Superfluid Density in the Underdoped $YBa_2Cu_3O_{7-x}$: Evidence for *d*-Density-Wave Order of the Pseudogap

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The investigation of the penetration depth $\lambda_{ab}(T, p)$ in $YBa_2Cu_3O_{7-x}$ crystals allowed one to observe the following features of the superfluid density $n_s(T, p) \propto \lambda_{ab}^{-2}(T, p)$ as a function of temperature $T < T_c/2$ and carrier concentration $0.078 \leq p \leq 0.16$ in CuO_2 planes: (i) $n_s(0, p)$ depends linearly on p; (ii) the derivative $|dn_s(T, p)/dT|_{T\to 0}$ depends on p slightly in the optimally and moderately doped regions ($0.10 < p \leq 0.16$), however, it rapidly increases with p further lowering; (iii) the latter finding is accompanied by the linear low-temperature dependence $[-\Delta n_s(T)] \propto T$ changing to $[-\Delta n_s(T)] \propto \sqrt{T}$. All these peculiarities can be treated in the framework of the *d*-density-wave scenario of electronic processes in underdoped high-T_c materials.

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In past years, much effort was devoted to study the nature and properties of pseudogap states of the high-T_c superconductors' (HTSC) phase diagram. This area corresponds to lower concentration p of holes per copper atom and lower critical temperatures T_c in comparison with the optimal value $p \approx 0.16$ and the maximum temperature of the superconducting transition. In the underdoped region, HTSC strongly differ from conventional materials, both in the normal and the superconducting states. This difference is likely to occur in p and T dependences of the superfluid density $n_s(T, p)$ of heavily underdoped HTSC.

It is well known that in clean BCS *d*-wave superconductors (DSC) the dependence $\Delta n_s(T) \equiv n_s(T) - n_s(0)$ is linear on temperature $T \ll T_c$: $[-\Delta n_s(T)] \propto T/\Delta_0$, where $n_0 \equiv n_s(0)$ and $\Delta_0 \equiv \Delta(0)$ are the superfluid density and the superconducting gap amplitude at $T = 0$. This dependence is confirmed by the measurements of the *ab*-plane penetration depth $\lambda_{ab}(T) = \sqrt{m^*/\mu_0 e^2 n_s(T)}$: $\Delta\lambda_{ab}(T) \propto T$ at $T < T_c/3$, where μ_0, m^*, and e are the vacuum permeability, the effective mass, and the electronic charge, respectively. The derivative $|dn_s(T)/dT|$ at $T \to 0$ determines the n_0/Δ_0 ratio. If thermally excited fermionic quasiparticles are the only important excitations even at $p < 0.16$, then the slope of $n_s(T)$ curves at $T \ll T_c$ is proportional to the $n_0(p)/\Delta_0(p)$ ratio: $|dn_s(T)/dT|_{T\to 0} \propto n_0(p)/\Delta_0(p)$. The measurements of $\lambda_{ab}(0)$ in underdoped HTSC showed that the superfluid density $n_0(p) \propto \lambda_{ab}^{-2}(0)$ increases approximately linearly with $p > 0.08$ reaching its maximum value at $p \approx 0.16$ [1,2].

When decreasing $p < 0.16$ and, hence, approaching the dielectric phase, the role of electron correlations and phase fluctuations becomes increasingly significant. The generalized Fermi-liquid models (GFL) allow for this through p-dependent Landau parameter $L(p)$ [3–5] which includes $n_0(p)$. The values of $\Delta_0(p)$ and $L(p)$ determine the doping dependence of the derivative $|dn_s(T)/dT|_{T\to 0} = L(p)/\Delta_0(p)$. In Ref. [3], the ratio $L(p)/\Delta_0(p)$ does not depend on p; the model [4] predicts $L(p)/\Delta_0(p) \propto p^{-2}$. The measurements of $YBa_2Cu_3O_{7-x}$ single crystals [6] and oriented powders [7] with the hole concentration $p \gtrsim 0.1$ showed that the slope of $n_s(T)$ dependences at $T \to 0$ is either slightly p dependent [6], which agrees with Ref. [3], or diminishes with decreasing $p \leq 0.16$ [7], which contradicts the GFL models [3–5].

Along with the above concept, there are a number of pseudogap concepts [8–11] proposed in order to describe the collapse of the single-particle density of states near the Fermi level, experimentally observed in underdoped HTSC at $T \gtrsim T_c$ by various techniques. At microwave frequencies, a breakdown of the normal skin-effect condition in some HTSC was also treated in terms of the pseudogap state [12]. In Ref. [13], the pseudogap order parameter was found to have the same *d*-wave symmetry as the superconducting one; it influences the quasiparticle spectrum at $T < T_c$ as well. In the precursor pairing model [14], based on the formation of pair electron excitations with finite momentum at $T > T_c$, this influence leads to a rise of $\Delta_0(p)$ and a decrease of $n_0(p)$ with p lowering. Hence, the decrease of the derivative $|dn_s(T)/dT|_{T\to 0} \propto n_0(p)/\Delta_0(p)$ is expected. The $n_s(T, p)/n_0$ dependences calculated in Ref. [15] show that their low-temperature slopes decrease with underdoping. An alternative behavior of $|dn_s(T)/dT|$ follows from the magnetic precursor *d*-density-wave (DDW) scenario of pseudogap [16]. In this model, a DDW order parameter $W(p, T)$ is directly introduced into the quasiparticle band structure. At low energies, the excitation spectrum of DDW consists of conventional fermionic particles and holes such as those of DSC with which it competes at $p < 0.2$. The DSC gap $\Delta_0(p)$ steadily vanishes with p decreasing, whereas the sum of zero-temperature squares $\Delta_0^2(p) + W_0^2(p)$ remains constant [17]. In the issue, the DDW model predicts a growth of the slope of the $n_s(T, p)/n_0$ curves at low T and $p < 0.1$.

The present Letter aims at the experimental verification of the theoretical predictions [3–5,14–17]. To fulfill the task, we investigated $\lambda_{ab}(T)$ dependences in a $YBa_2Cu_3O_{7-x}$ single crystal with the oxygen deficiency varied in the range $0.07 \leq x \leq 0.47$. The experiments were performed by the "hot-finger" technique [18] at the frequency of $\omega/2\pi = 9.4$ GHz and in the temperature range $5 \leq T \leq 200$ K. The initial $YBa_2Cu_3O_{6.93}$ crystal was grown in a $BaZrO_3$ crucible [19] and had a rectangular shape with dimensions being $1.6 \times 0.4 \times 0.1$ mm^3 [20]. To change the carrier density, we successively annealed the crystal in the air at various temperatures $T \geq 500\,^\circ$C [21]. Finally, five crystal states with critical temperatures $T_c = 92$, 80, 70, 57, 41 K were investigated. Using the empirical relation [22] $T_c = T_{c,\max}[1 - 82.6(p - 0.16)^2]$ with $T_{c,\max} = 92$ K at $p = 0.16$ ($x = 0.07$), we get the concentrations $p = 0.12, 0.106, 0.092, 0.078$ for the other four states of $YBa_2Cu_3O_{7-x}$ with lower T_c values and $x = 0.26, 0.33, 0.40, 0.47$, respectively. According to ac susceptibility measurements at the frequency of 100 kHz, the superconducting transition width amounted to 0.1 K in the optimally doped state ($p = 0.16$); however, the width increased with the decrease of p, having reached 4 K at $p = 0.078$. The temperature dependences of the ab-plane surface resistance $R_{ab}(T)$ and reactance $X_{ab}(T)$ are shown in Fig. 1 for each of the five crystal states. At $T < T_c/3$, all $R_{ab}(T)$ curves are linear on T. The residual losses $R_{ab}(T \to 0)$ do not exceed 40 $\mu\Omega$. In more detail, $R_{ab}(T)$ data will be discussed elsewhere [23]. The $X_{ab}(T)$ dependences in Fig. 1 are constructed with allowance made for both (i) the contribution $\Delta X_{ab}^{\mathrm{th}}(T)$ of thermal expansion of the crystal which essentially affects the measured reactance shift $\Delta X_{ab}(T)$ at $T > 0.9\,T_c$, and (ii) the additive constant X_0 which is equal to the difference between the values of $[\Delta X_{ab}(T) + \Delta X_{ab}^{\mathrm{th}}(T)]$ and $R_{ab}(T)$ at $T > T_c$: $X_{ab}(T) = \Delta X_{ab}(T) + \Delta X_{ab}^{\mathrm{th}}(T) + X_0$ [20]. So, in the normal state for each of the five crystal states, we have $R_{ab}(T) = X_{ab}(T)$ which implies the validity of the normal skin-effect condition. This finding enables one to extract the absolute values of the ab-plane penetration depth $\lambda_{ab}(T) = X_{ab}(T)/\omega\mu_0$ from $X_{ab}(T)$ curves at $T < T_c$.

Figure 2 shows the low-temperature sections of $\lambda_{ab}(T)$ curves. The linear extrapolation (dashed lines) of these dependences at $T < T_c/3$ gives the following $\lambda_{ab}(0)$ values: 152, 170, 178, 190, 198 nm for $p = 0.16, 0.12, 0.106, 0.092, 0.078$, respectively. The error in $\lambda_{ab}(0)$ is largely determined by the measurement accuracy of the additive constant X_0. In our experiments, the root-mean-square difference between $R_{ab}(T)$ and $X_{ab}(T)$ in the normal state corresponded to about 5 nm accuracy in the $\lambda_{ab}(0)$ value.

As follows from Fig. 3, halving of the concentration (namely, from $p = 0.16$ to $p = 0.078$) results in an approximately 2 times smaller $\lambda_{ab}^{-2}(0) = n_0\mu_0 e^2/m^*$ value. Similar behavior $n_0(p) \propto p$ within the range $0.08 < p \leq 0.16$ was observed by other groups [1,2]. It is easily seen that this dependence contradicts Uemura's relation $n_0(p) \propto T_c(p)$ [24]. The naive linear extrapolation of the dashed line in Fig. 3 at $p < 0.08$ leads to a nonphysical result: $n_0(p)$ is finite at vanishing p. To the best of our knowledge, there is no data of superfluid density

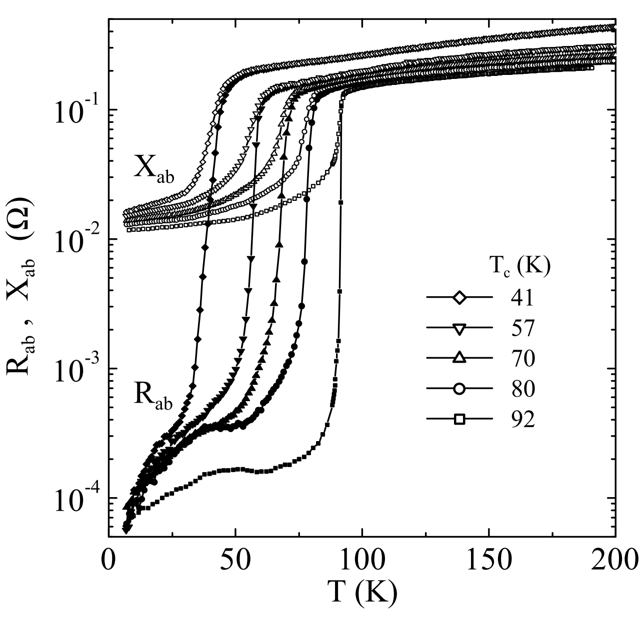

FIG. 1. Real $R_{ab}(T)$ (solid symbols) and imaginary $X_{ab}(T)$ (open symbols) parts of the ab-plane surface impedance of the $YBa_2Cu_3O_{7-x}$ single crystal for five different T_c values.

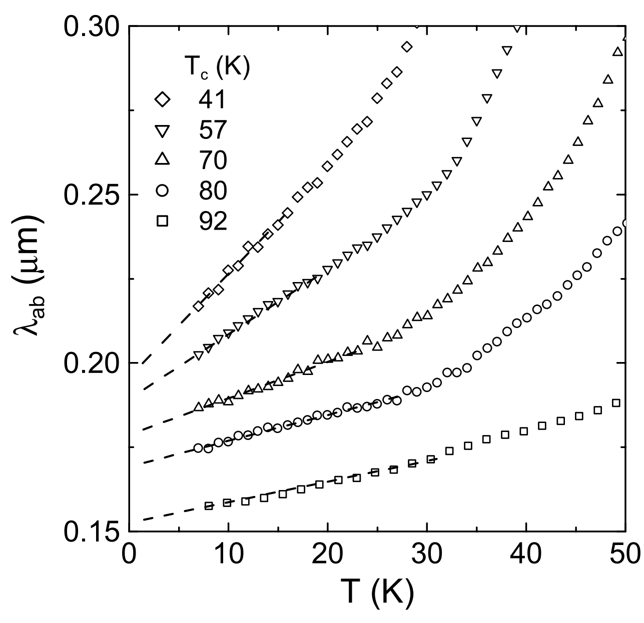

FIG. 2. Low-temperature dependences of $\lambda_{ab}(T)$ (open symbols) measured for five states of the $YBa_2Cu_3O_{7-x}$ crystal with $T_c = 92$ K, $T_c = 80$ K, $T_c = 70$ K, $T_c = 57$ K, and $T_c = 41$ K. Dashed lines are linear extrapolations at $T < T_c/3$.

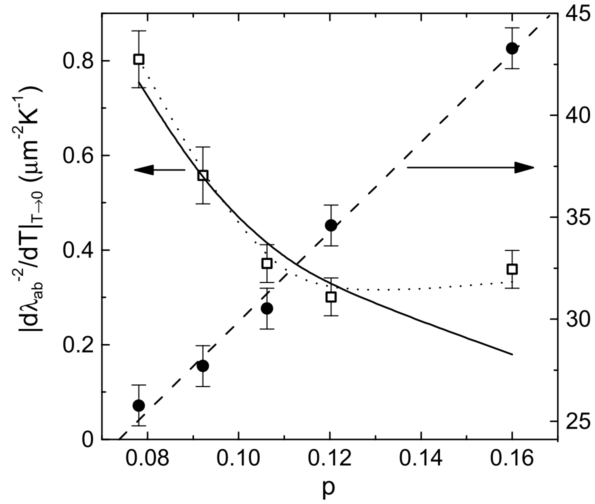

FIG. 3. The values of $\lambda_{ab}^{-2}(0) = n_s(0)\mu_0 e^2/m^*$ (right scale) and slopes $|d\lambda_{ab}^{-2}(T)/dT|_{T\to0} = \mu_0 e^2/m^*|dn_s(T)/dT|_{T\to0}$ (left scale) as a function of doping $p = 0.16 - \sqrt{(1 - T_c/T_{c,\max})/82.6}$ with $T_{c,\max} = 92$ K in $YBa_2Cu_3O_{7-x}$. Error bars correspond to experimental accuracy. The dashed and dotted lines guide the eye. The solid line is the $|dn_s(T)/dT| \propto p^{-2}$ dependence.

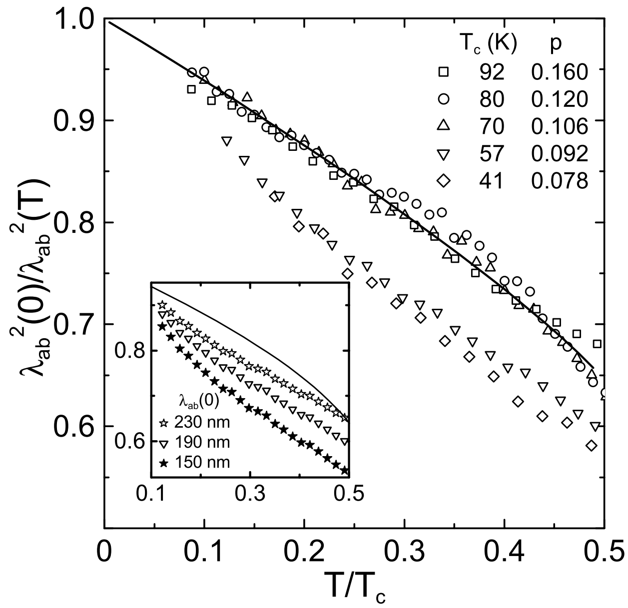

FIG. 4. The measured dependences of $\lambda_{ab}^2(0)/\lambda_{ab}^2(T) = n_s(T)/n_s(0)$ at $T < T_c/2$ in $YBa_2Cu_3O_{7-x}$ with different doping. The solid line is the $\lambda_{ab}^2(0)/\lambda_{ab}^2(T)$ dependence in the BCS d-wave superconductor (DSC). The inset shows the $n_s(T)/n_0$ experimental curve for $p = 0.092$ and the ones obtained using $\lambda_{ab}(0)$ increased (open stars) and decreased (solid stars) by 40 nm.

measurements in HTSC at $p < 0.08$. As for theoretical predictions, n_0 linearity on p extends down to $p = 0$ in the model [3], while in the DDW scenario [17,25] it exists in the underdoped range of the phase diagram where the DSC order parameter grows from zero to its maximal value (Fig. 1 from Ref. [25]). The latter agrees with our data.

In Fig. 3, we also show the slopes $|d\lambda_{ab}^{-2}(T)/dT|_{T\to0} \propto |dn_s(T)/dT|_{T\to0}$ of $\lambda_{ab}^{-2}(T)$ curves obtained from $\lambda_{ab}(T)$ dependences at $T < T_c/3$. The value of $|d\lambda_{ab}^{-2}(T)/dT|$ changes slightly at $0.1 < p \leq 0.16$ in accordance with Ref. [3]. However, it grows drastically at $p \lesssim 0.1$; namely, the $\lambda_{ab}^{-2}(T)$ slope increases 2.5 times with a p decrease from 0.12 to 0.08. The $|d\lambda_{ab}^{-2}(T)/dT| \propto p^{-2}$ dependence [4] is shown by the solid line in Fig. 3 and roughly fits the data at $p \leq 0.12$. The dotted line drawn through $|d\lambda_{ab}^{-2}(T)/dT|$ experimental points in Fig. 3 qualitatively agrees with the behavior of this quantity in the DDW model [17,25].

The temperature dependence of the superfluid density $n_s(T)$ at low T in the heavily underdoped $YBa_2Cu_3O_{7-x}$ proves to be one more check-up of the DDW scenario of pseudogap. $\lambda_{ab}^2(0)/\lambda_{ab}^2(T) = n_s(T)/n_0$ dependences obtained from the data in Fig. 2 are shown in Fig. 4 for different values of p. The solid line represents the DSC result. The evident peculiarities in Fig. 4 are the concavity of $n_s(T)/n_0$ curves corresponding to the heavily underdoped states ($p = 0.078$ and $p = 0.092$) and their deviation from DSC and the curves for $p = 0.16, 0.12, 0.106$. It should be noted that these peculiarities do not strongly depend on $\lambda_{ab}(0)$ values. This is demonstrated in the inset of Fig. 4, where the $n_s(T)/n_0$ experimental curve for $p =$ 0.092 is compared to the ones obtained using $\lambda_{ab}(0)$ increased (open stars) and decreased (solid stars) by 40 nm. Actually, the latter value is much higher than the experimental uncertainty.

The behavior of the superfluid density $n_s(T)/n_0$ in Fig. 4 contradicts the conclusions of the precursor pairing model [15], but agrees with the DDW scenario [17]. According to Ref. [17], at temperatures much smaller than the relevant energy scales W_0 and Δ_0, only the nodal regions close to the points $(\pi/2, \pi/2)$ and symmetry-related points on the Fermi surface will contribute to the suppression of the superfluid density. In a wide range of temperatures, the $n_s(T)$ dependence will be linear for the optimally and moderately doped samples, in which Δ_0 is larger than or comparable to W_0 and plays a leading role in the temperature dependence of the superfluid density. However, for the heavily underdoped samples, the situation is quite different. Though in the asymptotically low-temperature regime the suppression of the superfluid density is linear on temperature, there is an intermediate temperature range over which the suppression actually behaves as $\sqrt{T}$. It is worth emphasizing that the authors of Ref. [17] state that these features are independent of the precise $W_0(p)$ and $\Delta_0(p)$ functional forms. The only input that is needed is the existence of DDW order which diminishes with p increase and complementary development of the DSC order. The DDW order eats away part of the superfluid density from an

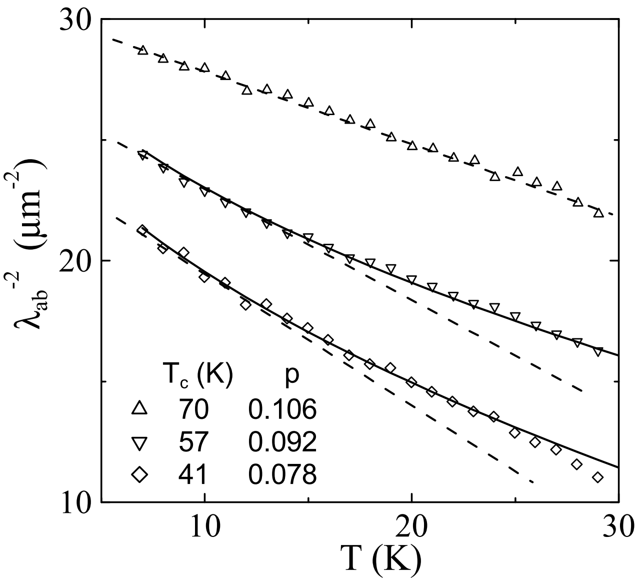

FIG. 5. Comparison of experimental $\lambda_{ab}^{-2}(T) \propto n_s(T)$ curves (symbols) with linear $[-\Delta\lambda_{ab}^{-2}(T)] \propto T$ (dashed lines) and root $[-\Delta\lambda_{ab}^{-2}(T)] \propto \sqrt{T}$ (solid lines) dependences for moderately doped ($p = 0.106$, $x = 0.33$) and heavily underdoped ($p = 0.092$, $x = 0.40$; $p = 0.078$, $x = 0.47$) $YBa_2Cu_3O_{7-x}$.

otherwise pure DSC system. Actually, in the intermediate temperature range $0.1\,T_c < T \lesssim 0.5\,T_c$, the experimental $n_s(T)$ curves in $YBa_2Cu_3O_{6.60}$ and $YBa_2Cu_3O_{6.53}$ with $p < 0.1$ are not linear but similar to $\sqrt{T}$ dependences. This is confirmed by Fig. 5, where the measured curves $\lambda_{ab}^{-2}(T) \propto n_s(T)$ are compared with linear ($\propto T$) in $YBa_2Cu_3O_{6.67}$ ($p = 0.106$) and $\sqrt{T}$ dependences $\Delta\lambda_{ab}^{-2}(T) = -3\sqrt{T}$ (λ_{ab} and T are expressed in μm and K) in $YBa_2Cu_3O_{6.60}$ ($p = 0.092$) and $\Delta\lambda_{ab}^{-2}(T) = -3.5\sqrt{T}$ in $YBa_2Cu_3O_{6.53}$ ($p = 0.078$). The dashed lines in Fig. 5 correspond to the linear at $T < T_c/3$ dependences of $\lambda_{ab}(T)$ presented in Fig. 2 and extended to higher temperatures. It is also interesting that these peculiarities of $\Delta\lambda_{ab}^{-2}(T)$ dependences in $YBa_2Cu_3O_{6.60}$ and $YBa_2Cu_3O_{6.53}$ are accompanied by inflection of the resistivity $\rho_{ab}(T)$ curves in the normal state of these samples which can be illustrated by Fig. 2 from Ref. [21].

Thus, three main experimental observations of this paper, viz. (i) linear dependence of $n_0(p)$ in the range $0.078 \leq p \leq 0.16$, (ii) drastic increase of the low-temperature $n_s(T)$ slope at $p < 0.1$, and (iii) the deviation of $\Delta n_s(T)$ dependence from universal BCS behavior $[-\Delta n_s(T)] \propto T$ at $T < T_c/2$ towards $[-\Delta n_s(T)] \propto \sqrt{T}$ with decreasing $p < 0.1$, evidence the DDW scenario [16,17,25] of electronic processes in underdoped HTSC. Nevertheless, the measurements of $n_s(T)$ at lower temperatures and in the high-quality samples with smaller carrier density are necessary for an ultimate conclusion.

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