Hole concentration dependence of penetration depth and upper critical field in $Bi_2Sr_2(Ca, Y)Cu_2O_{8+\delta}$ extracted from reversible magnetization

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We have measured the reversible magnetization of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) with different hole concentrations as a function of temperature and magnetic field. By employing the vortex fluctuation model, the values of the penetration depth and the upper critical field are obtained. As the hole concentration decreases, fluctuation effects become more remarkable due to the decrease in superfluid density. The upper critical field has a maximum value when the hole concentration is around 0.19.

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I. INTRODUCTION

Even though nearly twenty years have passed since the discovery of high-temperature superconductors (HTSCs), the dependence of superconducting and normal states on the hole concentration per Cu-O plane *p* is still controversial. The superconducting transition temperature T_c of HTSCs shows a domelike behavior concerning hole concentration. Namely, an optimally doped (OP) sample $(p=0.16)$ has a maximum transition temperature $T_{c,\text{max}}$ in a family among HTSCs, and T_c decreases as the doping level changes from the OP state to the underdoped (UD) or overdoped (OD) regime.1,2 In UD HTSC's, various crossover phenomena associated with the opening of a pseudogap (PG) and stripes resulting from the spin-charge separation^{3,4} were observed at temperatures even far above T_c . It is widely believed that the PG is closely related to the superconductivity of HTSCs. In spite of the large amount of research into this phenomenon, no consensus on the nature of PG or superconductivity exists.

For highly anisotropic layered materials such as Bi- and Tl-based HTSCs, the component of magnetization along the Cu-O layer can be ignored, because $(m_c/m_{ab})^{1/2} \ge 1$, where m_c is the effective mass in the direction perpendicular to the Cu-O plane and m_{ab} is the effective mass within the plane. The reversible magnetization, *M*, for a sample of randomly oriented grains is given by

$$
M = -\frac{M_0}{2} \ln \frac{\eta \sqrt{e} H_{c2}}{B} + \frac{k_B T}{2 \phi_0 s} \ln \frac{C' \kappa^2 k_B T}{\phi_0 s B},
$$
 (1)

where $M_0 = \phi_0 / (32\pi^2 \lambda^2)$ (λ is the in-plane penetration depth), $\eta \approx 1.2 - 1.5$ is determined by the cutoff of the vortex core size, H_{c2} is the upper critical field, *s* is the interlayer spacing, ϕ_0 is the flux quantum, and the constant $C' \approx 16.8$ ^{5–7} The first term results from the local London theory, while the second term results from the consideration of the thermal fluctuations of pancake vortices, which are strong for highly anisotropic layered materials. Since κ is included in Eq. (1) as ln κ , various fitting parameters such as M_0 and ηH_{c2} are insensitive to a precise value of κ . Thus, in

the following, we fix $\kappa = 100$ and $s = c/2$, where *c* is the crystallographic length of the unit cell along the *c* axis. Taking the $\ln B$ derivative of Eq. (1), we have

$$
\frac{dM}{d\ln B} = \frac{M_0}{2} - \frac{k_B T}{2\phi_0 s}.\tag{2}
$$

This result does not depend on η .

We have measured the reversible magnetization of polycrystalline Bi-2212 rectangular shaped samples with different hole concentrations $(0.1 < p < 0.22)$. In this paper, we calculate the superfluid density and the upper critical field from the reversible magnetization to investigate the effect of the hole concentration upon the reversible magnetization.

II. EXPERIMENTAL WORK

Polycrystalline Bi-2212 samples with different hole concentrations were synthesized by solid-state reaction in a stoichiometric mixture of $Bi₂O₃$, $SrCO₃$, $CaCO₃$, CuO , and Y_2O_3 as described elsewhere.⁸ The hole concentration was controlled by partially substituting Ca by Y. The powders were prepared in the appropriate ratio and calcinated several times in air with intermediate grindings for more than 100 h of firing time. The product was pressed into pellets and sintered at an appropriate temperature for each sample. The sintering temperature was increased linearly from 850 °C for $x=0$ to 900 °C for $x=0.5$ with varying doping concentrations *x*. In order to enhance the homogeneity of samples, the sintering process was carried out three times with intermediate grinding.

The superconducting transition temperature T_c for each sample was measured by ac susceptibility in $H_{\text{r.m.s.}} = 0.1$ Oe. Unlike $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ with an 1/8 anomaly⁹ and YBa₂Cu₃O_y with a 60 K plateau,¹⁰ it is well known that T_c 's of Bi-2212 follow the empirical parabolic $T_c(p)$ formula without any anomalies. $T_c(p)$ is given by

$$
T_c(p) = T_{c, \text{max}}[1 - 82.6(p - 0.16)^2],\tag{3}
$$

where $T_{c,\text{max}}$ corresponds to T_c for $p=0.16$, the optimal concentration.¹¹ Thus, the hole concentrations for each

\mathcal{X}		0.1	0.2	0.25	0.3	0.4
T_c (K)	72.0	84.0	91.7	92.8	89.0	67.8
	0.213	0.196	0.177	0.148	0.135	0.103

TABLE I. Superconducting transition temperature T_c and hole concentrations p of $Bi₂Sr₂Ca_{1-x}Y_xCu₂O_{8+\delta}$

sample were estimated from Eq. (3) with the corresponding T_c (see Table I).

Magnetization data were obtained in fields up to 60 kOe using a SQUID magnetometer. Before measuring the reversible magnetization, we measured the values of the irreversible fields at 30 K $(t = T/T_c \approx 0.35)$ and 50 K $(t \approx 0.6)$ on Bi-2212 with $p=0.196$ in order to check roughly the reversible range of magnetization. The irreversible fields at 30 and 50 K are \sim 20 and \sim 3 kOe, respectively. Based on this result, we measured the magnetization for all our samples at the temperature $t \ge 0.5$ under fields of 5 kOe $\le H \le 60$ kOe with an interval of 5 kOe. The values of magnetization have been corrected carefully for the normal-state background moment including the paramagnetic contribution.

The intrinsic inhomogeneity patterns of the moderately UD Bi-2212 from STM measurement are similar to that of the OD one,¹² and the global superconductivity is given by the proximity effect. Martin *et al.*¹³ predicted that anomalous behavior appeared once the modulation length scale for the inhomogeneity is of the order of the superconducting coherence length. We expect that this condition may be satisfied at a sample with $p<0.1$, and the same physics can be applied to our samples. Actually, we observed that the extremely UD Bi-2212 with $p \approx 0.07$ exhibited anomalous behavior, and are preparing a paper related to this topic.

III. RESULTS AND DISCUSSION

Figure 1 shows the equilibrium magnetization curves as a function of temperature for Bi-2212 with $p=0.177$ measured at different fixed fields from 10 to 60 kOe at intervals of

FIG. 1. Temperature dependence of the reversible magnetization for Bi-2212 with $p=0.177$, which clearly crosses at $T=90.5$ K. The inset shows the hole concentration dependence of λ^{-2} at T^* , λ^{*-2} . The ratio of λ^{*-2}/T_c is approximately 0.06 $(\mu m^2 K)^{-1}$.

10 kOe. It can clearly be seen that all $M(T)$ curves cross at T^* =90.5 K. The presence of T^* where *M* is independent of field is a common characteristic in highly anisotropic layered materials and reflects the strong thermal fluctuations of pancake vortices near T_c .^{14–16} As *M* is independent of field at T^* , Eq. (2) becomes zero, and then λ^{*-2} at T^* can be easily derived. We confirmed that all our samples exhibit this crossover in $M(T)$ although they have different hole concentrations and T_c 's. The inset of Fig. 1 shows the *p* dependence of λ^{*-2} calculated for the samples with different hole concentrations. As shown, λ^{*-2} shows a similar behavior to the *p* dependence of T_c and the ratio of λ^{*-2}/T_c is approximately $0.06~(\mu \text{m}^2 \text{ K})^{-1}.$

Figure 2 shows the isothermal equilibrium magnetization curves as a function of magnetic field *B* for Bi-2212 with $p = 0.177$. We find that the $M(B)$ data for all our samples are well described by Eq. (1) (the solid lines of Fig. 2) and are linear on ln *B* down to $T_c/2$, as shown in the inset of Fig. 2, which means our samples are in the dirty limit at least for the range $T_c/2 < T < T_c$. Using the linearity of *M* on ln *B*, λ^{-2} at different temperatures can be obtained from the slope of Eq. (2). Here, in order to derive λ^{-2} and ηH_{c2} , we use the values of the *c*-axis length given in Ref. 17.

Figure 3 shows the temperature dependence of λ^{-2} for Bi-2212 with different p from 0.103 to 0.213. The λ values at *t*= 0.6 and 0.9 for Bi-2212 with *p*= 0.177 are 2820 and 3840 Å, respectively, which are comparable with the experimental values of \sim 2500 and \sim 3750 Å obtained at the same temperature for Bi-2212 with the nearly same hole concentration by Kogan *et al.*¹⁸ This implies that our procedure for

FIG. 2. Magnetic field dependence of isothermal reversible magnetization for Bi-2212 with $p=0.196$. The solid lines are from Eq. (1). The inset shows the semilogarithmic plot on the data measured at different temperatures. It is clear that the magnetizations are linear on lnB.

FIG. 3. Reduced temperature dependence of the penetration depth λ for Bi-2212 with different hole concentrations. It is likely that in the OP and UD regimes, λ^{-2} at $T=0$ K is rapidly suppressed with underdoping.

analyzing data is reasonable. We find a change in the shape of λ^{-2} with doping and a severe suppression in λ^{-2} at around $T_c/2$ with underdoping. In addition, λ^{-2} shows a more pronounced downward curvature in the UD regime (the open symbols). These trends of λ^{-2} with *p* are consistent with the experimental results for La_{2−*x*}Sr_{*x*}CuO₄ derived from the ac susceptibility measurements¹⁹ and theoretical work.²⁰ Since magnetization becomes irreversible at low temperature, λ^{-2} cannot be determined by measuring high-field magnetization. However, it is likely from Fig. 3 that λ^{-2} at $T=0$ K decreases rapidly as *p* decreases. $\lambda^{-2}(T)$ is proportional to the superfluid density $\rho_s(T)$ which is a measure of the phase stiffness of the condensate and is expressed by $\rho_s(0)$ $= 4\pi^2 \langle v_x^2 N(E) \rangle / e^2$, where v_x is the Fermi velocity and $\langle \cdots \rangle$ represents the average taken over an energy shell $E_F \pm \Delta_{SC}$.¹⁹ It is well known that the PG starts to open at around *p* $= 0.19$, the critical hole concentration, and increases as underdoping progresses. The decrease of λ^{-2} , that is, the decrease of ρ_s is caused by the suppression of the density of states within the energy range $E_F \pm \Delta_{SC}$ due to the presence of the PG.

Figure 4 shows the temperature dependence of ηH_{c2} for Bi-2212 as p ranges from 0.103 to 0.213, which is given through fitting Eq. (1) to data. According to the Werthamer-Helfand-Hohenberg (WHH) theory,^{21,22} $H_{c2}(T)$ is linear in (T_c-T) near T_c . So, if η is constant up to T_c , ηH_{c2} also should be linear as (T_c-T) . As shown in Fig. 4, ηH_{c2} of Bi-2212 is linear nearly to T_c when $p=0.213$. However, the linearity of ηH_{c2} is not sustained near T_c and the deviation point decreases with underdoping. Because the superfluid density $\rho_s(T)$ decreases with underdoping, this effect enhances fluctuations as the temperature approaches T_c . Then, as expected, the uncertainties in the vortex core cutoff associated with η and H_{c2} may increase due to the enhanced fluctuation.

Figure 5 shows the p dependence of $H_{c2}(0)$, the upper critical field at $T=0$ K. Once the slope of H_{c2} near T_c is known, $H_{c2}(0)$ for a two-dimensional system is given by

FIG. 4. Reduced temperature dependence of the upper critical field H_{c2} for Bi-2212 with different hole concentrations. The continuous solid lines are from the linear fit.

$$
H_{c2}(0) = 0.59T_c \left| \frac{dH_{c2}}{dT} \right|_{T_c}
$$
 (4)

which is derived by Bulaevskii from WHH theory.²³ In order to obtain $H_{c2}(0)$ from Eq. (4), we used the slope of lines in Fig. 4 and $\eta = 1.4$ given by the Hao-Clem method which is appropriate for an intermediate temperature range. $6,24$ We note that $H_{c2}(0)$ is not symmetrical to $p=0.16$, i.e., as shown in Fig. 5, $H_{c2}(0)$ increases with decreasing p in the OD regime, but decreases around $p=0.183$ which is closer to 0.19 at which PG starts to open, rather than $p=0.16$ at which T_c is maximum. In the OD regime $(p > 0.19)$, the superconducting properties are determined by Δ_{SC} , but in the UD and slightly OD regime $(p<0.19)$, the situation is rather complex due to the appearance of PG. This behavior can be understood in the following terms. The upper critical field $H_{c2}(0)$ is given by $H_{c2}(0) = \phi_0 / [2\pi \xi(0)^2]$, where ξ is the coherence length $\left[\xi(0) = \hbar v / \pi \Delta_{SC}(0)\right]$.²⁵ In the presence of the PG, the magnitude of the total excitation gap is given by $\Delta(0) = [\Delta_{SC}(0)^2]$ $+\Delta_{PG}^2$ ^{1/2}.^{26,27} The superconducting energy gap can be writ-

FIG. 5. Hole concentration dependence of $H_{c2}(0)$ obtained from WHH theory. The hole concentration dependence of $H_{c2}(0)$ for UD and slightly OD region is different from that for OD region due to the presence of the PG. The solid lines are from the linear fits.

ten as $\Delta_{SC}(0) = [\Delta(0)^2 - \Delta_{PG}^2]^{1/2}$. The relative size of Δ_{PG} to $\Delta(0)$ increases as p decreases, which results in a decrease of $\Delta_{SC}(0)$ for $p < 0.19$. Consequently, $H_{c2}(0)$ on *p* has a maximum value around $p=0.19$ as p decreases from the OD regime.

The coherence length $\xi(0)$ for each sample can be extracted from the $H_{c2}(0)$ value of Fig. 5. For example, $\xi(0)$ values for Bi-2212 samples with $p=0.177$ and 0.103 are 2.5 and 4.2 nm, respectively. These are somewhat longer than the values of 1.3 and 2.5 nm reported by Li et $al.^{28}$ on Bi-2212 samples with the similar hole concentration. This discrepancy is mainly caused by the use of the different η values; Li *et al.* used $\eta = 0.35$, instead of $\eta = 1.4$. Although the choice of η affects the absolute values for H_{c2} , our primary conclusion about the *p* dependence of H_{c2} remains unchanged.

IV. CONCLUSIONS

We have measured the reversible magnetization of Bi-2212 with different hole concentrations as a function of temperature and magnetic field. In the OP and UD regimes, λ^{-2} is rapidly suppressed with underdoping. Due to the presence of the PG, the upper critical field $H_{c2}(0)$ has a maximum value when the hole concentration is around 0.19 rather than 0.16, the optimal hole concentration.

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