Effect of annealing on a pseudogap state in untwinned YBa$_2$Cu$_3$O$_{7-\delta}$ single crystals

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The effect of annealing both in the oxygen atmosphere and at room temperatures on physical properties such as the pseudogap ($\Delta^*(T)$) and excess conductivity ($\sigma'(T)$) of untwinned YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) single crystal with a small deviation from oxygen stoichiometry is studied. It was revealed that as the charge carrier density, $n_c$, increases, $T_c$ also slightly increases, whereas the temperature of the pseudogap opening, $T^*$, decreases noticeably, which is consistent with the phase diagram (PD) of cuprates. The excess conductivity in the vicinity of $T_c$ is represented by the Aslamazov-Larkin and Hikami-Larkin fluctuation theories, illustrating the three-dimensional to two-dimensional (i.e., 3D-2D) crossover with an increase in temperature. The crossover temperature $T_c$ determines the coherence length along the $c$ axis is $\xi_c(0) = 0.86 \text{Å}$, that is 2.6 times larger than for optimally doped YBCO single crystals with defects. Taking into account the short coherence length in high-temperature superconductors, in the model of free charge carriers the phase relaxation time of fluctuating Cooper pairs is determined, $\tau_{\varphi} (100 \text{K}) = (4.55 \pm 0.4) \cdot 10^{-13} \text{s}$, which is slightly (1.2 times) larger than in well-structured YBCO films, and as in films, does not depend on $n_c$. It is shown that $\Delta^*(T)$ at different annealing stages practically does not change its shape. As in the well-structured YBCO films, $\Delta^*(T)$ demonstrates maximum at $T_{\text{pair}} \approx 124 \text{K}$ which depends weakly on $n_c$. However, the maximum value of $\Delta^*(T_{\text{pair}})$ increases with increasing $n_c$ as it follows from the PD of cuprates. Comparing the experimental data with the Peters-Bauer theory we estimated the density of local pairs $<n \cdot n_1> \approx 0.3$ near $T_c$ that is a common value for high-temperature superconductors.

The mechanism of superconducting pairing in high-temperature superconductors (HTSCs), which makes it possible to obtain paired fermions at temperatures as higher as ~100 K, remains rather debatable$^1$–$^9$. To clarify the issue, the study of superconducting (SC) fluctuations has attracted considerable attention in the research of HTSCs$^{10–12}$ (and references therein). This because it is related to the nature of the pseudogap (PG), which is known to open in underdoped cuprates at a temperature $T^*$, much above the superconducting transition temperature $T_c$.$^{2–5,8–11}$ Above $T^*$ the dc resistivity, $\rho(T)$, of optimally doped (OD) and moderately underdoped cuprates is known to be linear$^{11}$. In the framework of the Nearly Antiferromagnetic Fermi Liquid model$^{14}$ it was proven that this linearity is a specific feature of HTSCs that is represented by the stability of the Fermi surface (FS). Notably, at $T < T^*$ not only all the properties of HTSCs measured by various experimental methods$^{13}$ change, but the density of electronic states at the Fermi level begins to decrease$^{16,17}$, which by definition is called a pseudogap$^{11,18}$. It is also assumed that below the PG temperature $T^*$ the rearrangement of the FS may begin$^{2,3,18,19}$. Accordingly, at $T^*$, the resistivity curve deviates downward from the linearity leading to excess conductivity $\sigma'(T)$ as the difference between determined conductivity $\sigma(T) = 1/\rho(T)$ and extrapolated normal-state conductivity $\sigma_0(T) = 1/\rho_0(T)$,$^{15,11,20,21}$

Importantly, the SC fluctuations are responsible for a relatively short part of the entire excess conductivity, ~15 K above $T_c$, which is thus called the fluctuation conductivity (FLC)$^{10–12,20}$ (and references therein). In such a scenario, the long-range coherence is lost at $T_c$ due to fluctuations of the phase of the superconducting order parameter$^{2,22,23}$. The FLC region would then be marked by (prefomed) fluctuating Cooper pairs (FCPs), which obey the Aslamasov-Larkin (AL)$^{24}$ and Hikami-Larkin (HL) (Maki-Thompson (MT) term)$^{25}$ fluctuation theories, and take the role of the precursor to the SC state.$^{1,26}$ In the FLC region, the FCPs behave in a good many way
The gradient was calculated by approximating the experimentally derived curves and confirmed the linear behavior. As mentioned above, the PG temperature $T_c$, the in-plane coherence length, $\xi_{ab}(T)$, which determines the pair size, decreases. At the same time, the bonding energy of the LPs, $\delta_b \sim 1/\xi_{ab}^2$ [27–29], noticeably increases. As a result, the LPs should change their properties in HTSCs as $T$ increases, which was observed experimentally [11, 22].

Nevertheless, there is still the question of the pairing mechanism, which allows the existence of bound fermions at temperatures significantly higher than $T_c$. Obviously, in HTSCs, in addition to the electron–phonon interaction, some other mechanism of interaction, most likely of a magnetic type should act (and references therein). As a result, it is proposed to consider both spin-density waves (SDW) [2, 3, 18], charge-density waves (CDW) [2–5] and charge order (CO) [19] (and references therein) in order to explain the pairing mechanism of HTSCs in the PG state. However, in YBCO, the CO onset temperature appears always to be lower than $T_c$ [19] and the temperature ranges and density of charge carriers indicated in the studies that assume the existence of the SDW, CDW and CO mechanisms differ significantly. As a result, the proposed new phase diagrams of cuprates are also very different [1, 3, 5, 8, 18, 19]. Thus, despite the tremendous efforts towards this end there is still no consensus on the physical nature of the PG (refer to [11, 18, 19, 28–32] and references therein).

It is well established now, that all properties of HTSC cuprates are determined by the density of charge carriers, $n_f$, which can vary over a wide range upon doping [1, 2, 3, 5, 10–13, 18–21]. In $\text{YBa}_2\text{Cu}_3\text{O}_7$–$\delta$, $n_f$ changes as a result of oxygen intercalation, and the maximum $T_c \sim 92$ K corresponds to a stoichiometric material (i.e. $\delta = 0$) [23]. To transfer YBCO to the so-called overdoped regime, it is necessary to use Ca doping [1]. Usually a set of samples with different $n_f$ is used for measurements [10, 11, 15–20]. In the manufacture of HTSC films, each $n_f$ value is determined by the manufacturing conditions (usually the oxygen pressure in the chamber) of each specific sample [9, 20]. As a result, film samples may differ in their structure, number of defects etc. The advantage of single crystals is that their $n_f$ can vary noticeably during annealing of the sample in an oxygen atmosphere [15, 20]. However, in the case of a strong change in $n_f$, various defects may also appear in the samples [21, 35] (cf. the thermodynamic parameters of defect processes in these high $T_c$ superconductors have been found to obey the thermodynamical $\mathbf{cB}\Omega$ model -where $c$ stands for a dimensionless factor that may be considered in a first approximation to be independent of temperature and pressure, $B$ is the isothermal bulk modulus and $\Omega$ the mean volume per atom [36, 37] - in a similar fashion as in the case of the so-called superionic conductors e.g. $\beta$-$\text{PbF}_2$) [38]. Therefore, it seems highly desirable to find out how the properties of the same sample, first of all FLC and PG, can change, if somehow the density of charge carriers in the sample varies in a relatively small range.

In the present study we take advantage of single crystals to study FLC and PG in untwinned YBCO single crystal with $n_f$ close to optimal doping ($T_c = 91.6$ K), when $n_f$ changes upon annealing in an oxygen atmosphere. We have studied three samples with different $n_f$. For a short notation, we name these samples A1, A2 and A3 (Table 1 and experimental methods). The fluctuation contributions to $\sigma'(T)$ were derived from the dc resistivity $\rho(T)$ measurements, and temperature dependences of PG, $\Delta \rho(T)$, as a function of $n_f$ were calculated. The results show that in the range of SC fluctuation near $T_c$, $\sigma'(T)$ is adequately interpreted by the AL and HL fluctuation theories. It was determined that $\Delta \rho(T)$, as expected, increases upon annealing and is in good agreement with the Peters–Bauer theory (PB) near ($T_c$). The implications of these findings will be discussed.

### Table 1. Determined parameters.

| Sample | $\rho(100)$ (µΩcm) | $T_c$ (K) | $\Delta m$ (µm) | $T_{\alpha}$ (K) | $T_{\beta}$ (K) | $T_{\gamma}$ (K) | $T_{\delta}$ (K) | $\Delta T$ (K) | $d_0$ (Å) | $\xi_{ab}$ (Å) | $C_{3D}$ |
|--------|------------------|---------|----------------|-----------------|----------------|----------------|----------------|--------------|----------|----------------|--------|
| A1     | 47.2             | 91.6    | 91.84          | 91.90           | 92.34          | 97.4           | 5.5            | 3.50         | 0.86     | 1.34           |        |
| A2     | 47.0             | 91.7    | 91.93          | 91.90           | 92.39          | 96.9           | 4.9            | 3.54         | 0.84     | 1.25           |        |
| A3     | 46.3             | 91.9    | 91.98          | 92.02           | 92.42          | 97.3           | 5.3            | 3.42         | 0.81     | 1.27           |        |

Results and Discussion

**Resistivity.** The dependence of the temperature of the resistivity (i.e. $\rho(T) = \rho_0(T)$) of untwinned $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ crystals are shown in Fig. 1. As $\rho(T)$ of the samples differ insignificantly, to simplify the figure on the upper panel (Fig. 1a), only the resistive curve of sample A1 is shown. The corresponding dependences of $\rho(T)$ for all three samples are shown as a three-dimensional (3D) graph in Fig. 1b. For temperatures above $T^* = (185 \pm 0.5)$ K (A1), $(182 \pm 0.5)$ K (A2), $(179 \pm 0.5)$ K (A3) and up to 300 K, all the $\rho(T)$ dependences are linear and are described by a gradient $a = d\rho/dT = 0.484$ (A1), 0.488 (A2) and 0.478 (A3) µΩ/cm/K, which slightly changes with annealing. The gradient was calculated by approximating the experimentally derived curves and confirmed the linear behavior of $\rho(T)$ with a mean-root-square error of 0.009 ± 0.002 in the specified temperature range for all samples. As mentioned above, the PG temperature $T^*$ was defined as a temperature at which the resistive curve deviates downward from the linearity (Fig. 1). The more precise approach to determine $T^*$ with accuracy $\pm 1$ K is to explore the criterion $|\rho(T) - \rho_0|/dT = 1$ (insert in Fig. 1a), where $a$ designates the slope of the extrapolated normal-state resistivity, $\rho_0(T)$, and $\rho_0$ is its intercept with the $y$ axis. Both methods give the same $T^*$.

In the process of annealing, with increasing the oxygen content, $T_c$ slightly increases, and $\rho(T)$ slightly decreases (Fig. 1b). This is not surprising, since the samples are actually on top of the PD. At the same time, $T^*$ decreases much more perceptibly, in full agreement with the PD of cuprates [5, 13, 14, 21] (and Tables 1, 2 and 3). The main difference between the untwinned YBCO and single crystal containing defects, presumably in the form of
twin boundaries (TB)\textsuperscript{21,40}, is much higher $T^*$ value. Usually, in optimally doped YBCO single crystals with $T_c \sim 91.1$ K, but containing defects in the form of TB, $T^* \sim 140$ K\textsuperscript{41}. It is assumed that the defects prevent the establishment of phase coherence of LPs (paired fermions) and, thus, effectively reduce $T^*$\textsuperscript{21,42}. At the same time, in well-structured YBCO films\textsuperscript{11,20}, a sample with $T_c \sim 88$ K has $T^* \sim 200$ K, which is much closer to $T^* = 185$ K, observed for the untwined YBCO single crystal A1 with $T_c = 91.6$ K. Therefore, it can be assumed that, by their properties, YBCO single crystals, which do not contain TB, are closer to well-structured films. This conclusion is supported by the analysis of the results of the FLC and PG study.

Fluctuation conductivity. In the resistive measurements, PG is evident as a deviation of the resistivity $\rho_{ab}(T) = \rho(T)$, determined in the $ab$ plane, from a linear dependence at high temperatures to smaller values (refer to Fig. 1). The result is excess conductivity expressed by $\sigma(T) = \sigma(T) - \sigma_{ab}(T) = [1/\rho(T)] - [1/\rho_{ab}(T)]$, or

| Sample | $R_{ab}(100$ K) \text{($10^{-4}$ m$^2$/C)} | $\rho(100$ K)$\times10^{-3}$ (µΩ cm) | $n_0$ (10$^{15}$ cm$^{-2}$) | $n_f$ (10$^{18}$ cm$^{-2}$) | $\mu_0$ (cm$^2$/V$\cdot$s) | $l$ (10$^{-8}$ cm) |
|--------|-----------------------------------|-----------------------------------|----------------------------|----------------------------|-----------------|-----------------|
| A1     | 2.40                              | 62.9                              | 2.60                       | 3.05                       | 38.2            | 110.0           |
| A2     | 2.39                              | 58.6                              | 2.62                       | 3.06                       | 40.8            | 117.8           |
| A3     | 2.38                              | 58.8                              | 2.63                       | 3.07                       | 40.5            | 117.1           |

Table 2. Determined parameters.
\[ \sigma'(T) = \left[ \rho_p(T) - \rho(T) / \rho(T) \right] \sigma_N(T), \]  

where \( \rho(T) = aT + b \) is the sample resistivity in the normal state that is extrapolated to the low temperature region\(^5,20,43\). As mentioned above, according to the model\(^14\), the linear dependence of \( \rho(T) \) above \( T^* \) is the normal state of HTSCs that characterizes by the stability of the FS\(^2,3,14,19\).

According to recent concepts\(^8-11,23\), a small value of the coherence length in conjunction with a quasi-layered structure of the HTSCs leads to the formation of a noticeable area of SC fluctuations on the \( T \) vs. \( \sigma'(T) \) dependence for the base sample A1. Expectedly at the vicinity of \( T_c \), in the interval \( T_g - T_c \), the data deviate to the right from the linear dependence, which indicates the presence of 2D Maki-Thompson (MT) contribution to the planes of CuO\(_2\) by means of the Josephson interaction\(^11,20,46\). This is the 2D FLC regime, which is perfectly approximated by the Hikami-Larkin theory 2D-MT equation for HTSCs\(^25\):  

\[ T_g \]  

that appears in all equations. From this it is clear that the correct determination of \( T_g \) plays a key role in the calculations of both FLC and PG. At the vicinity of \( T_c \), the coherence length in the \( c \) axis (\( \xi_c(T) \)) is greater than d. d \( \approx 11.7 \)\(^5\) is the \( c \) axis lattice parameter of the YBCO unit cell\(^25,46\). In this case, the FCPs associate throughout the superconductor and form a three-dimensional (3D) state of HTSC\(^20,25,46\). Therefore, at the proximity of \( T_c \), the FLC can be described by the 3D equation of the Aslamazov-Larkin (AL) theory\(^11,24\) with the critical exponent \( \lambda = -1/2 \), which determines the FLC in any 3D system:

\[ \sigma'_{\text{AL3D}} = \frac{C_{\text{3D}}}{\xi_{\text{c}}} \frac{e^2}{\hbar^2} \left( \frac{T_c}{T} \right)^{-1/2}. \]

Here \( \sigma'(T) \) and \( \varepsilon \) are determined with high accuracy\(^11,20,51\). Also in Fig. 2, the arrows show \( T_c \), the Ginzburg temperature \( T_G \), down to which the fluctuation theories are valid\(^46,47\), and the \( T \) of the 3D-2D crossover \( T_c \) limiting the area of 3D fluctuations. Notably, above \( T_c \) \( = 92.34 \)\(^K\) (refer to Fig. 2), the data deviate to the right from the linear dependence, which indicates the presence of 2D Maki-Thompson (MT) contribution to the FLC\(^2,3\). Having determined \( \varepsilon \), we construct the dependence \( \ln\sigma' / \ln\varepsilon \) (Fig. 3). Figure 3a shows the corresponding dependence for the base sample A1. Especially at the vicinity of \( T_c \)(\( \xi_c(T) = 5.21 \)), the FLC is well modelled by the AL fluctuation contribution (3) for the 3D system. In double logarithmic coordinates this is the dashed line (1) with slope \( \lambda = -1/2 \). As mentioned above, it implies that the classical three-dimensional FLC materializes in HTSC for \( T \sim T_c \) and \( \xi_c(T) > \xi_c(T) > d \)\(^1,12,20,43\). Above the crossover temperature \( T_G \) \( \xi_c(T) < d \)\(^1,21,25,46\), and this is no longer a 3D regime. However, as before, \( \xi_c(T) > d_01 \), where \( d_01 \approx 3.5 \)\(^\AA\) is the separation of the conducting planes of CuO\(_2\) in YBCO\(^5\). Thus, up to temperature \( T_{c1} \) \( \ln(\varepsilon_{c1}) = -2.8 \) (Fig. 3) \( \xi_c(T) \) connects the inner planes of CuO\(_2\) by means of Josephson interaction\(^11,20,46\). This is the 2D FLC regime, which is perfectly approximated by the Hikami-Larkin theory 2D-MT equation for HTSCs\(^25\):

\[ \sigma_{\text{MT2D}} = \frac{e^2}{8\hbar d_1} \frac{1}{1 - \alpha/\delta} \ln \left( \frac{\delta/\delta}{\alpha} + 1 \right), \]

where the experimental points deviate downward from the theory (Fig. 3) implying that the classical fluctuation theories are not valid. Thus, \( T_{c1} \) limits the region of SC fluctuations from above: \( \Delta T_{c1} = T_{c1} - T_c \). Conversely, \( T_{c2} \) limits the region of SC fluctuations from below: As a result, below \( T_{c2} \), designated as \( \ln(\varepsilon_{c2}) \) in (Fig. 3a,b), the experimental points also deviate downward from the theory (Fig. 3), suggesting the transition to the range of critical fluctuations or fluctuations of the SC order parameter \( \Delta \) just near \( T_c \), where \( \Delta < KT \).

The thin curves (3) in the figure are constructed according to the Lawrence-Doniach equation (LD)\(^44\):  

\[ \sigma'_{\text{LD}} = \frac{e^2}{16\hbar d_1} \frac{1}{1 + 2\alpha} \varepsilon^{-1}. \]

| Sample | \( \xi_c(\text{Å}) \) | \( v_F(10^7 \text{cm/s}) \) | \( m'/m_0 \) | \( \tau(100 \text{K}) (10^{-11} \text{s}) \) | \( \tau_{\sigma}(100 \text{K}) (10^{-13} \text{Å}) \) | \( \beta \) | \( \tau_{\sigma}(100 \text{K}) (10^{-13} \text{Å}) \) |
|--------|----------------|----------------|---------|----------------|----------------|-------|----------------|
| A1     | 11.00          | 1.04           | 4.91    | 1.06           | 54.68          | 12.03 | 4.55           |
| A2     | 10.50          | 1.01           | 5.04    | 1.17           | 59.83          | 13.49 | 4.43           |
| A3     | 10.35          | 1.04           | 4.92    | 1.13           | 60.73          | 13.61 | 4.46           |

Table 3. Determined parameters.
Figure 2. Dependence of $\sigma'^{-2}(T)$ for the untwinned YBa$_2$Cu$_3$O$_{7-\delta}$ single crystal after annealing in oxygen (sample A1). Arrows indicate $T_c$, $T'_{c \text{mf}}$, the Ginzburg temperature $T_G$ and 3D-2D crossover temperature $T_0$. A straight line is to guide the eye.

Figure 3. Panel a. $\ln \sigma'$ vs $\ln \varepsilon$ for the sample A1 (squares) in comparison with fluctuation theories: 3D AL (dashed line (1)), 2D MT (solid curve (2)) and LD (solid thin curve (3)). The $T_{01}$ ($\ln \varepsilon_{01}$) determines the range of the SC fluctuations, $T_0$ ($\ln \varepsilon_0$) is the temperature of the 3D-2D crossover and $T_G$ ($\ln \varepsilon_G$) is the Ginzburg temperature. Panel b: The same dependencies for all three samples: A1 - blue squares, A2 - green dots and A3 - red triangles.
The LD model works in case of defects in samples21,42,51. In our case, curves (3) lie far from the experimental points, which confirm the absence of defects (primarily TB) in our samples.

Notably, in this case the maximum distance between the MT curve (2) and the extrapolated AL straight line (1), Δlnτ′ ≈ 0.1, which is typical for nonmagnetic YBCO14,29. In magnetic superconductors, such as SmFeAsO85, the MT curve (2) always passes much higher than the extrapolated AL straight line (1)11, and in this case Δlnτ′ ≈ 0.831. Such behavior indicates the presence of a magnetic interaction in HTSCs, which is clearly absent in our non-magnetic untwinned single crystal.

At T0, ξ, is determined by the equation

\[ ξ(0) = d \sqrt{\frac{ε}{2}} \]

Taking into account that ln(ε0) = −5.21 (Fig. 3) and using Eq. (6), we get ξ(0) = (0.86 ± 0.02) Å (A1), which is almost 2.6 times the coherence length alongside the c axis obtained for OD YBCO with TB (Tc = 91.07 K)41. This is most likely due to the fact that in single crystals with TB the region of SC fluctuations is ΔT2 = Tg0 − Tc ≈ 1.5 K, that is extremely small. While for A1, ΔT2 = Tg0 − Tc = 97.4 K − 91.9 K = 5.5 K, that is, 3.7 times more. The result again underlines a noticeable difference in the behavior of YBCO single crystals with and without defects. Additionally, ξ, has already been defined by Eq. (6), we can calculate dc0 from the relation ξ, = d0 √ε0. For sample A1, calculations give d0 = (3.5 ± 0.2) Å, in good agreement with the results of structural studies50.

In Fig. 3b the dependences of lnτ′/lnεc are shown for all three samples A1–A3. It is seen that with increasing T, all characteristic temperatures also vary slightly. The increase in the absolute value of lnτ′ results in decrease in ξ, from 0.86 Å (A1) to 0.81 Å (A3) (Table 1), in full agreement with the theory of superconductivity, where ξ ≈ 1/Tc.26

In the above equations

\[ α = \frac{2}{d} \left( \frac{ξ(0)}{d} \right)^2 \]

is the coupling parameter;

\[ δ = \frac{β}{πΘ(8)} \left( \frac{ξ(0)}{d} \right)^2 k_B T \sqrt{ε} \]

is the pair-breaking parameter, and the phase relaxation time τ, is determined by the equation

\[ τ = \frac{πΘ}{8k_B} = A/ε \]

where A = 2.998 × 10^-17 s K. Here the factor β = 1.203(τab/ξ,ab) with l being the mean free path and ξ,ab the coherence length along the ab plane, takes into account the approximation of the clean limit (l > ξ,ab) that consistently occurs in HTSCs due to the smallness of ξ(T)25,44–46.

Comparative analysis of phase relaxation time. Having determined the parameters of the FLC analysis, it seems interesting to examine the physical meaning of the short coherence length ξ,ab(0) in the framework of the simple two-dimensional model of free charge carriers23,24,46. This approach allows us to define a set of additional important parameters of the samples, including τ, which is actually the lifetime of the FCPs in the range of SC fluctuation. In HTSCs all parameters, including τ, and Hall coefficient RH, are functions of temperature. Consequently, the corresponding calculations, including those in YBCO, are performed at T = 100 K, as is customary in the literature26,52–54.

From the FLC analysis, using Eq. (7), we find the coupling parameter α, and then the pair-breaking parameter δ of Eq. (8), which is always ~ 20,14, if all other parameters are correctly determined. Next, we calculate the parameter τ,βT (refer to Table 3), assuming in Eq. (9) ε0 = ε0 = 26. Since it is assumed that T = 100 K, in order to find τ, that is necessary to determine the coefficient β = 1.203(ξ,ab). For this it is necessary to know the mean free path l, which is determined by the density of charge carriers, n, and the coherence length in the ab plane (i.e. ξ,ab). The charge carrier density n can be calculated from the values of the Hall coefficient R, namely n = τl/(e R). Here ε is the electron charge, and the coefficient τ = l/(l + δτβT) = 0.2) Å, in good agreement with the results of structural studies50.

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where $\Delta(0)$ is the SC order parameter at $T = 0$ K. Taking into account that in YBCO $\Delta(0)/k_B T_c = 5.54,61,62$ and setting $\xi_0 = \xi_0(0)$, for the Fermi velocity we obtain $v_F = 1.04 \times 10^5$ cm/s, and for the effective mass of charge carriers $m^*_e = m_0 = (\rho l_0 m_e^2)/(v_F^2 m_0) = 4.91$ (Table 3). After this, the transport time of the normal carriers $\tau (100) K = l_0 v_F = 1.06 \times 10^{-13}$ s can also be calculated. All estimates obtained are in good agreement with similar results reported for optimally doped YBCO30,42-44,59.

Finally, we find $\beta (100 K) = [1.203(l/\xi_0(0))] = 12.03$. Now, using found $\Delta(100 K) = 5.68 \times 10^{-13}$ s K, we get the value of $\tau_0 (100 K) = 4.55 \times 10^{-13}$ s. Performing similar calculations for samples A2 and A3 and taking into account the corresponding changes of $R_H$ and $\xi_0(0)$ with increasing $n_f$ (Tables 2 and 3), we obtain $\tau_0 (100 K) = 4.43 \times 10^{-13}$ s and $\tau_0 (100 K) = 4.46 \times 10^{-13}$ s (Table 3) in good agreement with similar results obtained by measuring the magnetoresistance on YBCO-PrBCO superlattices63 and FLC on YBCO films43.

Nevertheless, the mean free path $l$ and the Hall mobility $\mu_H$ are about 2 times, and $\tau_0 (100 K)$ is about 1.2 times more than in OD YBCO films40, which is most likely a specific property of untwinned single crystals64. At the same time, $\tau_0 (100 K)/\tau_0 (100 K) \sim 4 \pm 0.2$, in excellent agreement with the theory41, which takes into account the clean-limit approximation ($\alpha > \xi$), as mentioned above. It should be emphasized that, as in well-structured YBCO films with different $n_f$, in the untwinned single crystals $\tau_0 (100 K)$ is practically independent on $n_f$. This result, apparently, can be considered as a common property of cuprates, at least of compounds based on YBCO.

### Pseudogap analysis

In resistive measurements of cuprates the pseudogap is a deviation at $T \leq T^*$ of the longitudinal resistivity $\rho(T)$ from linearity in the normal phase11,14,64. This results to the realization of excess conductivity $\sigma^\prime$ (refer to Eq. (1)). It is established that if there were no processes in HTSC leading to the opening of the PG at $T^*$, then $\rho(T)$ would preserve its linearity up to $T \sim T_c$. It is obvious that $\sigma^\prime(T)$ is a consequence of the PG opening and should enclose details about the magnitude and temperature dependence of the PG11,23. Conventional fluctuation theories, modified by Hikami-Larkin25 for HTSCs perfectly describe the experimental $\sigma^\prime(T)$ but only to about $\sim 110 K$10,11,31. For complete information about the pseudogap, an equation is needed that would describe the experimental curve in all temperature range from $T^*$ to $T_c$ and would contain PG explicitly. Such an equation was proposed earlier12:

$$\sigma^\prime(\varepsilon) = \frac{\rho^2 A_4}{16h\xi_0(0)^2 \sqrt{2\varepsilon_0^*} \sinh(2\varepsilon/\varepsilon_0^*)}$$

(11)

where $(1 - T/T^*)$ takes into consideration the number of LPs formed at $T \leq T^*$, and $(\exp(-\Delta/T^*))$ determines the dynamics of LPs destruction as $T$ approaches $T_c$.

Additionally to $T^*$, $\alpha$, and $\xi_0(0)$, already defined above, Eq. (11) includes the numerical coefficient $A_4$, which is equivalent to the C-factor in the FLC theory20,51-53, the theoretical parameter $\varepsilon_0^*$64,65, and $\alpha^* = \Delta^*(T_c)$. Here it is presumed that $\Delta^*(T_c) = \Delta(0)$64,67 with $\Delta$ being the order parameter of the sample in the SC state, as mentioned above. Importantly, all these parameters can be easily determined within the LP model11,12,20,31. We consider this for the case of A1 (refer to Figs 4, 5). In the region $\ln \varepsilon < \ln \varepsilon_0^*$ or, respectively, $\varepsilon < \varepsilon_0$ (113 K $< T < 155 K$), $\sigma^\prime \sim \exp(\varepsilon)$66. This feature turned out to be one of the main properties of most HTSCs11,23. As a result, in the specified temperature range, $\ln(\sigma^\prime)$ depends linearly with respect to $\varepsilon$ with a slope $\alpha^* = 5.8$ (insert to Fig. 4a), which gives the determination of the remaining parameters $\varepsilon_0^*$ for all samples, which, as established previously12,23, significantly impacts the dependence of the PG on $T_c^*$.

To determine $A_4$ we approximate the experiment by the dependence $\sigma^\prime(\varepsilon)$ calculated by Eq. (11), in the vicinity of 3D AL-fluctuations near $T_c$ (refer to Fig. 4). $\ln \sigma^\prime(\ln \varepsilon)$ is in essence a linear dependence of $\varepsilon$ (i.e. the reduced temperature) and has a slope $\lambda = -1/2$. To find $\Delta^\ast(T_c)$ used in Eq. (11), we construct the curve $\ln \sigma^\ast(1/T)$ using all the parameters found66 (refer to Fig. 5). Here, the gradient of the theoretical curve (11) is highly influenced by the value of $\Delta^\ast(T_c)$20,31. The best approximation is achieved when the Bardeen-Cooper-Schrieffer (BCS) ratio $\Delta^\ast(T_c)/k_BT_c = 5.0 \pm 0.1$, which corresponds to the strong-coupling limit characteristic for YBCO. Accordingly, we obtain $\Delta^\ast(T_c)/k_B = 229 K$ in good agreement with the experimental value $\Delta^\ast(T_c)/k_B \approx 228 K$ (see Fig. 6). Similar results were obtained for samples A2 and A3 (refer to Table 4).

Solving equation (11) for the PG, $\Delta^\ast(T)$, we obtain11,12 over the entire temperature range from $T^*$ to $T_c^\text{mf}$

$$\Delta^\ast(T) = T \ln \frac{\sigma^\prime(\varepsilon)}{\sigma^\prime(\varepsilon_0^*) \sqrt{2\varepsilon_0^*} \sinh(\varepsilon/\varepsilon_0^*)}$$

(12)

Here $\sigma^\prime(T)$ is the experimentally determined excess conductivity and the remaining parameters are already defined within the LP model (Tables 1 and 4). The fact that $\sigma^\prime(T)$ is given by Eq. (11) (refer to Fig. 4) suggests that Eq. (12) gives reliable values of both the magnitude and the temperature dependence of $\Delta^\ast(T)$. The dependence $\Delta^\ast(T)$ for sample A1, constructed by the formula (12), using the following parameters extracted from the experiment: $T_c^\text{mf} = 91.84$ K, $T^* = 185$ K, $\xi_0(0) = 0.86 \AA$, $\varepsilon_0^* = 0.17$, $A_4 = 33$, $\Delta^\ast(T_c)/k_B = 229 K$, shown in Fig. 6. Also shown are the dependencies $\Delta^\ast(T)$ for samples A2 and A3, calculated in a similar way with the corresponding set of parameters given in Tables 1 and 4.

All the curves in Fig. 6 have the shape typical for YBCO films, with a maximum at $T = T_{pair} \approx 124$ K, which is close to $T_{pair} \approx 130$ K usually observed in well-structured YBCO films54,55, and a minimum at $T \approx T_{01}^{\text{mf}}$. It can be
seen that, in full accordance with the phase diagram of cuprates, $\Delta^{*}_{\text{max}(T_{\text{pair}})}/k_B$ expectedly increases from $258$ K (A1) to $270$ K (A3) along with an increase in $n_f$. The BCS ratio $D^* = 2\Delta^* (T_G)/k_BT_c$ also increases from 5.0 to 5.3. At the same time, $T_{\text{pair}}$ practically does not change (Table 4), which is reasonable, given the high $T_c$ of the samples.

**Figure 4.** $\ln \sigma'$ vs $\ln \varepsilon$ (squares) for the untwined YBa$_2$Cu$_3$O$_{7-\delta}$ single crystal (sample A1) in the whole temperature range from $T^*$ to $T_{\text{G}}$. The solid curve is fit to the data with equation (11) with the set of parameters given in the text. $\ln(\varepsilon_{\text{c01}})$ and $\ln(\varepsilon_{\text{c02}})$ designate the interval of exponential dependence of $\sigma^{-1}(\varepsilon)$. In the inset: $\ln(1/\sigma')$ with respect to $\varepsilon^6$. Solid line indicates the linear part of the curve between $\varepsilon_{\text{c01}} = 0.20$ and $\varepsilon_{\text{c02}} = 0.68$. Corresponding $\ln(\varepsilon_{\text{c01}}) = -1.82$ and $\ln(\varepsilon_{\text{c02}}) = -0.4$ are marked by arrows in the main panel. $\alpha^* = 5.8$ is used to determine the parameter $\epsilon^{*\text{g0}} = 1/\alpha^* = 0.17$.

**Figure 5.** $\ln \sigma'$ as a function of $1/T$ (squares) for the sample A1 in the interval from $T^*$ to $T_{\text{mf}}$. The solid curve is fit to the data with equation (11) with a set of parameters given in Tables 2, 3 and 4. The best fit is obtained when Eq. (11) is calculated with $\Delta^*(T_G) = 229$ K, that is $D^* = 2\Delta^*(T_G)/k_BT_c = 5.0$.

**Table 4.** Experimental Parameters.
the results of the PB theory ref.6. Having normalized the temperature and PG, respectively, by $T/T_c$ (Fig. 7). As in the PB theory, the density of LPs in our samples increases (from $n^\ast_n > 0$, and filling factors (the Peters-Bauer model (PB)) 6). Besides, in the calculations the hopping $t$ and the bandwidth $W = 12t$ were used as energy scales. Taking into account the fact that Eq. (12) contains information on the density of local pairs, we tried to compare our data with the results of the PB theory ref.6. Having normalized the temperature and PG, respectively, by $T^\ast(T)$ and $\Delta_{\text{max}}^\ast$, and having adjusted the parameters, we obtained good agreement with the PB theory (refer to Fig. 7). In the process of fitting, $\Delta^\ast(T_0)/\Delta_{\text{max}}^\ast$ was coincided with the minimum value $<n^\ast,n^\dagger>$ at the lowest $T$, and $\Delta^\ast(T_0)/\Delta_{\text{max}}^\ast$ with the maximum value. Both temperatures in Fig. 7 are indicated by arrows. On the theoretical curve 2, these temperatures are indicated by inclined arrows. Importantly, the same fitting factors were used for all samples.

After fitting, good agreement was found between the experimental $\Delta^\ast(T)/\Delta_{\text{max}}^\ast$ and the theory of PB with the lowest interaction parameter $U/W = 0.2$, which corresponds to the density of local pairs $<n^\ast,n^\dagger> \approx 0.3$ near $T_0$ (Fig. 7). As in the PB theory, the density of LPs in our samples increases (from $<n^\ast,n^\dagger> > 0.292$ (A1) to $<n^\ast,n^\dagger> > 0.305$ (A3)) with an increase in the interaction energy, which corresponds to an increase in the BCS ratio $\Delta^\ast$ in our case. As the temperature increases both $<n^\ast,n^\dagger>$ and our data, as expected, decrease (refer to Fig. 7), which seems reasonable. Indeed, the number of FCPs should decrease along with $T^3–6,11$. Importantly, at $U/W = 0.2$ the experimental data is consistent with theory in a wide temperature range, actually in the whole range of SC fluctuations. Whereas, if we compare the data with the theory for larger values of $U/W$ (curves 2 and 3), the data will deviate from the theory already at $T/W \sim 0.2$. Notably, $<n^\ast,n^\dagger>$ $\approx 0.3$ was also obtained for FeSe single crystals near $T_c^\ast$. Accordingly, when approaching to $T_c$, the maximum $\Delta^\ast(T_0)$ separates both regimes 8–12,28–30. The minimum of strongly bound bosons, which obey BEC. Below $T_{\text{pair}}$, LPs must be converted to FCPs, which are subject to SC fluctuations. Whereas, if we compare the data with the theory for larger values of $U/W$ (curves 2 and 3), the data will deviate from the theory already at $T/W \sim 0.2$. Notably, $<n^\ast,n^\dagger>$ $\approx 0.3$ was also obtained for FeSe single crystals near $T_c^\ast$. Apparently, such a density of LPs near $T_c$ is typical of all HTSCs.

**Conclusions.** Using the LP model, we have studied the effect of annealing on the temperature dependences of FLC and PG in untwinned OD YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) single crystal with a slight increase in the oxygen index (7-8). The increase in oxygen content and, respectively, $n_i$ was carried out by annealing the single crystal both in an oxygen atmosphere (sample A1) and by exposure at room temperature (samples A2 and A3). It is found that with increasing $n_i$ in the sample, $T_0$ increases, and resistivity decreases. As expected, the increase in $T_0$ is rather small, since $n_i$ actually corresponds to the maximum of the phase diagram (PD). At the same time, $T^* \approx 140 K$, is quite large $T^* = 185 K$ (A1). It is assumed that defects interfere with the establishment of phase coherence of LPs (paired fermions) and, thus, effectively reduce $T^*$. Importantly, in well-structured YBCO films, the sample with $T_c \sim 88 K$ has $T^* \approx 200 K$, which is much closer to $T^* = 185 K$, observed for A1. This result suggests the conclusion that the investigated properties of untwined YBCO single crystals are noticeably closer to the well-structured films, which was confirmed by the results of analysis of both FLC and PG.
The present study demonstrated that in the range of SC fluctuations near Tc, FLC is consistent with the fluctuation theories of Aslamazov-Larkin (3D term) and Hikami-Larkin (2D-MT term), and demonstrate a 3D-2D crossover when the temperature is increased. Tc determines \( \xi(0) = 0.86 \text{Å} \) (A1), which is 2.6 times higher than in optimally doped defective YBCO single crystals. This is most likely due to the fact that the range of FLC is very small: \( \Delta T_g = T_{g0} - T_{g1} = 97.4 - 91.9 = 5.5 \text{K} \) (A1), which, however, is 3.7 times more than in single crystals containing defects, where \( \Delta T_g \approx 1.5 \text{K} \). According to the theory\(^6,9\), in the range of SC fluctuations, cuprates retain the finite value of superfluid density \( n_s(T) \), and the FCPs behave mainly as SC Cooper pairs, but without long-range order (known as "short-range phase correlations") that is confirmed by a number of experiments\(^6,7,26\). This result once again underlines a noticeable difference in the behavior of YBCO single crystals with and without defects.

\( T_{g0} \) also determines \( d_{01} \) (distance between the conducting CuO \(_2\) planes). In this case, regardless of the density of charge carriers, \( d_{01} \approx 3.5 \text{Å} \), in agreement with the determinations of structural studies\(^26\). This result, together with the presence of the fluctuation contribution of 2D-MT in FLC, confirms the good structure of the samples. It should be also noted that with increasing \( T_c \), with the presence of the fluctuation contribution of 2D-MT in FLC, confirms the good structure of the samples.

\( \tau_{\phi} \) is almost independent on \( n_s \) (Table 3), because as well as in the well-structured YBCO films\(^6,7\), \( \nu \) is almost independent on \( n_s \) (Table 3).

Having determined the parameters of the FLC analysis, the physical meaning of the short coherence length \( \xi_{s0}(0) \) in HTSCs was examined in the framework of the simple two-dimensional model of free charge carriers\(^6,7,26-34\). This approach allowed us to define a set of additional important parameters of the samples, including \( \tau_{\phi} \), which is actually the FCPs lifetime in the range SC fluctuations. Most of the calculated parameters are in good agreement with similar data obtained for OD YBCO\(^26,27,34,38,39\). It is shown that found \( \tau_{\phi}(100 \text{K}) = 4.49 \pm 0.06 \times 10^{-11} \text{s} \) (Table 3) is only slightly \((\sim 1.2 \text{ times})\) more than in well-structured YBCO films, but, as in films, in fact does not depend on \( n_s \). Accordingly, \( \tau_{\phi}(100 \text{K})/\tau(100 \text{K}) \approx 4 \) is in excellent agreement with the Bieri-Maki-Thompson theory, which takes into account the approximation of the clean limit \((I > \xi)\), which always takes place in HTSCs due to the small value of \( \xi(T) \). A certain role in this may be played by the presence of structural and kinematic anisotropy in the system\(^27-71\).

The PG analysis has shown that the \( \Delta^*(T) \) curves (Fig. 6) have the shape characteristic of YBCO films\(^12,24\), with a clear maximum at \( T = T_{pair} \approx 124 \text{K} \) and a minimum at \( T \approx T_{g0} \). According to the theory of systems with low \( n_s \)\(^6,9,27-30\), \( T_{pair} \) separates both BEC and BCS regimes of LPs formation\(^8,12,27-29\). In full accordance with the PD of cuprates, \( \Delta^*_{\max}(T_{pair})/k_B \) expectedly increases from 258 K (A1) to 270 K (A3) along with an increase in \( n_s \) and \( T_c \) (Fig. 6). The BCS ratio \( D^* = 2\Delta^*(T_c)/k_BT_c \) also increases from 5.0 to 5.3, suggesting the expected increase in bonding energy of the LPs\(^26,37,38\). At the same time, \( T_{pair} \) practically does not change (Table 3), which is understandable due to the high \( T_c \) of the samples. When approaching \( T_c \), the PG curves show behavior being typical for all HTSCs with a maximum \( \Delta^*(T) \) just below \( T_c \) and a minimum value at \( T = T_g \) (inset in Fig. 6). Thus, the approach within the LP model makes it possible to determine the exact values of \( T_g \) and, as a consequence, to obtain reliable values of \( \Delta^*(T_c) \approx \Delta(0)^224,65,66 \) (Table 4).

Finally, the shape of the \( \Delta^*(T) \) curves near \( T_c \) (Fig. 6), was found to be very similar to the temperature dependence of the density of LPs in HTSCs \(<n,n_0>\) calculated within the three-dimensional attractive Hubbard model for different values of temperature, interaction, and filling factors (the Peters-Bauer model (PB)\(^8\)). For the first time, an estimation of the density of local pairs \(<n,n_0>\) in the optimally doped YBCO was carried out by comparing the experimental data of \( \Delta^*(T) \) with the PB theory (Fig. 7). It was determined that \(<n,n_0> \approx 0.3 \) near \( T_g \), which, likely, is a typical value for HTSCs.
**Experimental methods.** The YBa$_2$Cu$_{3-δ}$O$_{7-δ}$ single crystals were grown by the solution-melt technology in a gold crucible, according to the procedure described in Refs. 4,27. As is well known, with an increase in the oxygen content a tetra-ortho structural transition occurs in YBa$_2$Cu$_3$O$_{6.8}$, which leads to a twinning of the single crystal and the creation of twin boundaries (TB), minimizing its elastic energy. To obtain an untwined sample, the crystal was untwined into a special cell at a temperature 420°C and a pressure 30–40 GPa, according to the procedure proposed previously. In order to obtain the uniform controlled oxygen content, the crystal after untwining was repeatedly annealed for seven days in an oxygen atmosphere at 420°C.

Rectangular crystals of about 1.7 × 1.2 × 0.2 mm were selected from the same batch to perform the resistivity measurements. The smallest parameter of the crystal corresponds to the c-axis. The experimental geometry was selected so that the transport current vector was parallel to the ab-plane. The four-point probe technique with stabilized measuring current of up to 10 mA was used to measure the ab-plane resistivity, ρ$_{ab}$(T) [40, and references therein]. Silver epoxy contacts were glued to the extremities of the crystal in order to produce a uniform current distribution in the central region where voltage probes in the form of parallel stripes were placed. The procedure for making contacts was completed by adding silver wires with a diameter of 0.05 mm and a three-hour annealing at a temperature of 200°C in an oxygen atmosphere. Contact resistances below 1 Ω were obtained. The temperature was measured using a Pt sensor having an accuracy of about 1 mK. The measurements were carried out in the temperature drift mode on two opposite directions of the transport current to eliminate the influence of the parasitic signal. The critical temperature, T$_c$, was determined by extrapolation of the linear part of the SC transition to its intersection with the axis T = T$_c$.

In order to change the oxygen content and, and obtain the appropriate values of n$_f$ and T$_c$, the sample was annealed for two days in an oxygen flow at temperature 620°C. After annealing, the crystal was cooled to room temperature within 2–3 minutes, mounted in a measuring cell, and annealed for 10–15 minutes (sample A1). All measurements were carried out by heating the sample. To study the effect of annealing at room temperature, the sample after the first measurements of ρ(T) was kept for 20 hours at room temperature (sample A2) and then repeated measurements were performed. The following measurements were carried out after additional exposure of the sample at room temperature for three days (sample A3). After this procedure, not only increased T$_c$ and decreased ρ(T), but unlike the data of the previous work, the PG temperature T$^*$ also decreased noticeably, whereas the value of PG increased, which is in full agreement with the PD for YBCO (refer to Refs. 1–5,21 and references therein).

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**Author Contributions**
A.L.S. performed the experiments. All the authors analyzed and discussed the results and contributed to the writing of the paper.

**Additional Information**
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