Doping dependence of phase coherence between superconducting
\(\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}\) grains

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Abstract

In the present work, we report a new finding on the doping level dependence of the phase coherence between superconducting \(\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}\) (Bi-2212) grains. The experimental results at the strongly underdoped and overdoped regimes deviate from the expectations based on the doping level dependence of the superfluid density at \(T = 0\) K. These findings appear to be governed by interplay between competing orders inside the superconducting dome of cuprate superconductors. Two quantum critical points are likely to exist at the underdoped and overdoped regimes beneath the superconducting dome.

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A quantum phase transition (QPT) is a phase transition driven by quantum fluctuations at 0 K without thermal fluctuations [1]. QPT between two or more phases can be tuned by varying the external parameters such as pressure, strength of the magnetic field, or the density of electrons. As the external parameter is varied at 0 K, QPT occurs at some critical point, which is called the quantum critical point (QCP). Although QCP exist only at 0 K, the influence of quantum criticality extends over a broad regime, i.e. a quantum critical region, above 0 K in the phase diagram. Close to the QCP, the correlation length diverges with a power-law dependence on the distance from the QCP [1].

Sebastian et al. [2] reported that the extrapolations of $T_{\text{MI}}$, at which the in-plane resistivity reaches its lowest value before diverging logarithmically at low temperature, and the Fermi temperature, $T_F$, which is extracted from the quantum oscillation experiments, to 0 K for the underdoped YBa$_2$Cu$_3$O$_{6+\delta}$ (YBCO) samples, have an intercept at $p \sim 0.08$ below the superconducting dome, where $p$ is the hole doping level. Vishik et al. [3] showed from angle-resolved photoemission spectroscopy (ARPES) on Bi-2212 that for $0.076 < p < 0.19$, gaps at the near-nodal and intermediate momenta were independent of doping, but for $p < 0.076$, the Fermi surface was fully gapped and the gap anisotropy decreases. Grissonnanche et al. [4] reported that the upper critical field at $T = 0$ K, $H_{c2}(0)$, versus the $p$ phase diagram for YBCO consists of two peaks, which are located at $p_1 \sim 0.08$ and $p_2 \sim 0.18$. These results strongly indicate the presence of two QCPs at the underdoped and overdoped regimes in the phase diagram of the cuprate superconductors.

Motivated by these findings, we investigate the doping dependence of phase coherence between the superconducting Bi-2212 grains over a sufficiently wide range of $p$. As $p$ decreases from the optimal doping to the underdoped limit of the superconducting dome, the superfluid density at $T = 0$ K decreased, whereas the phase coherence between the underdoped superconducting Bi-2212 grains was enhanced. In the overdoped regime, a sudden reversal in the doping dependence of phase coherence between the superconducting Bi-2212 grains was observed. These results strongly support the presence of QCPs at $p_{c1} \simeq 0.075$ and $p_{c2} \simeq 0.190$ in the superconducting dome of the cuprate superconductors.

Granular samples of Bi-2212 with different $p$ values were prepared using the solid state reaction method as described elsewhere [5]. Each of the samples was sintered twice with intermediate grinding to enhance the carrier homogeneity. The doping level for each sample was changed by partially substituting Ca with Y. The resistivity of samples was measured
with a current of $I = 1.0$ mA and the ac susceptibilities were measured under a small field of $H_{rms} = 0.1$ Oe to exclude the effect of magnetic field as much as possible. We consider the superconducting transition temperature, $T_c$, to be the temperature at which the diamagnetic signal appears. The $p$ values for all the samples were calculated from an empirical relationship between $T_c$ and $p$, $T_c/T_{c,\text{max}} = 1 - 82.6(p - 0.16)^2$, where $T_{c,\text{max}} = 94$ K for Bi-2212 [6].

In a granular superconductor, as the phases of the different superconducting grains are coupled by the Josephson tunneling of Cooper pairs, the resistivity approaches zero [7]. Therefore, we use a temperature, at which resistivity disappears, as a measure of the phase coherence between superconducting grains near $T_c$. We denote the temperature as $T_\phi$, which is same as or lower than $T_c$, depending on the material. For granular YBCO and La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) with a low anisotropy ratio, $T_\phi$ is almost equal to $T_c$, which means that the long-range phase coherence between superconducting grains is established with the superconducting transition almost simultaneously. On the other hand, for Bi- and Tl-based cuprate superconductors close to two dimensional (2D) systems due to the extremely anisotropic property, $T_\phi$ is much lower than $T_c$ due to the enhanced thermal fluctuations. Moreover, as $p$ decreases from the overdoped to underdoped regimes, the 2D anisotropic nature in Bi- and Tl-based cuprate superconductors become more remarkable, which makes the phase more sensitive to the thermal fluctuations. Therefore, Bi-2212 is a good candidate for examining the relationship between $T_\phi$ and $T_c$ as a function of $p$.

The superfluid density at $T = 0$ K, $\rho_s(0)$, is a measure of the phase stiffness of the superconducting order parameter for the thermal fluctuations, which destroys the long-range order [8]. This $\rho_s$ is closely related to the penetration depth, $\lambda$, as $\rho_s(T) \equiv \lambda^{-2}(T)$. The values of $\lambda$ for strongly underdoped Bi-2212 were calculated from the reversible magnetization measurements as described elsewhere [9]. As shown in the inset in Fig. 1, as $p$ decreases to the lower limit of the superconducting dome, $\lambda^{-2}(0)$, i.e. $\rho_s(0)$, approaches zero, which is consistent with the results of Anukool et al. [10] for Bi-2212. This fact leads one to expect that the temperature range of the phase incoherence between the superconducting grains increases with decreasing $p$ due to the enhanced phase fluctuations.

Figure 1 shows the temperature dependence of the resistivity and ac susceptibility for Bi-2212 with different $p$ values ranging from $p = 0.068$ to 0.213. Despite the considerable data on the samples, data from the representative samples is presented for the sake of clarity.
In contrast to the expectation of the enhanced phase fluctuation based on the decrease in $\rho_s(0)$ in the underdoped regime mentioned above, Fig. 1 reveals two important features from the temperature dependence of the ac susceptibility and resistivity for each sample. First, for $T < T_\phi$, while the diamagnetic signals of the overdoped and moderately underdoped Bi-2212 increase slowly and nonlinearly, those for strongly underdoped Bi-2212 increase rapidly and linearly like a single crystal with a single superconducting phase. Second, as $p$ moves from the overdoped to the strongly underdoped regime, the difference between $T_c$ and $T_\phi$, $\Delta T \equiv T_c - T_\phi$, decreases, approaching zero at $p \simeq 0.073$, and then increases again. Therefore, $\rho_s(0)$ is not the only factor determining the phase coherence between the strongly underdoped superconducting Bi-2212 grains.

To assess the degree of phase coherence between the superconducting grains near $T_c$, the $p$ dependence of $\Delta T$ normalized by $T_c$, $\Delta T/T_c$, is plotted in Fig. 2. The results show that the magnitude of $\Delta T/T_c$ remains at an almost constant value of 0.140 near $p = 0.15$, and decreases by approximately one order of magnitude at $p \simeq 0.073$, even though $\rho_s(0)$ is reduced significantly, and then increases to 0.102 again at $p \simeq 0.068$. Therefore, we can say that as soon as the superconductivity (SC) emerges for Bi-2212 at $p \simeq 0.075$, the long-range phase coherence between the superconducting grains is established, as in conventional superconductors. On the other hand, in the overdoped regime ($p > 0.16$, at which $T_c$ reaches its maximum), the magnitude of $\Delta T/T_c$ shows a nonmonotonic $p$ dependence with a broad dip with a minimum, rather than zero, at $p \simeq 0.19$.

In general, $\rho_s(0)$ is independent of $T_c$, because $T_c$ ($\rho_s(0)$) is determined by the maximum superconducting gap (density of state) on the Fermi surface \[11\]. On the other hand, it is widely recognized that a linear relationship exists between $T_c$ and $\rho_s(0)$ in the moderately underdoped regime \[12\]. Therefore, we superimpose $T_c/\rho_s(0)$ on Bi-2212 as a function of $p$ on Fig. 2 using $T_c$ and $\rho_s(0)$ given in Ref. 13 and in the inset of Fig. 1, where, for comparison, a constant is multiplied such that the values of $T_c/\rho_s(0)$ near $p = 0.15$ is approximately 0.140 K·$\mu$m$^2$. Sometimes, $T_c/\rho_s(0)$ is used as a measure of the strength of the fluctuations \[11\]. Fig. 2 shows good agreement between the $p$ dependence of $\Delta T/T_c$ and that of $T_c/\rho_s(0)$ from $p \simeq 0.10$ to 0.19, suggesting that the same mechanism for the $p$ dependence of $\Delta T/T_c$ is at play in $0.10 < p < 0.19$. On the other hand, $\Delta T/T_c$ deviates downward at $p \simeq 0.10$ in the underdoped regime and upward at $p \simeq 0.19$ in the overdoped regime, from the $p$ dependence of $T_c/\rho_s(0)$.  


Because SC in the cuprate superconductors emerges in close proximity to the long-range antiferromagnetic Mott insulator with a spin density wave (SDW) order \cite{14,15}, it is conjectured that a magnetic QCP, separating SC and coexistence SC + SDW orders, exists at the underdoped regime beneath the superconducting dome through the quantum interplay of SC and SDW \cite{16–18}. Actually, experimental evidences of the coexistence of SDW and SC have been observed unambiguously at the strongly underdoped regime from numerous NMR and neutron scattering experiments on cuprate superconductors \cite{19–24}. As shown in Fig. 2, the fact that $\Delta T/T_c$ is zero at $p \simeq 0.075$ means that the phase correlation length between the superconducting grains diverges as $|p - p_{c1}|$ vanishes ($p_{c1}=0.075$), which indicates the presence of QCP at $p \simeq 0.075$. Vishik et al. \cite{3} reported that in Bi-2212, the gap slope, which represents the degree of the increase of the $d$-wave gap as a function of momentum away from the node of the Fermi surface, changed suddenly at $p \simeq 0.075$, at which $\Delta T/T_c$ is zero, as shown in Fig. 2. The SDW volume fraction at $T = 0$ K, measured by muon spin rotation, disappears in the vicinity of the magnetic QCP ($p_{c1} \simeq 0.075$)\cite{25}. The strong coupling of the electrons to spin fluctuations at the magnetic QCP is likely to enhance the coherent motion of the Cooper pairs between the different superconducting grains.

Moon and Sachdev \cite{26} predicted the presence of a crossover line between the SC + SDW and SC orders, which begins at the magnetic QCP ($p_{c1}$) at $T = 0$ K and ends at $T_c$ of a $p_m$ ($> p_{c1}$) of the superconducting dome. Therefore, it is expected that as $p$ approaches from $p_{c1}$ to $p_m$, the coupling of the electron to spin fluctuation weakens, resulting in an increase in $\Delta T/T_c$. Based on these experimental results, it is argued that the crossover line ends at $p \simeq 0.10$, above which $\Delta T/T_c$ remains at an almost constant value of 0.140. The data of $\Delta T/T_c$ near $p = 0.075$ is described well by a solid line in Fig. 2, given by a function of $\text{Atanh}(|p - p_{c1}|/c)$, where $A$ and $c$ are 0.0140 and 0.014, respectively. As $\text{tanh}x \approx x$ for $x \ll 1$, we have $\Delta T/T_c \approx 10|p - p_{c1}|$ near $p_{c1}$.

Theoretically, it was suggested that a magnetic QCP triggers the emergence of orders with different symmetries from those of the parent transition \cite{27,28}. These orders include $d$-wave SC, charge density wave (CDW) with a checkerboard structure, and pseudogap (PG) state lacking long range order. In the La-based cuprate superconductor, the stripe-like charge order (CO) near $p = 1/8$ is associated with the anomalous suppression of $T_c$. Recently, Chang et al. \cite{29} and Ghiringhelli et al. \cite{30} reported the observation of an incipient CDW, which is a periodic modulation of the conduction electron density, by high-energy X-ray
diffraction in underdoped YBCO. The peak intensity of CDW emerged below $T_{CDW} \sim 135$ K, which is lower than $T^* \sim 220$ K at the same doping level, where $T^*$ is the temperature, at which PG opens. The peak intensity grows upon cooling to $T_c$ below which it is partially suppressed. This suggests that CDW and SC are competing orders [31].

Comin et al. [32] and da Silva Neto et al. [33] reported that the emergence of CDW order, which was observed previously in LSCO and YBCO, is a ubiquitous phenomenon in other cuprate superconductors, such as Bi$_2$Sr$_2$CuO$_{6+\delta}$ (Bi-2201) and Bi-2212 from scanning tunneling microscopy, ARPES and bulk resonant elastic X-ray scattering experiments. They also reported the competing nature of SC and CDW by observing that the onset of SC weakens the strength of CO. In particular, da Silva Neto et al. [33] found that in Bi-2212, going from $p = 0.12$ to the underdoped regime, the CO wave vector, $\delta$, increases from 0.25 (nearly commensurate) to 0.30 (incommensurate) in the narrow doping range ($\Delta p \sim 5.0 \times 10^{-3}$) near $p = 0.1$, which is in contrast to the expectation from a Fermi surface nesting mechanism. Such an increase in $\delta$ in the narrow doping range near $p = 0.1$ is in line with the steep suppression of $\Delta T/T_c$ near $p \simeq 0.1$ in Fig. 2.

As $p$ goes from $p = 0.1$ to the overdoped regime, both $\Delta T/T_c$ and $T_c/\rho_s(0)$ enter the suppression region at $p \simeq 0.15$. While $T_c/\rho_s(0)$ maintains a tendency to suppress with increasing $p$, but $\Delta T/T_c$ increases at $p \simeq 0.19$, as shown in Fig. 2. As suggested above, the $p$ dependence of $\Delta T/T_c$ in $0.10 < p < 0.19$ is dominated by the same mechanism, i.e. the coexistence of CDW, PG, and SC. The associated suppression in $\Delta T/T_c$ and $T_c/\rho_s(0)$ at $p \simeq 0.15$ can be explained naturally by the competing nature of CDW and PG on SC. Competition between orders implies that the order, which sets up first, tends to suppress the other one on the Fermi surface [27, 28]. Although both $T_{CDW}$ and $T^*$ decrease with increasing $p$, our experimental results suggest that the two ($T_{CDW}$ and $T^*$) are larger than $T_c$ up to $p \simeq 0.15$. Therefore the magnitudes of $\Delta T/T_c$ and $T_c/\rho_s(0)$ remain almost unchanged. In contrast, for $p > 0.15$, $T_{CDW}$, which is smaller than $T^*$, first becomes smaller than $T_c$, and $T^*$ then becomes smaller than $T_c$ at a slightly higher $p$, giving rise to an enhancement in $\rho_s(0)$. These considerations on $T_{CDW}$ and $T^*$ are consistent with the phase diagram given in Ref. 34. Despite the decrease in $T_c$ for $p > 0.16$, such enhancement in $\rho_s(0)$ keeps until both $T_{CDW}$ and $T^*$ vanish, which is responsible for the decrease in $\Delta T/T_c$ at $p > 0.15$.

It was pointed out that as $p$ increases, the CDW and PG orders vanish simultaneously at $T = 0$ K at a special $p$ in the superconducting dome, setting up a charge QCP, $p_{c2}$,
at some distance from the magnetic QCP \cite{27}. Very recently, Fujita et al. \cite{35} observed that the electronic symmetry breaking tendencies, which are closely related to CDW and PG, weaken with increasing $p$ and disappear close to $p \simeq 0.19$. Therefore, $\rho_s(0)$ reaches its maximum due to the disappearance of competing orders, CDW and PG, on SC at $p \simeq 0.19$. As a result, $\Delta T/T_c$ approaches the minimum value, not zero, at $p \simeq 0.19$, because both the CDW and PG orders lack a long range correlation length, which is in contrast to what is observed at $p \simeq 0.075$. The order for $p > 0.19$ in the superconducting dome is a pure $d$-wave SC, which results in an increase in $\Delta T/T_c$ due to the enhanced phase fluctuations by the decrease of $\rho_s(0)$ with further increasing $p$ from $p \simeq 0.19$. Through previous study, we reported that $H_{c2}(0)$ of Bi-2212 with $p > 0.16$ is a maximum at $p \simeq 0.19$ \cite{5}, similar to YBCO mentioned above. The Fermi surface, on which coherent Bogoliubov quasiparticles are detected, undergoes an abrupt transition from arcs to a closed contour at $p \simeq 0.19$ \cite{35, 36}. These experimental facts support that in addition to the magnetic QCP at $p_{c1} \simeq 0.075$, there is another QCP, i.e. a charge QCP, at $p_{c2} \simeq 0.19$ at $T = 0$ K beneath the superconducting dome.

In conclusion, the phase coherence between Bi-2212 superconducting grains with $p_{c1} \simeq 0.075$ and $p_{c2} \simeq 0.190$ is enhanced remarkably. In particular, our experimental result at the strongly underdoped regime deviates from the expectation based on the doping level dependence of the superfluid density at $T = 0$ K. These findings are governed by quantum interplay between the competing orders, SDW, CDW, PG, and SC, inside the superconducting dome of cuprate superconductors. Two quantum critical points at $p_{c1} \simeq 0.075$ and $p_{c2} \simeq 0.190$ are likely to exist beneath the superconducting dome.

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Figure Captions

Figure 1. Temperature dependence of the ac susceptibility and resistivity for Bi-2212 with different $p$ values. The solid arrow and dashed one indicate $T_c$ and $T_\phi$ for each sample, respectively. The inset shows the $p$ dependence of $\lambda(0)^{-2}$ as the Mott insulating state is approached.

Figure 2. $p$ dependence of $\Delta T/T_c$ ($\Delta T \equiv T_c - T_\phi$) and $T_c/\rho_s(0)$ for Bi-2212. The solid line near $p = 0.075$ shows a fit by a function of $Atanh(|p - p_{c1}|/c)$, where $A$ and $c$ are 0.0140 and 0.014, and the solid line near $p = 0.19$ is an eye-guide.
FIG. 1: Temperature dependence of the ac susceptibility and resistivity for Bi-2212 with different $p$ values. The solid arrow and dashed one indicate $T_c$ and $T_\phi$ for each sample, respectively. The inset shows the $p$ dependence of $\lambda(0)^{-2}$ as the Mott insulating state is approached.
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