Functional Materials 19, No.2 (2012)

Anisotropy of magnetoresistance in untwinned $YBa_2Cu_3O_{7-\delta}$ single crystals

R.V.Vovk, Z.F.Nazyrov, V.V.Kruglyak*, F.Y.Ogrin*

Kharkov National University, 4 Svoboda Sq., 61077 Kharkiv, Ukraine
*University of Exeter, Stocker road, Exeter, EX4 4QL, United Kingdom

Received January 25, 2012

In this work we study an influence of the disorientation angle between direction of constant magnetic field up to 15 kOe and *ab*-plane $\alpha \equiv \angle(\mathbf{H},ab)$ on the temperature dependences of the excess conductivity in the temperature interval of transition to superconducting state in untwinned $YBa_2Cu_3O_{7-\delta}$ single crystals with a small oxygen hypostoichiometry. At temperature $T > T_c$, the temperature dependence of the excess para-conductivity is interpreted within the Aslamazov-Larkin theoretical model of the fluctuation conductivity for layered superconductors. The cause of appearances of low temperature "tails" (para-coherent transitions) on the resistive transitions, corresponding to different phase regimes of the vortex-matter state is discussed.

Исследовано влияние угла разориентации между направлением постоянного магнитного поля $H = 15$ кЭ и направлением базисной *ab*-плоскости $\alpha \equiv \angle(\mathbf{H},ab)$ на температурные зависимости избыточной проводимости в области переходов в сверхпроводящее состояние раздвойникованных монокристаллов $YBa_2Cu_3O_{7-\delta}$ с малым отлонением от кислородной стехиометрии. Установлено, что при температурах $T > T_c$ температурные зависимости избыточной парапроводимости интерпретируются в рамках теоретической модели флуктуационной проводимости Асламазова-Ларкина для слоистых сверхпроводящих систем. Обсуждаются причины появления низкотемпературных хвостов (паракогерентных переходов) на резистивных переходах, соответствующих различным режимам фазового состояния вихревой материи.

1. Introduction

Creation of new functional materials with high current-carrying capacity remains one of the actual applied and fundamental problems of high-temperature superconductivity (HTSC) physics. An optimization of defective ensemble plays the main role in this [1]. A small coherence length ξ and a large penetration depth λ result in effective pinning in HTSC on small-scale defects, including oxygen vacancies and the introduction of impurities [2–4]. The impact of such defects on the phase state of the vortex matter is often difficult to explain due to the presence in the HTSC intergranural boundaries, twin boundaries (TB), clusters of inclusions and other defects which are powerful pinning centers. The presence "intrinsic" pinning due to the layered structure of HTSC compounds is also significantly affected [3].

In the present study we investigated magnetic conductivity in untwined $YBa_2Cu_3O_{7-\delta}$ single crystals under a fixed magnetic field and different values of angle α — between the magnetic field **H** and the *ab*-plane $(\mathbf{H},ab)$. Using as samples untwined single crystals we eliminated an influence of intergranural boundaries and TB allowing the selected geometry of the experiment to control the changes of the contribution of intrinsic pinning.

2. Experimental

In $YBa_2Cu_3O_{7-\delta}$ compounds it is relatively easy to obtain a given concentration of point defects by varying oxygen stoichiometry [5]. Therefore, measurement of resistive transitions to the superconducting state, which are sensitive to such concentration changes, allows to investigate an impact of the point defects on the phase state and on the dynamics of vortex matter. This is achieved by analyzing the fluctuation to the conductivity that was observed in HTSC compounds at temperatures near the critical temperature ($T \approx T_c$) [6].

$YBa_2Cu_3O_{7-\delta}$ single crystals were grown in a gold crucible using solution-melting method, with the methodology described previously [5]. The characteristic dimensions of the samples were 2.4×1.5×0.03 mm^3. $YBa_2Cu_3O_{7-\delta}$ oxygen saturating regime leads to the tetra-ortho structural transition that in its turn results in the crystal twinning in order to minimize its elastic energy. To obtain untwined samples we used a special cell at 420°C and pressure 30–40 GPa, in accordance with the procedure of Giapintzakis et al.[7]. To obtain homogeneous oxygen content the crystal was annealed again in oxygen flow at the temperature of 420°C for seven days.

Thereafter, they were annealed at the temperature of 200°C in oxygen atmosphere for three hours. This methodology results in contacts with resistance than 1 Ohm and allows measurements with a current value of 10 mA in the *ab*-plane. To form electric contacts the standard four-contact scheme was used. In this, silver paste was applied onto the crystal surface and the connection of silver conductors (with diameter 0.05 mm). All the measurements were performed in temperature drift mode using the method for two opposite directions of the transport current. This effectively eliminates the impact of the parasitic signal. A platinum thermo-resistor was used to monitor the temperature, whereas the voltage was measured across the sample and the reference resistor with V2-38 nano-voltmeters. Data from the voltmeters interface is automatically transferred to computer.

The critical temperature (T_c) was defined as the temperature corresponding to the main maximum in the $d\rho_{ab}(T)/dT$ dependence in the superconductive transition similarly to previous studies [8]. To produce oxygen hypostoichiometric samples, the crystal was annealed in oxygen flow, at the temperature of 620°C for 48 h. The measurements were performed 7 days after annealing to avoid the influence of the relaxation effects. The magnetic field at 15 kOe was created by an electromagnet, which could vary the field orientation relative to the crystal. An accuracy of the field orientation relative to the sample was better than 0.2°. A sample was mounted in the measuring cell so that the vector field **H** was always perpendicular to the vector of the transport current **j**.

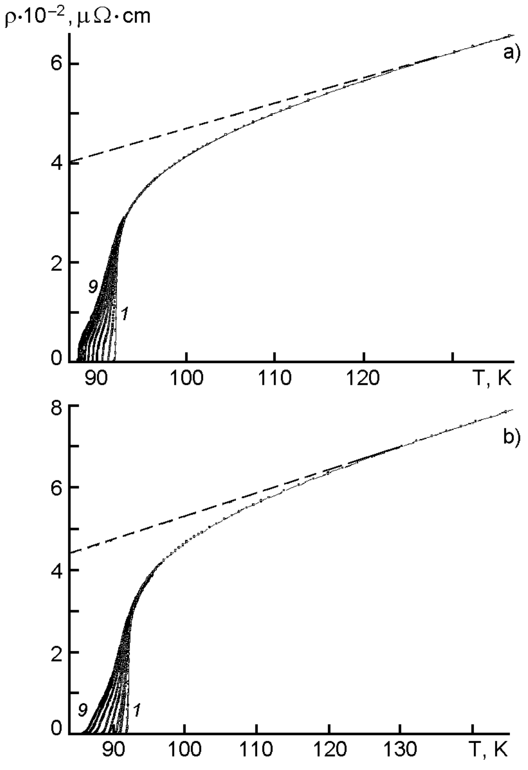

Fig. 1. Temperature dependences of resistivity in basal *ab*-plane $\rho_{ab}(T)$ for $YBa_2Cu_3O_{7-\delta}$ single crystal before (a) and after (b) lowering oxygen content, measured under magnetic field $\mathbf{H} = 0$ (curve *1*) and for $\mathbf{H} =$ 15 KOe for different angles $\alpha \equiv \angle(\mathbf{H},ab)$: 0; 5; 10; 20; 30; 45; 60 and 90° (curves *2–9* respectively). The dotted lines in the figure shows a linear extrapolation of the plots in the low temperature region.

For investigations of the resistive transitions in superconducting (SC) state we used the Kouvel-Fischer method [9]. This is based on the value analysis of $\chi = -d(\ln\Delta\sigma)/dT$, where $\Delta\sigma = \sigma - \sigma_0$ is the excess conductivity, which arises in the conducting subsystem due to the fluctuation pairing of carriers at $T > T_c$ [10] and determined by the phase state of vortex matter at $T < T_c$ [6]. Here $\sigma = \rho^{-1}$ is the experimentally measured value of conductivity, and $\sigma_0 = \rho_0{}^{-1} = (A + BT)^{-1}$ is a term, determined by extrapolating the high-temperature linear segment up to the area of the SC transition.

Assuming that $\Delta\sigma$ diverges as $\Delta\sigma$ $(T - T_c)^{-\beta}$ at $T \approx T_c$, from the derivative $\chi = -d(\ln\Delta\sigma)/dT$ it follows that $\chi^{-1} = \beta^{-1}(T - T_c)$, where β is an indicator that depends on the dimension and the phase state of the fluctuation and vortex subsystems [6, 10]. Thus, the identification of linear temperature dependence $\chi^{-1}(T)$ at the same time allows the determination of important dimensional parameters and characteristic temperatures of dynamic phase transitions in the SC carriers subsystem.

3. Results and discussion

Fig. 1 shows the temperature dependences of resistivity in the basal *ab*-plane $\rho_{ab}(T)$ measured under $\mathbf{H} = 0$ (curve *1*) and for a fixed magnetic field of $\mathbf{H} = 15$ KOe for different angles $\alpha \equiv \angle(\mathbf{H},ab)$ (curves *2–9*) before (a) and after (b) lowering the oxygen content.

As can be seen from the Fig. 1 with the temperature lowering from 300 K, as in the case of optimally doped as oxygen underdoped samples $\rho_{ab}(T)$ decreases almost linearly up to a certain characteristic temperature $T^* \approx 134$ K. Below this temperature it begins the systematic deviation of experimental points down from the linear dependence indicating the appearance of excess conductivity $\Delta\sigma$, as discussed above. According to the modern concepts such behavior of dependence $\rho_{ab}(T)$ at temperatures $T > T_c$ is conditioned by the so-called "pseudogap anomalies" (PG), which is discussed in more detail in [11]. It should be also noted that an application of a magnetic field and change the angle α at temperatures $T > 1.15T_c$ within experimental error have not effect on the behavior of the curves $\rho_{ab}(T)$, both leading to a significant broadening of the superconducting transition itself in comparison with the sharp ($\Delta T_c \approx 0.3$ K) transition observed at $H = 0$. In this case it is clear that there is a significant difference in the forms of themselves resistive transitions in superconducting state between ones measured in a magnetic field for the stoichiometric (a) and nonstoichiometric (b) composition of the sample. While in the first case on the tail of the superconducting transition there is a sharp "kink" in the second case — a monotone smoothing of low-temperature resistive transition takes place. The latter is reflected in the actual disappearance of the low-temperature peak in the temperature dependences of the derivative $d\rho_{ab}(T)/dT$ (Fig. 2b) in the underdoped sample. As can be seen from Fig. 2a, in the case of the original, stoichiometric composition of the sample such a peak is present for all values of α and its height increases with increasing α values. According to [3] an appearance of such features in the temperature dependences $\rho_{ab}(T)$ and $d\rho_{ab}(T)/dT$ shows the implementation in the first order phase transition corresponding to melting of the vortex lattice. A disappearance of these features in the case of underdoped experimental sample may indicate suppression of the transition. At the same time one should pay attention to the fact that in the second case the resistive transitions lie in some single universal dependence passing down the slope to the left of the main high-temperature peak depends $d\rho_{ab}(T)/dT$. According to [6], the temperature corresponding to this peak corresponds to the critical temperature in the mean-field approximation T_c^{mf}. This, in turn, may in-

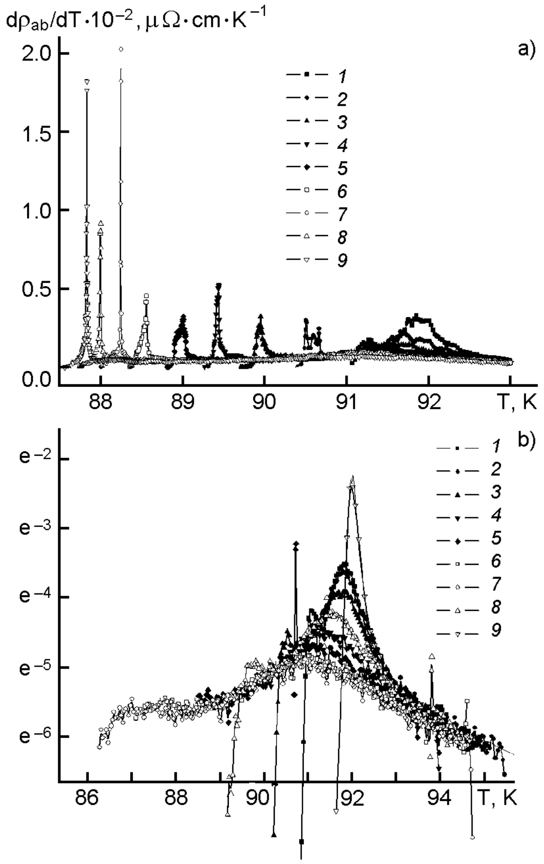

Fig. 2. Resistivity transitions to SC state for $YBa_2Cu_3O_{7-\delta}$ single crystal in $d\rho_{ab}/dT - T$ coordinates. The numbering of the curves is consistent to Fig. 1.

dicate that the implementation of the system of a new state of the conducting subsystem.

Fig. 3 shows the resistivity transitions to superconducting (SC) state in $[-d(\ln\Delta\sigma)/dT]^{-1} - T$ coordinates. In the inset of Fig. 3b the curve obtained for $\alpha = 60°$ is presented. This shows the characteristic temperatures T_{c0}, T_M and T_c corresponding to the end of the resistive transition in SC state, the melting point of the vortex lattice and the critical temperature in the mean-field approximation respectively [3, 6, 8]. In all curves in the high-temperature area of the SC transition we observe a linear area with slope $\beta \approx 0.5$ which according to [10] indicates realization of a three-dimensional (3D) regime of fluctuation carriers' existence in the system. At the same time the section corresponding to 3D regime is essentially unstable in the magnetic field, which is consistent with the results obtained in [6]. When increasing the temperature from T_c upwards an increase of the absolute value of β occurs, that suggests the realization of a 3D–2D crossover in the system [5, 11].

Application of magnetic field and increase of the angle α lead to a significant transformation of the form of the SC-transition, which expressed in the appearance of an additional low-temperature peak, so-called "para-coherent transition". It should be noted that there is a noticeable difference in the behavior of this maximum in the case of underdoped and stoichiometric compositions. While for the optimally doped sample with oxygen this transition is shifted toward lower temperatures without changing its shape. In the second case this shift is accompanied by a significant simultaneous increase of amplitude and width of the peak with increasing angle α. This peak rapidly shifts downwards to lower temperatures as the angle α increases with a simultaneous increase of width and amplitude of the peak. Similar behavior may be due to a decrease with an increase in the proportion α own intrinsic pinning, and thus increasing the role of bulk pinning due to formation in the volume of the sample of strong pinning centers.

It was established [2, 3] that the presence of strong pinning centers in the system leads to a spreading of the kink and the transition from an ordered vortex-lattice phase to a phase, so-called "vortex" or "Bragg" glass in which the vortex system is accommodated in the chaotic pinning's potential. This chaotic pinning potential violates the long-range order of vortex lattice thereby suppressing the first-order phase transition and results to formation of glassy state of vortices. In the resistive transitions appear "tails" whose amplitude is less than the resistance of viscous flow ρ_{ff}. These are probably due to a partial pinning of the vortex liquid.

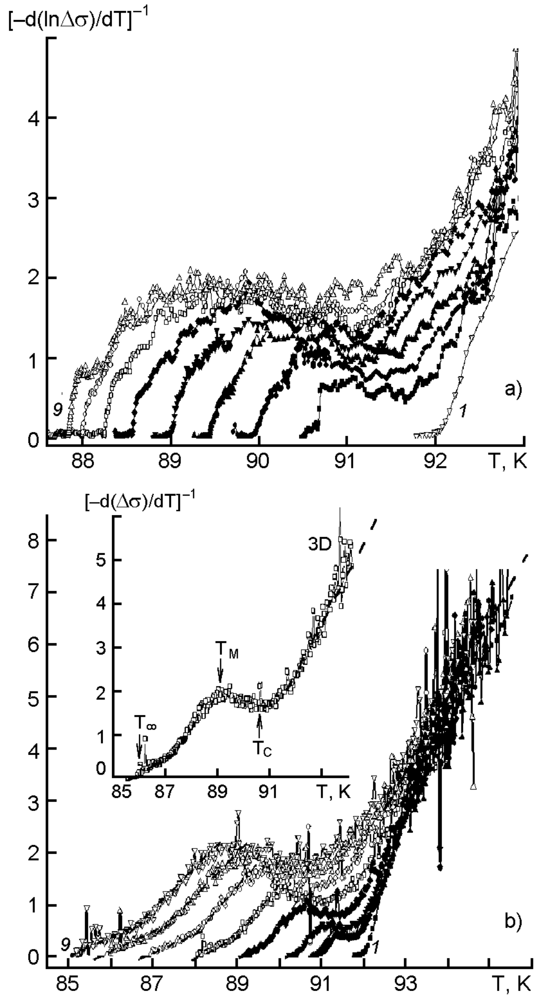

Fig. 3. Resistivity transitions to SC state for $YBa_2Cu_3O_{7-\delta}$ single crystal in $[-d(\ln\Delta\sigma)/dT]^{-1} - T$ coordinates. The numbering of the curves is consistent to Fig. 1. In the inset of Fig. 3 the curve obtained for $\alpha = 60°$. The dash lines correspond to the extrapolation of the areas corresponding to various FC regimes. Arrows show the characteristic temperatures T_{c0}, T_M and T_c, corresponding to the end of the resistive transition in SC state, the melting point of the vortex lattice and the critical temperature in the mean-field approximation, respectively.

Clusters of oxygen vacancies could have a significant role. In previous works, the influence of annealing at room temperature on excess conductivity for the same samples has investigated [11]. Immediately after annealing in oxygen atmosphere at temperature of 500°C the crystal had a critical temperature $T_c \cong 91.75$ K with a transition width $\Delta T_c \cong 0.3$ K. Then, the crystal was retained at room temperature for a week. As it was shown previously, this resulted in decrease of electrical resistance in the normal state $\rho(300\ K)$ of ≈3 % and an increase of $T_c \approx 0.25$ K. These changes are consistent with the concept of formation of clusters of oxygen vacancies in the process of ordering the vacancy subsystem [11]. This implies an increase of oxygen concentration in the volume of the crystal and a decrease of oxygen content in the volume of the clusters. In turn, a decrease of the concentration of carriers scattering centers in the volume of the crystal reduces the resistance $\rho(300\ K)$.

Taking into consideration the dome-shaped dependence T_c from carrier concentration with a maximum value of $T_c \cong 93$ K at $\delta \cong 0.93$ [12] we can assume that the redistribution of the labile oxygen leads to phase separation in the crystal and to formation of phase clusters with different value of T_c. Considering the presence of percolation paths of current flow on the main volume of the crystal this process should lead to an increase of the measured value of T_c. We presuming that in the single crystal of the present work there coexists a point pinning potential created by the isolated oxygen vacancies and a volume pinning potential with a suppressed superconducting order parameter formed by the clusters of oxygen vacancies.

As was determined in [6] that in the case of "Bragg glass" state the dependence $\chi(T)$ should be observed scaling in $\chi(T_c - T_{c0})$ – $(T - T_{c0})/(T_c - T_{c0})$ coordinates. In this, T_{c0} is the critical temperature of the transition in para-coherent area determined at the point of intersection with the linear interval approximating the so-called para-coherent area with the axis of temperature. T_c is the temperature corresponding to the mean-field critical temperature determined as the maximum in the curves $d\rho_{ab}(T)/dT$ [8].

Fig. 4 shows these curves scaled as $\chi(T_c - T_{c0})/\varepsilon_\alpha$ – $(T - T_{c0})/(T_c - T_{c0})$. We took into consideration changing the self pinning's contribution with an increase of the disorientation angle $\alpha \equiv \angle(\mathbf{H},ab)$ by introducing the reduced value $\chi(T_c - T_{c0})$ accounting the anisotropy parameter [2] $\varepsilon_\alpha = (\sin^2\alpha + \cos^2\alpha)^{1/2}$, where $\varepsilon = 6 \div 9$. In Fig. 4, the best scaling in the experimental curves is observed in the para-coherent area at $T < T_M$. At higher temperature spread of the curves becomes significant and this is due to influence of the pinning of superconducting fluctuations on the cluster inclusions as well as possible to strengthen the role of some specific mechanisms of the quasiparticle interaction [13, 14].

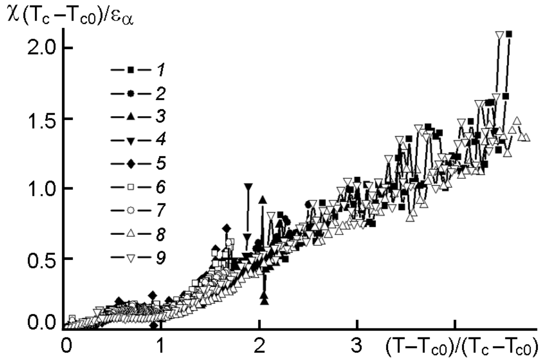

Fig. 4. Resistivity transitions to SC state for $YBa_2Cu_3O_{7-\delta}$ crystal in (reducing) $\chi(T_c - T_{c0})/\varepsilon_\alpha$ – $(T - T_{c0})/(T_c - T_{c0})$ coordinates. The numbering of the curves is consistent to Fig. 1.

4. Conclusions

To conclude, the FC near T_c is described by 3-dimensional Aslamazov-Larkin model for layered superconducting systems. Application of constant magnetic field to $YBa_2Cu_3O_{7-\delta}$ single crystals with a small oxygen hypostoichiometry, in distinction to similar stoichiometric samples, leads to degradation of the additional para-coherent transition in the excess conductivity temperature dependences in basic *ab*-plane in the area of resistive transition to SC state. Increasing the angle $\alpha \equiv \angle(\mathbf{H},ab)$ in case of underdoped by oxygen sample leads to synchronous increasing of amplitude and width of the peak corresponding to this transition, and its shift to the area of lower temperatures. This can be due to a decrease of the self intrinsic pinning of the vortex subsystem contribution with an increase of α and strengthening the role of volume pinning due to presence of the clusters of oxygen vacancies in the experimental sample. Consequently, at temperatures below the critical $T < T_c$, there is a suppression of the dynamic phase transition type liquid vortex-vortex lattice and the formation of transition in the system type liquid vortex-vortex "Bragg" glass.

This work was supported in part by European Commission CORDIS Seven Framework Program, Project No. 247556.

References

1. A.I.Chroneos, I.L.Goulatis, R.V.Vovk, *Acta Chim. Slov.*, **54**, 179 (2007).
2. A.V.Bondarenko, A.A.Zavgorodniy, D.A.Lotnik et al., *Low Temp. Phys.*, **34**, 645 (2008).
3. W.K.Kwok et al., *Phys. Rev. Lett.*, **69**, 69, 3370 (1992)
4. A.A.Zavgorodniy, R.V.Vovk, M.A.Obolenskii, A.V.Samoilov, *Low Temp. Phys.*, **36**, 143 (2010).
5. R.V.Vovk, M.A.Obolenskii, A.V.Bondarenko et al., *J. Alloys Compd.*, **464**, 58 (2008).
6. R.M.Costa, I.C.Riegel, A.R.Jurelo, J.L.Pimentel Jr., *J. Magn. Magn. Matter.*, **320**, 493 (2008).
7. J.Giapintzakis, D.M.Ginzberg, P.D.Han, *J. Low Temp. Phys.*, **77**, 155 (1989).
8. R.V.Vovk et al., *Philosophical Magazine*, **91**, 2291 (2011).
9. J.S.Kouvel, M.E.Fischer, *Phys. Rev.*, **136**, 1616 (1964).
10. L.G.Aslamasov, A.I.Larkin, *Fizika Tverdogo Tela*, **10**, 875 (1968).
11. R.V.Vovk, M.A.Obolenskii, A.A.Zavgorodniy et al., *Low Temp. Phys.*, **33**, 931 (2007).
12. J.L.Tallon, C.Berbhard, H.Shaked et al., *Phys. Rev. B*, **51**, 12911 (1995).
13. V.M.Apalkov, M.E. Portnoi, *Phys. Rev. B*, **66**, 121303 (2002).
14. P.G.Curran, V.V.Khotkevych, S.J.Bending et al., *Phys. Rev. B*, **84**, 104507 (2011).

Анізотропія магнітопровідності у роздвійникованих монокристалах $YBa_2Cu_3O_{7-\delta}$

Р.В.Вовк, З.Ф.Назиров, В.В.Кругляк, Ф.Я.Огрін

В роботі досліджено вплив кута разорієнтації між напрямком постійного магнітного поля $H = 15$ кЕ і напрямком базисної ab-площини $\alpha \equiv \angle(\mathbf{H},ab)$ на температурні залежності надлишкової провідності в області переходів в надпровідний стан роздвійникованих монокристалів $YBa_2Cu_3O_{7-\delta}$ з малим відхиленням від кисневої стехіометрії. Встановлено, що при температурах $T > T_c$ температурні залежності надлишкової парапровідності інтерпретуються в рамках теоретичної моделі флуктуаційної провідності Асламазова-Ларкіна для шаруватих надпровідних систем. Обговорюються причини появи низькотемпературних "хвостів" (паракогерентних переходів) на резистивних переходах, що відповідають різним режимам фазового стану вихорової матерії.