Functional Materials 14, No.1 (2007) © 2007 — Institute for Single Crystals

Excess conductivity and pseudogap state in $YBa_2Cu_{3-y}Al_yO_{7-\delta}$ single crystals with a system of unidirectional twin boundaries

R.V.Vovk, M.A.Obolenskii, A.V.Bondarenko

V.Karazin Kharkiv National University,
4 Svobody Sq., 61077 Kharkiv, Ukraine

Received June 30, 2006

The conductivity in the basis plane of $YBa_2Cu_{3-y}Al_yO_{7-\delta}$ single crystals with a pre-specified topology of plane defects has been investigated. The results indicate that the doping with Al results in increased number of the effective scattering centers of the normal carriers. The excess conductivity for the analyzed single crystals within a wide temperature interval corresponds to an exponential temperature dependence. Near T_c, it is described well by the Aslamazov-Larkin theoretical model. A partial substitution of Cu with Al results in a considerable expansion of the temperature interval of pseudo-gap anomaly existence in the *ab*-plane.

Исследована проводимость в базисной плоскости монокристаллов $YBa_2Cu_{3-y}Al_yO_{7-\delta}$ с заданной топологией плоских дефектов. Показано, что внесение примеси Al приводит к возрастанию числа эффективных центров рассеяния нормальных носителей. Избыточная проводимость исследованных образцов в широком интервале температур подчиняется экспоненциальной температурной зависимости, а вблизи T_c удовлетворительно описывается теоретической моделью Асламазова-Ларкина. При этом частичная замена Cu на Al приводит к значительному расширению температурного интервала существования псевдощелевой аномалии в *ab*-плоскости.

The transport properties in high-temperature superconductors (HTSC) are dependent on the type and concentration of defects and the oxygen stoichiometry [1–3]. Doping of YBaCuO compounds with aluminum is known to result in substitution of copper atoms in the CuO planes [1, 2]. The influence extent of such changes on the transport properties (i.e. electric conductivity) is still unclear. For example, in [1], an insignificant increase of the electric resistance in the basic plane (ρ_{ab}) was reported for $YBa_2Cu_{3-y}Al_yO_{7-\delta}$ single crystals with $y \leq 10$ %. On the contrary, concentration a two-fold increase of ρ_{ab} for the same aluminum concentration was observed in [2]. Such disagreement seems to be due to non-homogeneous distribution of aluminum in the crystal bulk, because an uncontrolled aluminum doping in the course of the crystal growth in alundum crucible. In particular, the non-homogeneous distribution of aluminum is evidences by the wide transitions to superconducting state ($\Delta T_c \geq 2$ K) and their stepwise shape [1, 2]. There is also a significant scatter in the superconducting parameters. Moreover, twin boundaries (TB) always exist in YBCO single crystals [3]. The contribution of these defects is difficult to determine in experiment. The aim of this work is to study the effect of low aluminum doping (up to 5 %) in various conductivity regimes in the YBCO single crystals with high T_c and unidirectional TB system at the transport current vector $\mathbf{I} \| \mathrm{TB}$, when the influence of twin planes on carrier scattering processes is minimal.

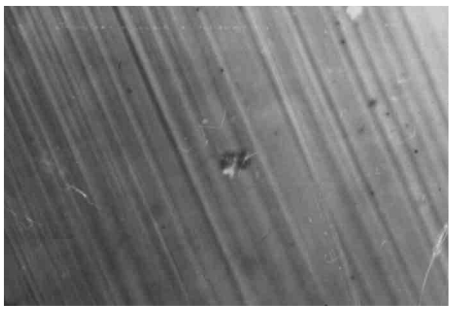

Fig. 1. Surface photo of a K2 single crystal, where its characteristic twin grid is visible (polarized light, ×440).

The $YBa_2Cu_{3-y}Al_yO_{7-\delta}$ single crystals were grown using the technique described in a previous study [3]. The Y_2O_3, $BaCO_3$, CuO powders of special purity grade were used as initial materials. To obtain aluminum doped single crystals, Al_2O_3 (0.2 wt. %) was added to the raw blend. The growth and oxygen saturating regimes for $YBa_2Cu_{3-y}Al_yO_{7-\delta}$ crystals are identical to those described for undoped single crystals [3]. For the resistivity measurements, thin 0.5×0.5 mm^2 crystals with unidirectional TB were selected (Fig. 1). This geometry was selected so that it was possible to cut out 0.2 mm wide bridges with parallel TB and 0.3 mm distance between the pair of contacts (see Fig. 2). The electric resistance in the *ab*-plane was measured using the standard four-contact method using dc current up to 10 mA. The temperature was measured by a platinum thermo-resistor.

Fig. 1 is a photograph of the aluminum doped crystal with its characteristic twin grid. As is known, the trivalent impurity ions are the defect-originating centers [4] and when their density increase, the period of the domain twin boundary structure diminishes. As a result, the neighboring micro twins become overlapped and a "tweed" type structure is formed [4]. It is seen from the Figure that in studied $YBa_2Cu_{3-y}Al_yO_{7-\delta}$ crystals, the "tweed" structure was not observed, perhaps due to low Al concentration, and the distance between TB was two or three times smaller than in pure crystals.

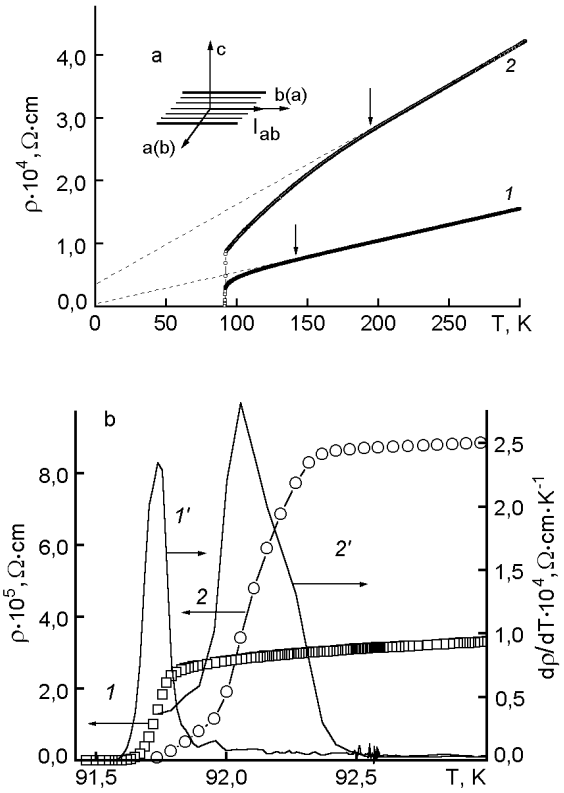

Fig. 2. Temperature dependences of electric resistivity $\rho_{ab}(T)$ for K1 and K2 single crystals (curves *1* and *2*, respectively). The T^* — PG transitions temperatures are indicated by arrows. Inset: superconducting transition for the same samples in $\rho_{ab} - T$ and $d\rho_{ab}/dT - T$ coordinates. The curves in the inset and in the figure are numbered identically.

The temperature dependences of electric resistivity in the *ab*-plane, $\rho_{ab}(T)$, for the YBCO (K1) and $YBa_2Cu_{3-y}Al_yO_{7-\delta}$ (K2) crystals are shown in Fig. 2. The inset represents the resistivity transitions into the superconducting state for the same samples in $R - T$ and $dR/dT - T$ coordinates. It is seen that in both cases, the dependences are metallic, but the ρ_{ab}(300 K)/ρ_{ab}(0 K) ratio varies and its value is 40 and 12 for K1 and K2 crystals, respectively. The ρ_{ab}(0 K) value was determined by extrapolating the linear $\rho_{ab}(T)$ conductivity section to zero

Table. Parameters of experimental samples.

Sample	T_c, K	ρ_{ab}(300), μΩ ·cm	T^*, K	Δ^*_{ab}, meV	ε_0	α_{3D}	α_{2D}	ξ_c(0), Å
K1	91.74	155	143	88.4	0.064	−0.499	−1.012	1.48
K2	92.05	421	199	58.1	0.157	−0.506	−0.990	2.32

temperature, as shown in Fig. 2. The resistivity value in *ab*-plane for the K1 and K2 crystals at room temperature was 155 and 421 $\mu\Omega \cdot$ cm and their critical temperatures are 91.7 and 92.1 K, respectively. The data obtained for the samples are presented in Table. According to literature data on the dependence of T_c on aluminum concentrations [1, 2], we can conclude that Al concentrations in the K2 crystal is less that 5 %, when the oxygen concentration is $\delta \le 0.1$ [3]. At the same time, the narrow superconducting transition width $\Delta T_c \le 0.5$ K for both samples evidences the homogeneous distribution of oxygen and Al over the crystal volume. As the transport current vector, **I**, was parallel to the TB for all the samples, the increase of electric resistivity in the Al-doped crystal having a smaller interplane spacing cannot be explained by the increase of TB density. Thus, the observed two-fold increase of ρ_{ab} value could be probably caused either by the current carrier density reduction or by the appearance of efficient carrier scattering centers. Taking under consideration the increase of Hall signal at comparable Al concentrations [1], we that the observed ρ_{ab} increase can be concluded to be defined mainly by increasing number of scattering centers. The change in $\rho_{ab}(300\ \mathrm{K})/\rho_{ab}(0\ \mathrm{K})$ ratio is also indicative to this conclusion. The role of such centers can be played by the trivalent Al impurity [4] as well as by the increasing number of vacancies. The last supposition is supported by the increase of twin density in the Al-doped crystal which, in its turn, could be due to increasing non stoichiometry degree resulting from the rising vacancy concentration.

It is seen from Fig. 2 that when the temperature is reduced below a characteristic value T^*, a deviation of $\rho_{ab}(T)$ from the linear dependence occurs, thus indicating the appearance of excess conductivity. According to recent studies, the excess conductivity is caused by the transition into the pseudo-gap regime (PG) [5–7]. There are two main scenarios for the onset of the pseudo-gap anomaly in HTSC systems. According to the first, the appearance of the PG is due to "dielectric" type, short-range order fluctuations, which occur in underdoped compounds (see, e.g., [5]). The second scenario is the formation of Cooper pairs already at temperatures substantially higher than the critical $T^* >> T_c$, with a subsequent establishment of their phase coherence at $T < T_c$ [6, 7]. As is shown in Table and in Fig. 2, for the aluminum doped crystal, the linear section of the $\rho_{ab}(T)$ dependence is significantly narrower as compared with the dopant-free crystal, and the temperature T^* is shifted towards higher temperatures for more than 55 K. This indicates the respectively expanded temperature range of excess conductivity existence. The value of $\Delta\sigma$ is usually determined from the equation:

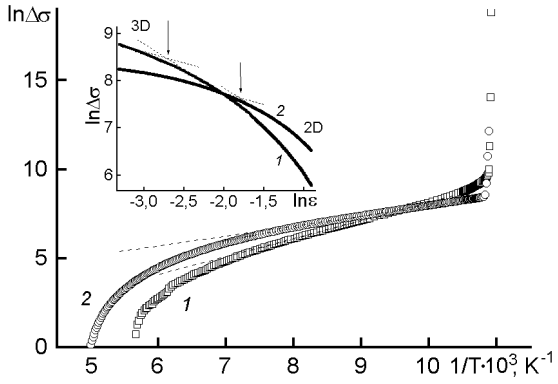

Fig. 3. Temperature dependences of excess conductivity in *ab*-plane for K1 and K2 crystals in $\ln(\Delta\sigma) - 1/T$ and $\ln(\Delta\sigma) - \ln\varepsilon$ coordinates (inset). The curves are numbered as in Fig. 2. The dashed lines in Fig. 3 show approximation of the experimental curves by Eq.(2) and in the inset, by the straight lines with $\mathrm{tg}\alpha \approx -0.5$ (3D-regime) and $\mathrm{tg}\alpha \approx -1.0$ (2D-regime). The 2D-3D crossover points are shown by arrows.

$$\Delta\sigma = \sigma - \sigma_0, \qquad (1)$$

where $\sigma_0 = \rho_0^{-1} = (A + BT)^{-1}$ is the conductivity value determined by extrapolating the linear section to zero temperature and $\sigma = \rho^{-1}$ is the experimental conductivity in the normal state. The measured $\Delta\sigma(T)$ dependences are presented in Fig. 3 in $\ln\Delta\sigma - 1/T$ coordinates. It is seen that in a rather wide temperature range these dependences are well approximated by the exponential relationship:

$$\Delta\sigma \sim \exp(\Delta^*_{ab}/T), \qquad (2)$$

where Δ^*_{ab} is a quantity defining a certain thermal activation process through an energy gap, "pseudo-gap". The $\Delta\sigma(T)$ exponential dependence was observed before in YBaCuO films [7]. As it was shown in [7], the experimental data approximation could be significantly extended by introducing the

$(1 - T/T^*)$ factor. In this case, the excess conductivity is proportional to density of superconducting carriers $n_s \sim (1 - T/T^*)$ and in inverse proportion to the number of pairs $\sim\exp(-\Delta^*/kT)$, destroyed by the thermal motion. Therewith, T^* is considered as the mean-field temperature of the superconducting transition, while the temperature range $T_c < T < T^*$, where the pseudo-gap state exists is defined by rigidity of the order parameter phase, which in its turn depends on the oxygen deficit or the dopant concentration. The quantity Δ^* obtained from (2) for our experimental samples is presented in Table. It is seen that the aluminum alloying results to a significant decrease of the absolute PG value ($\Delta^*_{K1}/\Delta^*_{K2} \approx 1.52$). It follows from Fig. 3 that $\Delta\sigma$ increases sharply near T_c. According to the Aslamazov-Larkin theoretical model [8], the excess conductivity near T_c is due to fluctuation pairing of the carriers. This contribution of pairs to the conductivity at $T > T_c$ for the two-dimensional (2D) and three-dimensional (3D) cases is described by the following relations:

$$\Delta\sigma_{2D} = \frac{e^2}{16\hbar d}\varepsilon^{-1}, \qquad (3)$$

$$\Delta\sigma_{3D} = \frac{e^2}{32\hbar\xi_c(0)}\varepsilon^{-1/2}, \qquad (4)$$

where $\varepsilon = (T - T_c)/T_c$; e, electron charge, $\xi_c(0)$, the coherence length along the **c** axis at $T \to 0$ and d is a characteristic size of the two-dimensional layer. In our case, T_c is defined as the critical temperature value corresponding to the maximum in the $d\rho_{ab}(T)/dT$ dependences, in the of the superconducting transition interval (Fig. 2(b)).

In the inset to Fig. 3, the $\Delta\sigma(T)$ dependences in $(\ln\Delta\sigma - \ln\varepsilon)$ coordinates are presented. It is seen that near T_c these dependences are approximated well by a straight line with a slope $\mathrm{tg}\alpha_1 \approx -0.5$, in accordance to the power index $-1/2$ in Eq.(4), thus evidencing the 3D character of fluctuating superconductivity within this temperature range. As the temperature further rises, the $\Delta\sigma$ decline rate increases appreciably ($\mathrm{tg}\alpha_2 \approx -1$). This points to the FC dimensionality change. As it follows from Eqs.(3) and (4), at the 2D-3D crossover point we have:

$$\xi_c(0)\varepsilon_0^{(-1/2)} = d/2. \qquad (5)$$

In this case, having determined the ε_0 value and using literature data on the interplane spacing dependence on δ [9] ($d \approx 11.7$ Å), we can calculate the coherence length values $\xi_c(0)$. The calculations have shown that at doping with Al, the coherence length $\xi_c(0)$ changes from $\xi_c(0) = 1.48$ Å for YBaCuO to $\xi_c(0) = 2.32$ Å for the $YBa_2Cu_{3-y}Al_yO_{7-\delta}$ crystal, and the 2D-3D crossover point temperature becomes shifted significantly (see Table and Fig. 3 (b)).

Thus, the increasing electroresistance in the linear section of $\rho_{ab}(T)$ dependences and changing ratio $\rho_{ab}(300\ \mathrm{K})/\rho_{ab}(0\ \mathrm{K})$ in cases of partial Cu substitution by Al evidence the increasing number of effective current carrier scattering centers. The excess conductivity $\Delta\sigma(T)$ of YBCO and $YBa_2Cu_{3-y}Al_yO_{7-\delta}$ single crystals answers within a wide temperature range $T_f < T < T^*$ to exponential temperature dependence and near T_c it can be approximated by the Aslamazov-Larkin theoretical model. The low doping with aluminum (up to $y \approx 0.05$) results in the expanded temperature interval of PG regime existence, thereby narrowing the linear section of the $\rho(T)$ dependence in *ab*-plane.

References

1. P.H.Hor, J.Bechtold, Y.Y.Xue, et al., *Physica C*, **185–189**, 2311 (1991).
2. R.B.Van Dover, L.F.Schneemeyer, J.V. Waszczak et al., *Phys. Rev. B*, **39**, 2932 (1989).
3. M.A.Obolenskii, A.V.Bondarenko, V.I. Beletskii, et al., *Fiz. Nizk. Temp.*, **16**, 1103 (1990).
4. G.Lacayo, G.Kastner, R. Herrmann, *Physica C*, **192**, 207 (1992).
5. M.V.Sadovskii, *Usp.Fiz.Nauk*, **171**, 515 (2001).
6. P.Pieri, G.C.Strinati, D.Moroni, *Phys. Rev. Lett.*, **89**, 127003 (2002).
7. D.D.Prokofyev, M.P.Volkov, Yu.A.Boyko, *Fiz. Tverd. Tela*, **45**, 1168 (2003).
8. L.G.Aslamazov, A.I.Larkin, *Fiz. Tverd. Tela*, **10**, 1104 (1968).
9. G.D.Chryssikos, E.I.Kamitsos, J.A.Kapoutsis, et al., *Physica C*, **254**, 44 (1995).

Надлишкова провідність і псевдощілинне становище у монокристалах $YBa_2Cu_{3-y}Al_yO_{7-\delta}$ з системою односпрямованих двійникових меж

Р.В.Вовк, М.О.Оболенський, О.В.Бондаренко

Досліджено провідність у базисній площині монокристалів $YBa_2Cu_{3-y}Al_yO_{7-\delta}$ із заданою топологією площинних дефектів. Показано, що внесення домішок Al викликає зростання кількості ефективних центрів розсіювання нормальних носіїв. Надлишкова провідність досліджених зразків у широкому інтервалі температур відповідає експоненціальній температурній залежності, а поблизу T_c задовільно описується теоретичною моделлю Асламазова-Ларкіна. При цьому часткова заміна Cu на Al приводить до значного розширення температурного інтервалу існування псевдощілинної аномалії в *ab*-площині.