained results are p=0.176 for sample Y<sub>0.975</sub>Ca<sub>0.025</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>7-\delta</sub> and p=0.189 for Y<sub>0.8</sub>Ca<sub>0.2</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>7-\delta</sub>. Thus both of our samples are overdoped.

On Fig. 1 (a, b) resistivity vs. temperature dependencies at different magnetic fields are presented for sample with x=0.025 and x=0.20, respectively.



Fig. 1. Resistivity vs. temperature dependencies at different magnetic fields for (a)  $Y_{0.975}Ca_{0.025}Ba_2Cu_3O_{7-\delta}$  and (b)  $Y_{0.80}Ca_{0.20}Ba_2Cu_3O_{7-\delta}$  samples.

The observed double step transitions could be interpreted as a result of vortex dynamics in intra- and inter-granular regions. The intergranular transition is more stable in more ovedoped sample and persists up to the highest magnetic field used. This result is in consent with our previous investigations [6] and shows that S-N-S intergrain connections established in overdoped samples are more stable in magnetic field. The intra-grain behavior is believed to be determined by the collective flux creep, while the inter-grain one is usually more complicated. It is not clear whether it is governed by Josephson – vortex pinning and creep or by Josephson current across the grains [7].

The normal state resistivity  $(\rho_n)$  has been determined from the intersection of two straight lines extrapolations of the resistivity above the critical temperature and the steepest slope in the transition. For determination of the irreversibility line  $(H^{\rho}_{irr}(T))$  the criterion 0.01. $\rho_n$  has been used, while  $0.9.\rho_n$  criterion determines the  $H_{c2}(T)$ dependence. On Fig.2(a) the  $H^{\rho}_{irr}(T)$  and  $H_{c2}(T)$  lines as a function of reduced temperature (T/T<sub>c</sub>) are presented for both samples. The diminished separation between H<sub>irr</sub> and  $H_{c2}$  lines for sample  $Y_{0.975}Ca_{0.025}Ba_2Cu_3O_{7\text{-}\delta}$  indicates better pinning in it, in consent with our previous M(H) investigations [5]. It should be mentioned that for both samples the irreversibility lines show positive curvature (PC) and no tendency of saturation could be seen at low temperatures (up to  $T/T_c \sim 0.65$  for sample with x=0.20).  $H_{c2}(T)$  lines for both samples have similar behavior, which is better seen in rescaled presentation on Fig.2(b). These results are in contradiction with the theory of conventional superconductors, where the s-wave pairing is assumed [1]. More reliable description of observed PC suggest methods accounting for the influence of quantum critical point [8] and the mixing of  $d_{xy}$  and  $d_{x2-y2}$  components due to the magnetic field [9]. In spite of that, simple WHH formula for  $H_{c2}(0)$  estimation is often used.





Fig. 2. (a)  $H_{irr}^{\rho}$  and  $H_{c2}$  as a function of reduced temperature  $(T/T_c)$  for both samples. (b) Rescaled  $H_{c2}$  vs. temperature dependencies for both samples. The lines represent the quadratic fits to the data and underlined the positive curvature of the  $H_{c2}(T)$  dependencies.

On Fig. 3 irreversibility lines determined by different methods are presented for the sample with x=0.20. The  $H^{\rho}_{irr}(T)$  line was determined as it is explained above.  $H^{ac}_{irr}(T)$  line has been determined by third harmonic AC magnetic susceptibility measurements. The observation of high harmonics signal ( $\chi_n$ ) implies the nonlinear response of the flux system. The irreversibility in magnetization behavior is a result of bulk pinning, surface or edge barrier effects and contributes to the third harmonics ( $\chi_3$ ) signal. In case of superposition of DC and AC magnetic fields, when  $H_{ac}$ <= $H_{dc}$  the flux creep is non - linear and the irreversible dynamical regime determines the  $\chi_3$  signal [10].



Fig. 3.  $H_{irr}$  vs. temperature dependencies for sample  $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$  obtained by three different methods.

The vortex lattice in HTS is highly disordered due to the small coherence length and anisotropy. In such bulk disordered system a second order phase transition and sharp equilibrium phase boundary between vortex -glass and vortex-liquid thermodynamic phases were proposed to exist [11, 12]. According to this model a power -low behavior is expected between voltage and current E(J, T=T\_g) ~ J^{(z+1)(D-1)} or  $\rho(J, T=T_g) ~ J^{(z+1)(D-1)}$  at phase transition temperature  $T_{\text{g}},$  where  $\tilde{D}$  is the dimensionality of the system and z is the so called dynamical exponent. The phase transition temperature (T<sub>g</sub>) determined at different magnetic fields in fact defined the irreversibility line, H<sup>1-</sup> <sup>4</sup><sub>irr</sub> [13]. To the best of our knowledge I-V curves were not used for determination of irreversibility line for Ca substituted samples. In spite of the fact that I-V curves were recorded changing the temperature at intervals of 1K the method is reliable for H<sub>irr</sub>(T) determination. As it is shown on Fig.3 a satisfactory agreement between the results obtained by these three different methods exists. The same correlation was found for the other sample also.

For the purpose of comparison of two samples with different level of overdoping rough estimation of  $H_{c2}(0)$  could be done by WHH formula. For sample  $Y_{0.975}Ca_{0.025}Ba_2Cu_3O_{7-\delta}$  we obtain  $H^*_{c2}(0)=344$  T and for  $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta} - H^*_{c2}(0)=178$  T. Thus increasing the level of overdoping decreases the  $H^*_{c2}(0)$  value. The star index reminds that the obtained values are not exactly the  $H_{c2}(0)$ . The coherence length was also calculated. Our estimations give  $\xi=0.96$  nm for  $Y_{0.975}Ca_{0.025}Ba_2Cu_3O_{7-\delta}$  and  $\xi=1.34$  nm for  $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$ . In contrast to  $H_{c2}(0)$  behavior the coherence length increases when the level of overdoping is raised.

#### 4. Conclusions

In conclusion we demonstrate that the behavior of magnetoresistive transition depends on the level of overdoping. The observed PC of  $H_{irr}(T)$  and  $H_{c2}(T)$  lines support the non-conventional character of superconductivity and the speciality of ovedoped state. A good consent is demonstrated between H<sub>irr</sub>(T) lines determined by different methods: magnetoresistive measurements, third harmonics AC magnetic susceptibility and current-voltage characteristics at different magnetic fields. The decreased  $H_{c2}(0)$  and growing coherence length have close relation with increasing level of carrier concentration in investigated samples. These results support the assertion that important characteristics of cuprates depends on the level of doping.

#### Acknowledgments

The authors are grateful to G. Fuchs for the helpful comments. The financial support(ed) through the EURATOM (Project FU07-CT-2007-00059) and Bulgarian Science Fund through the Association EURATOM-INRNE is also greatly acknowledged.

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