Reversible magnetization and critical fluctuations in systematically doped YBa$_2$Cu$_3$O$_{7-\delta}$ single crystals

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The temperature and field dependence of reversible magnetization have been measured on a YBa$_2$Cu$_3$O$_{7-\delta}$ single crystal at six different doping concentrations. It is found that the data above 2 T can be described by the scaling law based on the GL-LLL (lowest Landau level approach based on Ginzburg-Landau theory) critical fluctuation theory yielding the values of the slope of upper critical field $-dH_{c2}(T)/dT$ near $T_c$. This set of values is self-consistent with that obtained in doing the universal scaling for the six samples. Based on a simple Ginzburg-Landau approach, we determined the doping dependence of the coherence length $\xi$ which behaves in a similar way as that determined from $\xi = h\nu_F/E_{sc}$ the superconducting energy scale. Our results may suggest a growing coherence length towards more underdoping.

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In hole doped high temperature superconductors the transition temperature $T_c$ and the maximum quasiparticle gap (or called as the pseudogap) behave in an opposite way: the former drops down but the latter rises up towards more underdoping [1]. Although consensus has been reached on the doping dependence of some quantities, such as the transition temperature $T_c$, the superfluid density $\rho_s$ and the condensation energy, etc., it remains still highly controversial about the doping dependence of the upper critical field or the coherence length in the underdoped region. In practice, however, to directly determine $H_{c2}(0)$ has turned out to be a difficult task due to its very large values. An alternatively way to derive $-dH_{c2}(T)/dT$ near $T_c$ is to measure the reversible magnetization or conductivity and then analyze the data based on the critical fluctuation theory. Using the Lawrence-Doniach model for layered structure of superconductors, Ullah-Dorsey obtained expressions for the scaling functions of various thermodynamic and transport quantities around $T_c$ [2]. Moreover, Tešanović et al. pointed out that the scaling of magnetization due to critical fluctuations near $H_{c2}(T)$ can be represented in terms of the Ginzburg-Landau (GL) mean field theory on a degenerate manifold spanned by the lowest Landau level (LLL) [2]. By using a nonperturbative approach to the Ginzburg-Landau free energy functional, $M(T)$ curves are evaluated explicitly for quasi-2D superconductors in a close form as:

$$\frac{M}{(HT)^{1/2}} = B f \left[ A \frac{T - T_c(H)}{(HT)^{1/2}} \right], \quad (1)$$

$$f(x) = x - \sqrt{x^2 + 2}, \quad (2)$$

where $A$ and $B$ are independent of $T$ and $H$, but $A$ is dependent on both the GL parameter $\kappa$ and $[dH_{c2}/dT]_{T_c}$, $B$ depends on $\kappa$. This scaling behavior is expected specially in a high magnetic field. Many experiments were tried to test these scaling laws and obtain the values of the mean-field transition temperature $T_c(H)$ and the slope $-dH_{c2}(T)/dT$ [1, 2, 3]. However, due to the sample diversity, the scaling produced values of $T_c(H)$ and $-dH_{c2}(T)/dT$ that did not agree with each other. As a consequence, the universal scaling for superconducting diamagnetization fluctuations is still elusive. In this paper, we systematically investigate the diamagnetization fluctuations in the vicinity of $T_c$ of a YBa$_2$Cu$_3$O$_{7-\delta}$ single crystal with six oxygen doping levels. The results indicate the plausibility of the existence of a universal scaling in the framework of 2D GL-LLL approximation theory. The doping dependence of $H_{c2}(0)$ are also reliably obtained.

The YBa$_2$Cu$_3$O$_{7-\delta}$ single crystal used here was grown by top-seeded solution-growth using the Ba3-Cu5-O solvent. Details for crystal growth were presented elsewhere [4]. It has a shape of platelet with lateral dimensions of 3.50 mm × 1.98 mm, thickness of 0.44 mm, and a mass around 15.56 mg. The different concentrations of oxygen were achieved by post-annealing the sample at different temperatures in flowing gas followed by a quenching in liquid nitrogen. The detailed annealing procedures are as follows: the as-grown YBa$_2$Cu$_3$O$_{7-\delta}$ single crystal was first annealed at 400 °C for 180 hours with flowing oxygen and slowly cooled down to room temperature. The resulted crystal (S1) is close to optimally doped with a $T_c = 92.0$ K. Then the following doping status on this specific sample were achieved by annealing it in flowing oxygen in sequence: $T_c = 85.3$ K (520 °C for about 110 h, S2), 79.5 K (540 °C for 120 h, S3), 68.5 K (580 °C...
Table I. Parameters at six different annealed states

| sample | $T_c(K)$ | $p$  | $T_{c0}(K)$ | $\Delta H_{c2}/|dH_{c2}/dT|$ (T/K) |
|--------|---------|------|-------------|----------------------------------|
| S1     | 92.0    | 0.146| 92.7 ± 0.6  | 3.45 ± 0.01                      |
| S2     | 85.3    | 0.127| 85.7 ± 0.3  | 3.23 ± 0.01                      |
| S3     | 79.5    | 0.117| 80.0 ± 0.5  | 3.03 ± 0.01                      |
| S4     | 68.5    | 0.103| 70.1 ± 0.5  | 2.00 ± 0.01                      |
| S5     | 58.6    | 0.093| 62.6 ± 0.5  | 1.82 ± 0.02                      |
| S6     | 30.5    | 0.070| 36.7 ± 1.0  | 1.43 ± 0.05                      |

for 110 h, S4), 58.6 K (680 °C for 130 h, S5). The last sample (S6) was annealed at 520 °C for 90 h with flowing N$_2$ gas yielding $T_c$ = 30.5 K.

The magnetization was measured by a Quantum Design superconducting quantum interference device (SQUID) magnetometer in both so-called zero-field-cooled (ZFC) and field-cooled (FC) modes with fields ranging from 10 G to 5 T parallel to c-axis. In the reversible regime, the data measured using ZFC and FC modes coincide very well. In SQUID measurement, a scanning length of 3 cm was taken, the SQUID response curves in reversible regime were fully symmetric to avoid artificial signal.

The $T_c$s of the sample at six annealed states were determined by the deviation point of magnetization from the normal state background (as shown in Fig. 1(a), ZFC mode, $H=10$ Oe). One can see that the superconducting transitions are rather sharp near the transition point. The gradually rounded foot of $M(T)$ curves in more underdoped samples may be attributed to the easy flux motion since the system becomes more 2D like. The relationship between the $T_c$s and the oxygen doping level $p$ (determined by using the phenomenological relation $T_c/T_{c,max}^{P_{max}} = 1 - 82.6(p - 0.16)^2$ taking $T_{c,max}^{P_{max}} = 93.6K$) has been summarized in Table I.

Presented in Fig. 1(b) are the $M(T)$ curves in the reversible regime under the applied magnetic fields $H = 0.5$, 1, 2, 3, 4, 5 T. A crossing point at $(T^*, M^*)$ appears in each set of $M(T)$ curves. Such crossing behavior of the $M(T)$ curves in high magnetic field has been well described by 2D or 3D GL-LLL scaling theory, and is a general consequence of fluctuations in the vortex state. It is interesting to note, however, that the value of $M^*/T^*$ is not a constant in our six sets of $M(T)$ data, being contradicting with the theoretical prediction. This deviation from the prediction of the 2D GL-LLL scaling theory were widely observed in Y123, Hg1223 and Bi2212 single crystals and may be attributed to the doping induced change of the anisotropy.

Despite the deviation mentioned above, excellent 2D scaling curves are obtained for each set of data with different $p$ and $H \geq 2$ T, as shown in Fig. 2 where $M(H,T)/(TH)^{1/2}$ is scaled as a function of the variable of $[T - T_c(H)]/(TH)^{1/2}$. We also performed 3D scaling of $M(T)$ for our six samples. The quality of the 2D scal-
ing are, however, better than that of 3D scaling for the five sets of $M(T)$ curves of S2, S3, S4, S5 and S6. For S1 (close to optimally doped) the quality of 3D scaling is as good as that of 2D scaling with a narrower scaling region. This may suggest a 2D-3D crossover between $p = 0.127$ and 0.146 for our deoxgenated YBCO crystals.

To fulfill this 2D scaling, two variables $T_{c0}$ and $dH_{c2}/dT$ are employed in $T_c(H)$ as the fitting parameters: $T_c(H) = T_{c0} - H (dH_{c2}/dT)^{-1}$. The values of $T_{c0}$ and $-dH_{c2}/dT$ resulted from the fit are also listed in Table I. The critical fluctuation region $\delta T \equiv T_{c0} - T_c$ increases with decreasing $p$, indicating a larger fluctuation regime for more underdoped YBCO. As shown in Table I, both the $T_{c0}$ and $|dH_{c2}/dT|_{T_c}$ drop down towards more underdoping. Another interesting phenomenon shown by Fig.2 for the scaled curves is that there is a new type of crossing point for different samples. This new type of crossing point may indicate a universal scaling among different samples.

In the following we will check the feasibility of the universal scaling for six samples. As all $M(T)$ data are measured on the same platelet of YBCO single crystal, this allows us to do universal scaling for six sets of $M(T)$ curves. The vertical axis $M/M_{(TH)}^{0.5}$ (y-axis) is intact because our experiments were done on the same sample with different doping concentrations and the GL parameter $\kappa$ is weakly doping dependent. According to the 2D LLL-scaling theory, an analytical formula of the $x$-axis for the scaling is written as [3]:

$$x = \frac{L}{\kappa} \left| \frac{dH_{c2}}{dT} \right|_{T_c} \left[ \frac{T - T_c(H)}{(TH)^{1/2}} \right]$$

(3)

where $L$ is a constant, related to $s$, the interlayer spacing. From this expression for $x$ axis, it is evident that a full 2D scaling analysis of diamagnetization fluctuations has to include a material-dependent scaling factor $A = \frac{L}{\kappa} \left| \frac{dH_{c2}}{dT} \right|_{T_c}$. Based on the 2D LLL-scaling curve in Fig. 2, such full 2D scaling is performed by multiplying the “$x$-axis” of each 2D LLL-scaled curve in Fig. 2 by a factor $A^\prime = A(p)/A(p = 0.127)$ to make all curves collapse on a single branch. Here we use the values of $|dH_{c2}|_{T_c}$ obtained in 2D scaling analysis and leave $\kappa$ as a free fitting parameter. In doing the universal scaling we put the 2D-scaled data of S2 as a reference, i.e., the “$x$-axis” is kept as it is. The resulted collapse of curves from the six samples is shown in Fig. 3. together with the doping dependence of $\kappa$. It is found that the scaling quality is predominantly controlled by $|dH_{c2}/dT|_{T_c}$ and $\kappa$ depends weakly on the doping level. Within the experimental uncertainty the quality of the data collapse is reasonably good. For a comparison, a theoretical curve generated using Eq. (2) is also plotted in Fig. 3. This theoretical curve describes the data rather well except for a deviation for the sample $p = 0.146$ ($T_c = 92.0$) on which we believe the system becomes more 3D like.

We now discuss the results obtained by the universal scaling analysis. According to the Werthamer-Helfand-Hohenberg (WHH) theory for a 2D system, the value of $H_{c2}(0)$ is given by $H_{c2}(0) = 0.697T_c \times |dH_{c2}/dT|_{T_c}$. Displayed in Fig. 4(a) is the dependence of $H_{c2}(0)$ on $p$ based on the obtained values of $|dH_{c2}/dT|_{T_c}$ and $T_c$ in Table I. Our data show a rough linear correlation between $H_{c2}(0)$ and $p$: $H_{c2}(0) = 2620(p - 0.058)$. We noted that such linear $H_{c2}(0) - p$ was also obtained for underdoped Bi-2212 polycrystal [14]. The spin-ordering quantum transition theory predicts that $H_{c2}(0)$ increases with doping in the underdoped regime [15]. Quantitatively, this theory certainly deserves consideration in interpreting our data.

It is interesting to note that in the underdoped regime the linearity of $H_{c2}(0) - p$ leads to a linear dependence of superfluid density $\rho_s(0)$ on $p$ by the relation: $H_{c2}(0) = \Phi_0/2\pi \xi^2 = \Phi_0 \kappa^2/2\pi \lambda_{ab}^2$. Based on the fact that $\kappa$ is weakly $p$ dependent, which is indeed the case for our samples and also found previously in underdoped YBCO [16], it is easy to have $\lambda_{ab}^2(0) \propto \rho_s(0) \propto p$. This linear correlation was recently verified in Bi2212 [14], La$_{2-x}$Sr$_x$CuO$_4$ and HgBa$_2$CuO$_{4+\delta}$ [17].

The coherence length $\xi_{ab}(0)$ of each sample can also be extracted from the $H_{c2}(0)$ value of Fig. 4(a). For example, we have $\xi_{ab}(0) = [\Phi_0/2\pi H_{c2}(0)]^{1/2} = 12.2$ Å for $H_{c2}(0) = 220$ T for the sample of $T_c = 92.0$ K. In Fig. 4(b) $\xi_{ab}$ is summarized and depicted as a function of $p$ in the underdoped regime. As $H_{c2}(0)$ is reduced to zero at the point $p = 0.058 \pm 0.002$, very close to the critical point $p_c = 0.050$ for superconductivity. This implies that $H_{c2}(0) = T_c$ simultaneously drop to zero at the critical point of the phase diagram, indicative of the complete suppression of $\rho_s(0)$ at $p_c \approx 0.05$. Another consistent
way to reckon the coherence length is using $\xi = \hbar v_F / E_{ac}$ with $E_{ac} = nk_BT_c$ the superconducting energy scale. $v_F$ is the Fermi velocity taking $2.5 \times 10^7 cm/s$ \cite{10} and is almost doping independent. In Fig.4(b) we present also the coherence length calculated in this way with $n \approx 20$. It is clear that the coherence length obtained in these two ways coincide rather well. Our results here may indicate a growing coherence length in more underdoped region, being consistent with our earlier report \cite{20}.

In summary, we have systematically investigated the critical fluctuations on a YBa$_2$Cu$_3$O$_{7-\delta}$ single crystal at six different doping concentrations. It is found that the data above $2T_c$ can be described by the universal scaling law based on the 2D GL-LLL critical fluctuation theory. Thus the values of the slope of upper critical field $-dH_c^2(T)/dT$ near $T_c$ (and thus $H_c^2(0)$) can be reliably extracted. The coherence length derived from both $H_c^2(0)$ and $\xi = \hbar v_F / E_{ac}$ show a similar growing behavior towards more underdoping.

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\[ \text{FIG. 4: (color online)} \text{(a) The doping dependence of the upper critical field } H_c^2(0) \text{ (phase coherence) derived by doing the scaling. The solid line is a guide to the eye. (b) Doping dependence of the coherence length determined from two different ways (see text).} \]