

## Transverse resistance in $HoBa_2Cu_3O_{7-\delta}$ single crystals

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The transverse electrical resistance of HoBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals is investigated in the temperature range  $T_c - 300$  K for optimally-doped ( $T_c \approx 91$  K) and oxygen-poor ( $T_c \approx 51$  K) samples. With decreasing temperature, the resistivity of the optimallydoped samples has been found to transit from the regime of scattering on phonons and defects to the regime of "semiconductor" character and, near  $T_c$ , of the fluctuation conductivity. The oxygen-poor samples have been revealed to exhibit only a variable range hopping conductivity of "semiconductor" character, which near  $T_c$  transits into the fluctuation conductivity. A significant anisotropy of the residual resistivity and characteristics of the fluctuation conductivity is observed for samples of both types.

*Keywords*: Superconducting cuprates; holmium; electrical resistance measurements; anisotropy.

#### 1. Introduction

Identification of the charge carriers scattering mechanisms is important for understanding the origin of a series of nontrivial phenomena peculiar to high-temperature superconductors (HTSCs) in the normal state.<sup>1</sup> To these phenomena one ascribes the pseudogap<sup>2,3</sup> and fluctuation<sup>3–5</sup> anomalies, the metal-to-insulator transition,<sup>6,7</sup> the incoherent electronic transport and a series of other effects.<sup>8–10</sup>

The most easy-to-use HTSC compound for studying the aforementioned properties is the 1-2-3 system (YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>). This is stipulated by its rather high critical temperature ( $\approx 90$  K) above the nitrogen liquefaction temperature<sup>11</sup> and a

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relative simplicity to change the system's physical properties by varying the oxygen  $content^{12-14}$  and/or substituting its constitutive elements.<sup>15,16</sup>

However, as it was revealed in a recent work,<sup>17</sup> substituting Y by Ho in the case of oxygen-nonstoichiometric samples can substantially affect the ordering processes in the oxygen subsystem and stimulate the appearance of specific diffusion mechanisms, such as the ascending diffusion.

The system HoBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>, like other crystals from the ReBaCuO family (Re=Y or lanthanides), is essentially anisotropic.<sup>15</sup> In consequence of a ferroelastic tetra-ortho transition, in the course of saturating the system with oxygen, twins appear in crystals,<sup>18,19</sup> that give rise to an additional anisotropy<sup>20</sup> and act as a further scattering channel.<sup>21</sup> The redistribution of the labile oxygen leads to the appearance of inhomogeneities in the system.<sup>22,23</sup> Accordingly, these phenomena are of interest from both the fundamental and the applications-related viewpoints.

Previously,<sup>24</sup> we studied the basal-plane resistance in HoBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals and found that the effect of twins on the resistance is rather weak. The temperature dependences of the basal-plane resistance of HoBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals in the temperature range  $T_c$  – 300 K can be presented as a result of electron scattering on phonons and defects, in conjunction with the fluctuation conductivity near  $T_c$ .

Here, we investigate the transverse (along the *c*-axis) resistance of HoBa<sub>2</sub>Cu<sub>3</sub>- $O_{7-\delta}$  single crystals, when the effect of the twin boundaries is absent. The goal of this study is to identify the anisotropy of charge scattering mechanisms and parameters of the fluctuation conductivity as well as the effect of the oxygen content on the scattering processes.

## 2. Experimental Methods

The HoBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals were grown by the solution-melt technique in a gold crucible, similar to the procedure used for the growth of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals.<sup>25,26</sup> For resistive measurements, crystals of a rectangular shape with the dimensions 2 × 1.8 × 0.5 mm<sup>3</sup> and 1.9 × 1.9 × 0.5 mm<sup>3</sup> were selected from a onegrowth series. In what follows, the samples will be referred to as sample 1 and 2, respectively, denoting their state after annealing as 1a and 2a. The smallest crystal dimension corresponds to the *c*-axis. The electrical contacts to the samples were formed by applying a silver paste on the crystal surface followed by attaching silver leads of 0.05 mm diameter and a three-hour annealing in an oxygen atmosphere at 200°C. Such a procedure allowed us to obtain a contact transient resistance of less than 1  $\Omega$  and to conduct resistive measurements at transport currents of up to 10 mA in the *ab*-plane<sup>24</sup> and along the *c*-axis in the present work.

To obtain samples with the optimal oxygen content and a high  $T_c$ , they were annealed in an oxygen flow for three days at a temperature of 420°C. To reduce the oxygen content, the samples were annealed in air for a day at 500°C. The resistance measurements were performed in the temperature-sweep mode, upon both warming up and cooling down the samples. All measurements were done three days later after each annealing step that ensured a homogeneous oxygen distribution over the sample volume at room temperature.<sup>27</sup>

# 3. Results and Discussion

Figure 1 depicts the temperature dependences of the normalized transverse resistivity in the initial (curves 1 and 2) and the annealed (curves 1a and 2a) states. The curves are rather similar and close to each other, especially prior to annealing. Both curves 1 and 2 exhibit a minimum which is likely caused by the competition of the "metallic-like" and the "semiconductor-like" run of  $\rho_c(T)$ .

The 500°C annealing in air invoked a rise in the oxygen deficit up to  $\delta \approx 0.18$ ,<sup>28</sup> that has led to a substantial increase of the total resistivity value and the appearance of the  $\rho_c(T)$  dependences of the completely "semiconductor" character (curves 1a and 2a).

The shape of the derivatives  $d\rho_c/dT$  in the superconducting transition region is rather sensitive to the sample homogeneity. An analysis of these derivatives allows one to draw conclusions on the sample constitution, see e.g. Ref. 29.

Figure 2 displays the derivatives  $d\rho_c/dT$  in the superconducting transition region for both samples before and after annealing. In the initial state, the oxygen distribution is rather uniform as the derivative maxima (Fig. 2, curves 1 and 2)



Fig. 1. (Color online) Temperature dependences of the normalized transverse electrical resistivity of the two HoBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals: 1, 2 — before annealing, 1a, 2a — after annealing. Symbols — experiment, lines — fits to Eqs. (1)–(4).



Fig. 2. (Color online) The derivatives  $d\rho_c/dT$  in the vicinity of the superconducting transition for the two HoBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals. The curve numbering is as in Fig. 1.

are symmetric and have a nearly equal width. After annealing, the  $d\rho_c/dT$  maxima have substantially broadened, an asymmetry has appeared and, for sample 2, an additional maximum has appeared close to 54 K (Fig. 2, curve 2a) that points to the presence of a phase with this  $T_c$  in the sample.<sup>30</sup>

The temperature dependences of the transverse resistance (Fig. 1) have been approximated in the whole temperature range  $T_c - 300$  K by an expression accounting for the normal-state resistivity  $\rho_n^c$  as well as the fluctuation conductivity,  $\Delta \sigma_c^{AL}$ , in the 3D Aslamazov–Larkin model<sup>31</sup>

$$1/\rho^c = \sigma^c = (\rho_n^c)^{-1} + \Delta \sigma_c^{\text{AL}}, \qquad (1)$$

$$\Delta \sigma_c^{\rm AL} = \frac{se^2}{32\eta \xi_{ab}^2 \sqrt{2\varepsilon_0 \sinh(2\varepsilon/\varepsilon_0)}} \,. \tag{2}$$

This expression for the fluctuation conductivity is chosen to limit its influence,<sup>32</sup> where  $\varepsilon = \ln(T/T_c)$  is the reduced temperature,  $T_c$  is the critical temperature in the mean-field approximation,  $T > T_c$ ,  $\xi_{ab}$  is the coherence length in the *ab*-plane,  $\varepsilon_0$  determines the temperature interval for the superconducting fluctuations  $\varepsilon_0 = \ln(T_{\text{fluct}}/T_c)$ , and  $T_{\text{fluct}}$  is the characteristic temperature above which the superconducting fluctuations can be neglected.

It should be noted that according to Ref. 33, the appearance of fluctuating Copper pairs above  $T_c$  causes a decrease of the density of one-electron states at the Fermi level, that is, leads to the appearance of the pseudogap at  $T \approx T_{\text{fluct}}$ . However, it is difficult to account for the corresponding changes in the normal resistance because of the much more stronger effect of the fluctuation conductivity which acts simultaneously and determines  $\rho(T)$  near  $T_c$ .

The normal-state resistance can be presented in the form

$$\rho_n^c = (\rho_0^c + \rho_{ph}^c)(1 + bT^2) \exp(T_0/T)^m,$$

$$\rho_{ph}^c(T) = C_3 \left(\frac{T}{\theta}\right)^3 \int_0^{\theta/T} \frac{x^3 dx}{(e^2 - 1)^2}.$$
(3)

Here  $\rho_0^c$  is the residual resistivity across the layers, which characterizes scattering on defects and  $\rho_{ph}^c$  is the resistivity contribution owing to scattering on phonons.<sup>34</sup>  $C_3 \propto (\varepsilon_F)^{-1}$ , where  $\varepsilon_F$  is the Fermi energy,  $kT_0$  is the activation energy and the presence of the exponent m is due to the fact that in Ref. 35, a variable range hopping conductivity has been observed in this system.

After annealing,  $\rho_n^c$  has acquired a completely "semiconducting" character,  $C_3=0$  and

$$\rho_n^c = \rho_0^c (1 + bT^2) \exp\left(T_0/T\right)^m.$$
(4)

Since  $\rho_n^c = 1/\sigma = 1/e n_0 \mu^{36,37}$  ( $n_0$  is the concentration of the conductivity electrons), the parameter b is connected with the temperature dependence of the mobility  $\mu^{-1} = \rho_0 (1 + bT^2)$ .

The parameters of Eqs. (1)–(4) are compiled in Table 1. These provide a minimal fitting error for the transverse resistance for all samples. The data in Table 1 attest to the following features. The fitting parameters for both samples are rather similar for both, the initial and the annealed states. This is because, in contrast to Ref. 24, the applied current in both samples is parallel to the twin planes. The difference in  $T_c^c$  in the annealed state is somewhat larger than in the initial state, that can be attributed to a sample inhomogeneity appearing as a consequence of the annealing process.

The value of  $\rho^c$  (300 K) before annealing is close to the value of the maximal metallic resistivity value  $1 \div 5 \text{ m}\Omega \text{cm}.^{38}$  The ratio  $\rho^c$  (300 K)/ $\rho_0^c \approx 2$  is typical for disordered alloys. The annealing has enhanced the oxygen deficit and, consequently, the degree of disorder in the samples. This leads to a significant (in  $\approx 25$  times) rise of  $\rho^c$  (300 K). Such an increase of the transverse resistivity may be associated with a decrease of the density of electronic states at the Fermi level, that is also peculiar to conductors with a short electron mean free path and is caused by the electron–electron interaction.<sup>39</sup>

When comparing the data with those of Ref. 24, one finds a considerable anisotropy of the residual resistivity  $\rho_0^c/\rho_0^{ab} \simeq 300$  prior to annealing and  $\simeq 1000$  after annealing. The increase of the anisotropy with an increase of the disorder degree also attests to a change in the electronic characteristics in consequence of annealing. A certain role in this may be played by other specific mechanisms of the quasiparticle scattering.<sup>40–44</sup> A comparison of the data with Ref. 24 points to some

State	Optimally-doped		After annealing	
Parameters/samples	1	2	1a	2a
$T_c^c, \mathbf{K}$	91.3	91.6	51.2	50.4
$\rho^c$ (300 K), $\mu\Omega {\rm cm}$	9.02	9.12	144	146
$\rho_0^c / \rho \ (300 \text{ K})$	0.541	0.486	0.792	0.856
$ ho_0^c,  \mathrm{m}\Omega\mathrm{cm}$	4.88	4.43	114	125
$\rho_0^c / \rho_0^{ab}$ (Ref. 24)	320		950	
$C_3/\rho$ (300 K)	1.12	1.34	_	
$\theta^c$ , K	480	494	—	—
$\theta^c/\theta^{ab}$	1.5			
$b_0 \times 10^6,  \mathrm{K}^{-2}$	0	0.2	-5.9	-6.4
m	1	1	0.54	0.53
$T_0, \mathbf{K}$	46	46	193	194
$\xi_{ab}(0), \text{\AA}$	35	57	183	161
$\xi_{ab}(0)/\xi_c(0)$	33		110	
$\Delta T_{\rm fluct}^c = T_c - T_{\rm fluct},  \mathrm{K}$	0.7	0.5	17.6	2.9
$\Delta T_{\rm fluct}^{ab} / \Delta T_{\rm fluct}^c$ (Ref. 24)	16		1.5	
$\varepsilon, \%$	0.7	1	3	4

Table 1. Fitting parameters for Eqs. (1)–(4) for the transverse resistivity in the HoBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals.

anisotropy of the Debye temperature  $\theta$ . It is likely caused by the anisotropy of the interatomic distances and the force constants for the HoBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystal.

Before annealing, the parameter b > 0 and is small, that is, the mobility  $\mu$  decreases with increasing temperature which attests to a weak scattering on phonons.<sup>36</sup> After annealing b < 0 and its value has increased by an order of magnitude. The mobility after annealing grows with increasing temperature which attests to the dominating scattering on ionized defects,<sup>36</sup> in this case on vacancies in the oxygen subsystem. Accordingly, the change of the parameter b suggests that at small  $\delta$  the charge carriers are primarily scattered on phonons, whereas at large  $\delta$  on the oxygen vacancies.

The exponent *m* varies from 1 before annealing to  $m \approx 0.5$  after annealing. The values  $m \approx 1/4 \div 1/2$  are typical for the variable range hopping conductivity.<sup>38,45</sup> The activation energy of the process,  $kT_0$ , increases by about a factor of five as a consequence of annealing. A comparison of the data with Ref. 24 reveals a notable anisotropy of characteristics of the fluctuation conductivity. The anisotropy of the coherence length grows from  $\xi_{ab}(0)/\xi_c(0) \approx 30$  before annealing to  $\approx 110$  in the annealed state. For the temperature range of the superconducting fluctuations,

the anisotropy decreases from  $\Delta_{\rm fluct}^{ab}/\Delta T_{\rm fluct}^c \approx 16$  in the initial state down to  $\approx 1.5$  in the annealed state.

# 4. Conclusion

The results of the study of the electrical resistance in the  $HoBa_2Cu_3O_{7-\delta}$  single crystals allow one to draw the following conclusions:

- (i) For the optimally-doped samples the temperature dependence of the transverse resistance is stipulated by scattering on defects and phonons. With decreasing temperature, this dependence transits into the dependence of "semiconductor-like" character and, further on, to the fluctuation conductivity near  $T_c$ .
- (ii) Annealing enhances the oxygen deficit and leads to the dependence of purely "semiconductor-like" character with a variable range hopping conductivity and a transition to the fluctuation conductivity near  $T_c$ .
- (iii) A significant anisotropy of the residual resistivity and characteristics of the fluctuation conductivity has been observed.

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