## Reversible magnetization and critical fluctuations in systematically doped $YBa_2Cu_3O_{7-\delta}$ single crystals

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The temperature and field dependence of reversible magnetization has been measured on a YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystal at six different doping concentrations. It is found that the data above 2 T can be described by a scaling law based on critical fluctuation theory in a lowest-Landau-level approach in Ginzburg-Landau theory yielding the values of the slope of upper critical field  $-dH_{c2}(T)/dT$  near  $T_c$ . A universal scaling has been found in the five underdoped samples. Based on a simple Ginzburg-Landau approach, we determined the doping dependence of the upper critical field  $H_{c2}(0)$  and the coherence length  $\xi$ . Our results may indicate a growing coherence length with increasing underdoping.

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In hole-doped high-temperature superconductors the transition temperature  $T_c$  and the maximum quasiparticle gap (also called the pseudogap) behave in opposite ways with respect to doping: the former decreases down but the latter increases with increasing underdoping.<sup>1</sup> Although some 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., the doping dependence of the upper critical field and the coherence length in the underdoped region still remain highly controversial. In practice, however, to directly determine  $H_{c2}(0)$ has turned out to be a difficult task due to its very large values. An alternative 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 critical fluctuation theory.

Using the Lawrence-Doniach model for layered-structure superconductors, Ullah and Dorsey obtained expressions for the scaling functions of various thermodynamic and transport quantities around  $T_{c}$ .<sup>2</sup> 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).<sup>3</sup> By using a non-perturbative approach to the Ginzburg-Landau free energy function, M(T) curves are evaluated explicitly for quasi-two-dimensional (2D) superconductors in a closed form as

$$\frac{M}{(TH)^{1/2}} = Bf\left(A\frac{T - T_{\rm c}(H)}{(TH)^{1/2}}\right),\tag{1}$$

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

where *A* and *B* are independent of *T* and *H*, but *A* is dependent on both the GL parameter  $\kappa$  and  $|dH_{c2}/dT|_{T_c}$ , and *B*  $\propto 1/\kappa$ . This scaling behavior is expected especially in a high magnetic field. Many experiments were tried to test these scaling laws leading to values of the slope  $-dH_{c2}/dT$ .<sup>4–7</sup> However, due to the sample diversity, the scaling produced

values of  $-dH_{c2}/dT$  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<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> (YBCO) 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  $H_{c2}(0)$  is also reliably obtained.

The YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystal used here was grown by top-seeded solution growth using a Ba3-Cu5-O solvent. Details for crystal growth are presented elsewhere.<sup>8</sup> The crystal has the shape of a platelet with lateral dimensions of 3.30  $\times 1.98$  mm, thickness of 0.44 mm, and the 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<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystal was first annealed at 400 °C for 180 h with flowing oxygen and slowly cooled down to room temperature. The resulting crystal (S1) is close to optimally doped with a  $T_c=91.5$  K. Then the following dopings of this specific sample were achieved by annealing it in flowing oxygen in sequence:  $T_c = 84.4 \text{ K}$  (520 °C for about 110 h, S2), 78.6 K (540 °C for 120 h, S3), 68.0 K (580 °C for 110 h, S4), 58.4 K (680 °C for 130 h, S5). The last sample (S6) was annealed at 520 °C for 90 h with flowing N<sub>2</sub> gas, yielding  $T_c$  = 30.3 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 the c axis. In the reversible regime, the data measured using ZFC and FC modes coincide very well. In SQUID measurements, a scanning length of 3 cm was taken; the SQUID response curves in the reversible regime were fully symmetric to avoid artificial signals.

The  $T_{\rm c}$ 's of the sample in the six annealed states were



FIG. 1. (Color online) (a) Temperature dependence of the magnetization measured in the warming-up process with a ZFC mode at H=10 Oe for the samples at six different doping levels. (b) Magnetization vs temperature at magnetic fields of 0.5, 1, 2, 3, 4, and 5 T. A crossing point (or a narrow area) marked as  $(T^*, M^*)$  appears for each sample showing a typical behavior of critical fluctuation.

determined by the intersection of the highest-slope line drawn through the data below  $T_c$  with the normal-state background extrapolation<sup>9</sup> [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 the M(T) curves in the 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<sup>10</sup>  $T_c/T_c^{max} = 1-82.6(p-0.16)^2$ , taking  $T_c^{max} = 93.6$  K] 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, and 5 T. A crossing point (or a small area) at or near  $(T^*, M^*)$  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. However, the value of  $M^*/T^*$  is not a constant in our six sets of

TABLE I. Parameters in six different annealed states.

Sample	$T_{\rm c}$ (K)	р	$T_{c0}$ (K)	$\left  dH_{\rm c2}/dT \right   ({\rm T/K})$
<b>S</b> 1	91.5	0.143	92.7±0.6	$3.45 \pm 0.01$
S2	84.4	0.126	85.7±0.3	$3.23 \pm 0.01$
<b>S</b> 3	78.6	0.116	$80.0 \pm 0.5$	$3.03 \pm 0.01$
S4	68.0	0.102	$70.1 \pm 0.5$	$2.00 \pm 0.01$
S5	58.4	0.093	$62.6 \pm 0.5$	$1.82 \pm 0.02$
<b>S</b> 6	30.3	0.070	$36.7 \pm 1.0$	$1.43 \pm 0.05$

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FIG. 2. (Color online) Scaling curves based on the 2D GL LLL critical fluctuation theory (see text) for the samples at six different doping levels. From this scaling one can determine the slope  $|dH_{c2}(T)/dT|_{T_c}$  for each sample. Interestingly, a different type of crossing point exists for the scaled curves of different samples.

M(T) data. This phenomenon does not necessarily mean the failure of the 2D GL LLL scaling theory;<sup>3</sup> it has been widely observed in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub>,<sup>11</sup> HgBa<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>8+x</sub>,<sup>12</sup> and Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub> (Bi2212) single crystals.<sup>13</sup> It may be attributed to the bilayer structure and the doping-induced change of the fluctuation region.

Despite the deviation mentioned above, excellent 2D scaling curves are obtained for each set of data with different p and  $H \ge 2$  T, as shown in Fig. 2, where  $M(H,T)/(TH)^{1/2}$  is scaled as a function of the variable  $[T-T_c(H)]/(TH)^{1/2}$ . We also performed 3D scaling of M(T) for our six samples. The quality of the 2D scaling is, 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 region. This suggests a 2D-3D crossover between p=0.126 and 0.143 for our deoxygenated YBCO crystals.

To satisfy 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  $-dH_{c2}/dT$  resulting 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  $T_{c0}$  and  $|dH_{c2}/dT|_{T_c}$  drop down with increasing underdoping. Another interesting phenomenon shown by Fig. 2 for the scaled curves is that there is a different type of crossing point for different samples. This type of crossing point may indicate a universal scaling among different samples.

As all M(T) data are measured on the same platelet of a YBCO single crystal, this allows us to test the universal scaling for six sets of M(T) curves based on 2D LLL scaling as shown in Fig. 2. In the following we will check the feasibility of the universal scaling (full 2D LLL scaling) for these six samples. In full 2D LLL scaling for our data, the vertical axis  $M/(TH)^{0.5}$  (*y* axis) is dependent only on the GL parameter  $\kappa$ , and scaling of the unit on the horizontal axis (*x* axis) is written as<sup>3</sup>



FIG. 3. (Color online) A universal scaling curve obtained by collapsing the six scaling curves for different doping concentrations based on the closed form of the scaling equation of Tešanović *et al.* (Ref. 3). The thick solid line is a theoretical curve after Eq. (2). The inset shows the relative  $\kappa$  value derived from the universal scaling.

$$x = \frac{L}{\kappa} \left| \frac{dH_{c2}}{dT} \right|_{T_c} \left( \frac{T - T_c(H)}{(TH)^{1/2}} \right), \tag{3}$$

where L is a constant, related to s, the interlayer spacing. From this expression for the *x* axis, it is evident that a full 2D LLL scaling analysis of diamagnetization fluctuations has to include a material-dependent scaling factor  $A = \frac{L}{\kappa} \left| \frac{dH_{c2}}{dT} \right|_{T_c}$ . In doing the universal scaling we first put the 2D-scaled data of S2 as a reference, i.e., the values of the "x axis" and "y axis" are multiplied by two free prefactors A(p=0.126) and B(p=0.126)=0.126) to get a best fit to the theoretical curve (thick solid line in Fig. 3). Based on the 2D LLL scaling scenario, such full 2D LLL scaling is performed by multiplying by a factor  $A(p) = A(p=0.126) [H'_{c2}(p)/H'_{c2}(p=0.126)] [\kappa(p=0.126)/\kappa]$ the value of the x axis, and multiplying by 1/B(p) = [1/B(p)]=0.126)][ $\kappa/\kappa(p=0.126)$ ] the value of the y axis of each scaled curve in Fig. 2 to make all curves collapse on a single branch. Here we use the values of  $H'_{c2} = \left| \frac{dH_{c2}}{dT} \right|_{T_c}$  obtained in 2D scaling analysis and leave  $\kappa/\kappa(p=0.126)$  as the only free fitting parameter for each sample. The resulting collapse of curves from the six samples is shown in Fig. 3, together with the doping dependence of  $\kappa/\kappa(p=0.126)$ . It is found that the scaling quality is predominantly controlled by  $|dH_{c2}/dT|_{T_a}$ and  $\kappa$  depends weakly on the doping level. Within the experimental uncertainty the quality of the data collapse is reasonably good and the data fit the universal theoretical curve [Eq. (2)] of the 2D GL LLL theory very well except for a deviation for the sample p=0.143 ( $T_c=91.5$  K), where 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<sup>14</sup> theory, the value of  $H_{c2}(0)$  is given by  $H_{c2}(0) = 0.69T_c |dH_{c2}/dT|_{T_c}$ . Displayed in Fig. 4(a) is the dependence of  $H_{c2}(0)$  on p based on the obtained values of



FIG. 4. (Color online) (a) The doping dependence of the upper critical field  $H_{c2}(0)$  (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  $H_{c2}(0)$  (see text). The solid line is drawn to guide the eye.

 $|dH_{c2}/dT|_{T_c}$  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 note that such a linear  $H_{c2}(0)-p$  was also obtained for an underdoped Bi2212 polycrystal.<sup>15</sup>

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,<sup>16</sup> it is easy to have  $\lambda_{ab}^{-2}(0) \propto \rho_s(0) \propto p$ . This linear correlation was recently verified in Bi2212,<sup>15</sup> La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> and HgBa<sub>2</sub>CuO<sub>4+ $\delta$ </sub>.<sup>17</sup>

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  $T_c = 91.5$  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±0.002, very close to the critical point  $p_c = 0.050$  for superconductivity, it is easy to see that  $H_{c2}(0)$  and  $T_c$  simultaneously drop to zero at the critical point of the phase diagram, indicative of the complete suppression of  $\rho_s(0)$  at  $p_c$  $\approx 0.05$ . Our results here may indicate a growing coherence length in the more underdoped region, which is consistent with our earlier report.<sup>18</sup>

In summary, we have systematically investigated the critical fluctuations in a  $YBa_2Cu_3O_{7-\delta}$  single crystal at six different doping concentrations. It is found that the data above 2 T

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can be described by a universal scaling law based on the 2D GL LLL critical fluctuation theory. Thus the values of the slope of upper critical field  $-dH_{c2}(T)/dT$  near  $T_c$  [and thus  $H_{c2}(0)$ ] can be reliably extracted. The coherence length derived from  $H_{c2}(0)$  increases toward more underdoping.

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