Superfluid response in monolayer high- T_c cuprates

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We have studied the doping dependence of the in-plane and out-of-plane superfluid density $\rho^s(0)$ of twomonolayer high- T_c superconductors, HgBa₂CuO_{4+ δ} and La_{2-x}Sr_xCuO₄, using the low-frequency ac susceptibility and the muon-spin-relaxation techniques. For both superconductors, $\rho^s(0)$ increases rapidly with doping in the underdoped and optimally doped regimes and becomes nearly doping independent above a critical doping, $p_c \sim 0.20$.

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Measurements of the magnetic penetration depth have been important in probing the order parameter and in testing theories of high- T_c superconductors (HTS's).^{1–3} In the hole doped HTS, the low-temperature dependence of the in-plane penetration depth $\lambda_{ab}(T)$ is linear and doping independent, indicating the presence of nodes in the superconducting energy gap.^{1,4} The *c*-axis penetration depth λ_c is a key parameter for some theories describing the mechanism of hightemperature superconductivity.^{5–13} It is sensitive to the electromagnetic anisotropy of the system and has been used to test the pairing symmetry and properties of interlayer dynamics.^{3,8,9}

In a recent study of the spin and charge response of HTS, it was found that both the superfluid density $\rho^s(0) \sim \lambda^{-2}(0)$ and the muon-spin-relaxation, (μ SR), rate show dramatic changes at a critical doping $p_c \sim 0.20$, slightly above optimal doping, in pure and Zn-doped La_{2-x}Sr_xCuO₄ (La-214) and Bi_{2.1}Sr_{1.9}Ca_{1-x}Y_xCu₂O_{8+y} (Bi-2212) at zero temperature.¹⁴ The sharp changes in the superfluid density with the disappearance of a spin-glass phase transition near p_c suggested a change in symmetry of the ground state. The existence of such a special doping has been demonstrated in many other physical quantities¹⁵ and the $\rho^s(0)$ and μ SR data could be linked to the presence of a quantum phase transition at p_c , which is in turn related to the opening of the normalstate pseudogap.

To elucidate further the changes in the ground state across the phase diagram of HTS, we have studied the doping dependence of the zero-temperature in-plane and out-of-plane superfluid responses ρ_{ab}^s and ρ_c^s for two-monolayer HTS materials: HgBa₂CuO_{4+ δ} (Hg-1201) and La-214. This study allows us to determine both in-plane and out-of-plane responses as a function of doping and to perform a direct comparison between two simple HTS's with different degrees of disorder. We find that in both systems, the superfluid density is strongly doping dependent below p_c and shows abrupt changes around p_c . For Hg-1201, the effect is sharper and there is actually a peak in the superfluid density at p_c .

The Hg-1201 samples were prepared in Houston using a method similar to that described in Ref. 16. Their doping level can be continuously varied from very underdoped to heavily overdoped regime by adding or removing oxygen.

Unlike La-214, where the doping is varied by Sr substitution for La, which may cause pronounced disorder effects, the variation of oxygen concentration in Hg-1201 is known to induce small lattice disorder.¹⁷ The Hg-1201 samples were characterized using magnetization and thermoelectric power measurements. The doping level p was determined by both thermopower¹⁸ and the universal relation $T_c = T_{c,max}$ $[1-82.6(p-0.16)^2]$.¹⁹ The La-124 samples were synthesized in Cambridge using solid-state reaction followed by oxygenation. Effort was made to ensure high purity and homogeneity. All La-214 powders were dried, reacted, ground, milled, repressed, and resintered at least four times. The phase purity was verified by powder x-ray diffraction as well as extensive transport and thermodynamic measurements. No signal of impurities or inhomogeneity was captured in microanalytical spectroscopic studies.²⁰ The T_c values as well as lattice parameters of these samples were in good agreement with published data. In La-214 p is taken to be equal to the Sr concentration. The heat-capacity anomalies and acsusceptibility transitions are sharp.

We have measured the magnetic penetration depth λ using the low-field ac susceptibility technique for grainaligned powders.^{9,21} The superfluid density is inversely proportional to the square of the in-plane penetration depth. To determine the in-plane and c-axis penetration depths separately, the grains were magnetically aligned in a static field of 12 T at room temperature. X-ray powder-diffraction scans²² for both La-214 and Hg-1201 samples showed that more than 90% of the grains had their CuO_2 planes aligned. The ac-susceptibility measurements were performed down to 1.2 K with a homemade susceptometer using miniature coils with $H_{ac} = 1-3$ G (rms) at f = 333 Hz. The absence of weak links among grains was confirmed by the linear response of the signal with H_{ac} from 0.3 to 3 G rms and f from 33 to 333 Hz. We also used a commercial susceptometer to confirm some of our findings. Considering the grains to be approximately spherical, as indicated by scanning electron microscopy, the data were analyzed using London's model.^{2,23} The ac-susceptibility data were also confirmed by standard transverse field μ SR experiments performed on unaligned powders at 400 G.24

Figure 1 shows the data for [panel (a)] T_c , $\lambda_{ab}^{-2}(0)$ and [panel (b)] $\lambda_c^{-2}(0)$ for La-214. The T_c and $\lambda_{ab}^{-2}(0)$ data were



FIG. 1. (a) Doping dependence of the superconducting transition temperature T_c and inverse square of the zero-temperature in-plane penetration depth for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (La-214) measured by the ac-susceptibility technique. (b) Doping dependence of the inverse square of the zero-temperature out-of-plane penetration depth.

published in an earlier paper and are included here for comparison.¹⁴ $\rho_{ab}^{s}(0)$ is nearly doping independent in the overdoped regime, but drops fast below p = 0.19. This suppression of the superfluid density for p < 0.19 was previously discussed in terms of a competition between quasistatic magnetic correlations and superconductivity.¹⁴ It has also been linked to the strong reduction in entropy as well as condensation energy associated with the opening of the normal-state pseudogap.^{15,24}

 $\rho_c^s(0)$ shows similar behavior as its in-plane counterpart. However, in contrast to the nearly linear doping dependence of $\rho_{ab}^s(0)$ on p, $\rho_c^s(0)$ shows a stronger doping dependence below p_c corresponding to $1/\lambda_c^2 \propto p^n$ with $n \sim 2.7$. This difference in the doping dependence between $\rho_{ab}^s(0)$ and $\rho_c^s(0)$ is probably associated with the unconventional interlayer coupling of electrons in high- T_c oxides, and is worthy of further theoretical and experimental investigation.

Figure 2(a) shows the doping dependence of T_c and $\rho_{ab}^s(0)$ for Hg-1201. Similar to La-214, $\rho_{ab}^s(0)$ is relatively doping independent in the overdoped regime and shows a sharp drop below 0.19. A similar *p* dependence of ρ_{ab}^s has been found for Bi-2212,²⁵ and recently also reported for Y_{0.8}Ca_{0.2}Ba₂Cu₃O_{7-\delta} (Ca:Y-123) and Tl_{0.5-y}Pb_{0.5+y} Sr₂Ca_{1-x}Y_xCu₂O₇ (Pb:Tl-2212).²⁶ The maximum of $\rho_{ab}^s(0)$



FIG. 2. (a) Doping dependence of the critical temperature T_c and inverse square of the zero-temperature in-plane penetration depth for HgBa₂CuO_{4+ δ} (Hg-1201). (b) Doping dependence of the inverse square of the zero-temperature out-of-plane penetration depth.

is located at p_c for all high- T_c compounds. It suggests that the observed doping dependence of $\rho_{ab}^s(0)$ below p_c is common to all HTS compounds and is not due to a structural transition or inhomogeneity. The relatively dopingindependent $\rho_{ab}^s(0)$ for $p > p_c$ in La-214 and Hg-1201 is in agreement to Bi-2212,²⁵ but seems to differ from the data for Tl₂Ba₂CuO_{6+ δ}, Ca:Y-123, and Pb:Tl-2212.^{26,27} The mecha-



FIG. 3. Doping dependence of the anisotropic ratio $\lambda_c(0)/\lambda_{ab}(0)$ for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (La-214) and HgBa₂CuO_{4+ δ} (Hg-1201).

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nism causing this difference is unknown and is certainly worthy of further investigation. Nevertheless, it is clear that the maximum of $\rho_{ab}^{s}(0)$ is located at p_{c} for all high- T_{c} cuprates.

Figure 2(b) shows the doping dependence of the *c*-axis superfluid density for Hg-1201. A sharp change from large to low superfluid response is also observed around p_c . This is the sharpest change in $\rho_c^s(0)$ ever being reported and together with the observed peak at p_c could be related to its tetragonal crystal structure and the fact that Hg-1201 is more ordered than La-214. It is worth noting that a significantly weaker glassy response has been observed in Hg-1201.²⁸ We may speculate that this observation suggests that the sharper changes near p_c may be linked to a quantum critical point for which disorder causes smoothing of the doping dependence of various physical properties and associated phase transitions.

The interlayer distance between the CuO₂ planes may be a key parameter for optimal T_c . This has been emphasized by Uemura recently.²⁹ Indeed, for the same in-plane superfluid density, T_c is higher if the interlayer distance is shorter. Therefore, the interlayer coupling seems to be essential for obtaining higher T_c . The observed variation of T_c cannot be explained by the simple Kosterlitz-Thouless transition where T_c is solely determined by the 2D superfluid density. The similar doping dependence of $\rho_c^s(0)$ to $\rho_{ab}^s(0)$ observed here supports this view and indicates the fundamental role of the *c*-axis electrodynamics to the overall superconducting condensation. As a matter of fact, $\lambda_c(0)$ for both monolayer cuprates studied here is small above p_c and agrees, for example, with the interlayer tunneling model of Anderson and co-workers.^{5–7} Large superfluid response above p_c seems to occur in connection with a crossover from two-dimensional to three-dimensional transport, as suggested by the doping dependence of the anisotropy in λ (Fig. 3) and the associated behavior of the anisotropy of the normal-state resistivity.^{30,31}

In summary, for the two-monolayer high- T_c cuprates, La-214 and Hg-1201, both the in-plane and the *c*-axis superfluid response remain relatively constant above p_c , but drop rapidly below p_c . We have found a peak in $\rho^s(0)$ at p_c for Hg-1201 indicating the strongest superconductivity at the point where the spin-glass phase transition (the glass transition temperature T_g versus *p* curve) vanishes and the normalstate gap extrapolates to zero.¹⁴ The rapid change and peak may be due to a change in the superconducting ground state. Furthermore, we have observed that the doping dependence of $\rho_c^s(0)$ in La-214 follows a power law of ≈ 2.7 .

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