Phase Diagram of Cuprate Superconductors at Ultrahigh Magnetic Fields

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We have investigated the field-temperature (H-T) diagram of the superconducting and pseudogapped states of $\mathrm{Bi_2Sr_2CaCu_2O_{8+y}}$ over a wide range of hole doping $(0.10 \le p \le 0.225)$. Using interlayer tunneling transport in magnetic fields up to 60 T to probe the density-of states (DOS) depletion at low excitation energies we mapped the pseudogap closing field H_{pg} . We found that H_{pg} and the pseudogap onset temperature T^* are related via a Zeeman relation $g\mu_BH_{pg}\approx k_BT^*$, irrespective of whether the magnetic field is applied along the c-axis or parallel to $\mathrm{CuO_2}$ planes. In contrast to large anisotropy of the superconducting state, the field anisotropy of H_{pg} is due solely to the g-factor. Our findings indicate that the pseudogap is of singlet-spin origin, consistent with models based on doped Mott insulator.

1 Introduction

In the phase diagram of cuprate superconductors, the most salient and fiercely debated feature is the normal state pseudogap [1], whose link to the superconductivity with high transition temperature (T_c) is still unresolved. The central issue is whether the pseudogap originates from spin or charge degrees of freedom and, in particular, whether it derives from some precursor of Cooper pairing that acquires the superconducting coherence at T_c . Experimentally, the situation appears deeply conflicted. On the one hand, photoemission [2] and surface tunneling spectroscopy [3, 4] show the pseudogap continuously evolving into a superconducting gap below T_c . The reports of anomalous and large Nernst effect in the normal state [5] led to claims of vortex-like excitations surviving up to temperatures close to T^* . On the other hand, intrinsic tunneling measurements revealed a double gap structure [6], indicating the pseudogap distinct even below T_c . With very different magnetic field sensitivities [7], the two gap features have been viewed by some as being unrelated [8].

Recently we have shown that in magnetic fields along the c-axis, the field H_{pg} that closes the pseudogap Δ_{pg} relates to T^{\star} via a simple Zeeman relation [9] (Fig. 1), suggesting that Δ_{pg} is controlled by the spin- rather than orbital degrees of freedom. However, several 'precursor superconductivity' scenarios, for example, those based on BCS-Bose Einstein crossover [10] or on intermediate coupling [11] models, argue that Zeeman scaling is compatible with the superconducting origin of the pseudogap. Conventionally, the upper critical field $H_{c2} \cong \Phi_0/2\pi\xi^2$ is determined not directly by the gap, but by the coherence

length ξ (the size of the Copper pair). The orbital motion of the Cooper pairs with increasing field eventually leads to diamagnetic pair breaking, restoring the normal state. Ginzburg-Landau description of anisotropic 3D superconductor gives $H_{c2}^{ab} = \Phi_0/2\pi\xi_{ab}\xi_c$ (for the field in the ab-plane) and $H_{c2}^c = \Phi_0/2\pi\xi_{ab}^2$ (for the field along the c-axis), where Φ_0 denotes the flux quantum [12]. In cuprates, the field anisotropy $\gamma = H_{c2}^{ab}/H_{c2}^c = \xi_{ab}/\xi_c$ is large [13], since the coherence length ξ_c along the c-axis (~ 2 Å) is much shorter than the in-plane ξ_{ab} (~ 30 Å). In the 'precursor' view, one would similarly expect an orbital frustration of preformed pairs (related to their center of mass motion) at the pseudogap closing field.

Here we will discuss our experiments probing the field anisotropy of H_{pg} . We find that while in the superconducting state anisotropy is large, $H_{pg}(T)$ displays only a small anisotropy of the g-factor, independently known from the magnetic susceptibility measurements [14]. This rules out the diamagnetic pair-breaking at H_{pg} . Furthermore, given the scales for H_{pg} (here, $\sim 70-100$ T) and T^{\star} (here, ~ 100 K), the Zeeman splitting for the spin degrees of freedom is not in correspondence with pair-breaking via a conventional paramagnetic (Pauli) effect [15]. The observed absence of orbital frustration naturally points to a singlet spin-correlation gap closed with a triplet spin excitation at H_{pg} .

2 Experimental

The magnetic field range required to close the pseudogap is immense in the underdoped regime, but decreases rather fast with doping [9]. Thus, to explore the field anisotropy,

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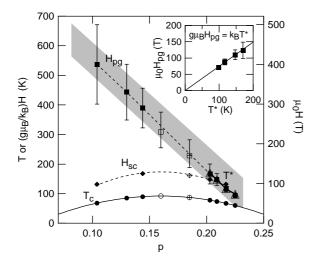


Figure 1. Doping dependence of low-temperature H_{pg} (squares) and H_{sc} (diamonds) in $\mathrm{Bi_2Sr_2CaCu_2O_{8+y}}$ (Bi-2212) together with T^\star (open triangles) and T_c (circles). The hole concentration p was obtained from the empirical formula $T_c/T_c^{max}=1-82.6(p-0.16)^2$ [8] with $T_c^{max}=92$ K. The shaded band covers $T^\star(p)$ in cuprates determined by several techniques (taken from ref. [1]). Inset: For $H\|c$ the pseudogap closing field H_{pg} and T^\star follow a simple Zeeman scale $g\mu_B H_{pc}^{pc} \approx k_B T^\star$ with g=2.0 down to the hole doping level p=0.225.

we have chosen to work with overdoped Bi-2212 crystal annealed in 200 atm O₂ for three days at 375°C to obtain a sharp transition at $T_c(H=0) \approx 60$ K, corresponding to the hole doping p = 0.225. Measurements were performed at 100 kHz in a 60 ms pulse 60 T magnet at National High Magnetic Field Laboratory (NHMFL) in Los Alamos using a lock-in technique. Negligible eddy-current heating was verified by the consistency of the data taken with successive pulses to different target fields. In overdoped samples, T^* can be very close to T_c , or may be below T_c (Ref. [6]). In our crystal, a semiconducting-like $(d\rho_c(T)/dT < 0)$ upturn in the c-axis resistivity $\rho_c(T)$ – a consistent signature of the pseudogap [9, 14] – is very obvious when T_c is suppressed by only a ~ 10 T field (Fig. 2). Here, the upturn – a result of the depletion in the quasiparticle density of states (DOS) near the Fermi energy – onsets at $T^{\star} \sim 100$ K. Fig. 2 illustrates how at high fields (~ 55 T) the upturn is suppressed, extending the metallic $(d\rho_c(T)/dT > 0)$ region to lower temperatures where the high-field $\rho_c(T)$ systematically approaches the normal ungapped resistivity $\rho_c^n(T)$ (Ref. [6]).

3 Results and Discussion

In the superconducting state, $\rho_c(H)$ becomes finite above the irreversibility field H_{irr} (\equiv zero resistivity field $H_{0\rho}$, see Fig. 3). A characteristic peak is observed at a higher field H_{sc} . This peak arises from a competition between two parallel tunneling conduction channels [16]: of Cooper pairs (Josephson tunneling that decreases with increasing field) and quasiparticles (dominating at high fields). At H_{sc} ,

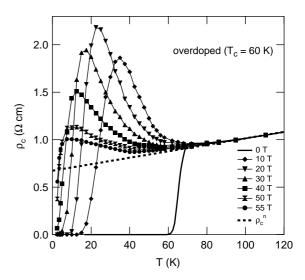


Figure 2. c-axis resistivity vs temperature in overdoped Bi-2212 (with the hole doping level p=0.225) up to 55 T $\parallel c$. The normal state resistivity $\rho_c^n(T)$ is shown as dashed line. Here the pseudogap temperature $T^\star \sim \! 100$ K.

the quasiparticle and the Josephson tunneling currents are comparable. Above the peak, the magnetoresistance is negative until the pseudogap is quenched at H_{pg} when $\rho_c(H)$ reaches ρ_c^n (Fig. 3a).

 H_{sc} is a good measure – a reliable lower bound on H_{c2} (Ref. [16]). The temperature dependence of H_{sc} in Fig. 4a shows that not only the initial slope for the two field alignments is very different, namely, $dH_{sc}^{ab}/dT|_{T_c} = -3$ T/K is much larger than $dH_{sc}^{c}/dT|_{T_c} = -0.27$ T/K, but also the overall curvature changes from concave to convex when the field is rotated from the out- to in-plane. This is reflected in the strong temperature dependence of the anisotropy ratio $\gamma_{sc} = H_{sc}^{ab}/H_{sc}^{c}$, which is ~ 12 close to 55 K but decreases by a factor of 3 near $0.5T_c$ (Fig. 4b). The irreversibility anisotropy (also T-dependent) is even larger; $H_{irr}^{ab}/H_{irr}^{c} \approx 20-30$ near 30 K, as shown in Figs. 4c and 4d.

To quantify the anisotropy of the pseudogapped state we use an identical procedure for $H \parallel c$ and $H \parallel ab$ to evaluate the excess quasiparticle resistivity $\Delta \rho_c$ due to the densityof-states (DOS) depletion associated with the pseudogap (the difference between ρ_c and ρ_c^n) (Ref. [9]). $\Delta \rho_c(H)$ follows a power-law at high fields above H_{sc} [9, 16], and when taken at each temperature to the limit $\Delta \rho_c \rightarrow 0$ it gives the value of the pseudogap closing field $H_{pq}(T)$. Fig. 5 (top) illustrates that for the in-plane applied field $\rho_c(H)$ has to be extrapolated somewhat further to reach the ungapped normal state value than for H||c. The values of $H_{pq}(T)$ can be independently tracked from the high-field scaling behavior of $\Delta \rho_c$ for $H \to H_{pg}$ shown in Fig. 5 (bottom). However, in contrast to H_{sc} and H_{irr} , $H_{pg}(T)$ is temperatureindependent below $\sim 0.8 T^{\star}$, as shown in the H-T diagram in Fig. 6. The anisotropy ratio $\gamma_{pg}=H_{pg}^{\parallel ab}/H_{pg}^{\parallel c}\approx 1.35$ holds throughout the entire temperature range below $T^\star.$

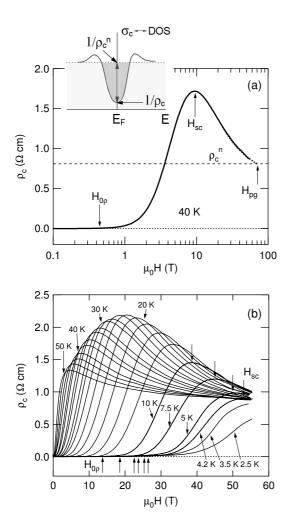


Figure 3. (a) $\rho_c(H)$ is marked by three characteristic fields: (zero-resistivity) $H_{0\rho} \equiv H_{irr}, H_{sc}$, and H_{pg} . The ungapped state value ρ_c^n (dashed line) is reached at the pseudogap closing field H_{pg} . The data are for the crystal with $T_c = 60$ K of Fig. 2. Inset: ρ_c is the inverse of the interlayer tunneling conductivity σ_c near Fermi energy $E = E_F$. (b) The peak field at H_{sc} , and the irreversibility field H_{irr} both strongly upshift on cooling. The data shown here are for $H \| c$.

Hence, we conclude that a Zeeman scaling relation also holds for $H\|ab$. This can be written as $g^{\parallel c}\mu_B H_{pg}^{\parallel c}(T=0)=g^{\parallel ab}\mu_B H_{pg}^{\parallel ab}(T=0)\approx k_B T^\star(H=0)$, with the g-factor anisotropy $g^{\parallel c}/g^{\parallel ab}\equiv\gamma_{pg}$. The value of γ_{pg} is in excellent agreement with the (spectroscopic splitting) g-factor anisotropy of ~ 1.3 obtained independently from the measurements of uniform spin susceptibilities [14] in fields $\|ab$ and $\|c$. Note that, given the scales of H_{pg} and T^\star , the observed absence of orbital anisotropy at the pseudogap closing field appears inconsistent with the simple paramagnetic Pauli pair-breaking effect, $H_p|_{T=0}=1.84T_c|_{H=0}$ [15], and the one deduced for anisotropic singlet pairing, $H_p=1.58T_c$ [18]. It is fully consistent with a singlet-spin (pseudo)gap closed by a triplet excitation at H_{pg} arising in the spin-charge separation scenarios for high- T_c based on a doped Mott insulator [19, 20, 21].

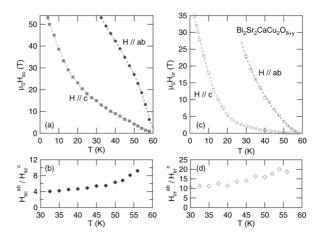


Figure 4. Anisotropy of the characteristic fields in the superconducting state. (a) The peak field $H_{sc}(T)$ and (c) the irreversibility field $H_{irr}(T)$ for $H\|c$ and $H\|ab$. H_{irr} was determined using a $\rho_c=0.01\rho_c^n$ criterion, consistent with our experimental resolution. Large anisotropy of H_{sc} and H_{irr} is seen in the ratios for the two field configurations in (b) and (d) respectively.

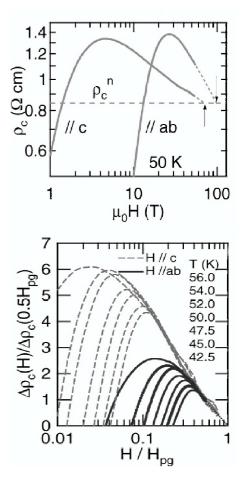


Figure 5. Top panel: c-axis resistivity ρ_c vs field for $H\|c$ and $H\|ab$ at T=50 K. Above the peak (at H_{sc}) it follows a power-law field dependence $[\Delta\rho_c(H)-\Delta\rho_c(0)]\propto H^{\alpha}$ $(\Delta\rho_c=\rho_c-\rho_c^n)$. At each temperature the pseudogap is closed at $H_{pg}(T)$ when $\rho_c(H)=\rho_c^n(H)$. Power-law fits indicated by thin dotted lines point to $H_{pg}^{\parallel c}$ somewhat lower than $H_{pg}^{\parallel ab}$. Bottom panel: The high-field collapse on the same scaling curve of $\Delta\rho_c(H)$ plotted vs H/H_{pg} for many temperatures enables us to independently track $H_{pg}(T)$ for $H\|c$ and $\|ab$.

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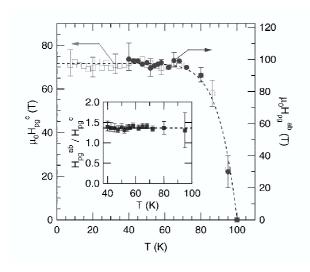


Figure 6. The pseudogap closing field $H_{pg}(T)$ for $H \parallel c$ (left hand side) and $H \parallel ab$ (right hand side) in Bi-2212 with p=0.225. The error bars indicate the uncertainties in the power-law fits. The ratio $H_{pg}^{\parallel ab}(T)/H_{pg}^{\parallel c}(T) \approx 1.35$ is temperature independent (inset) and corresponds to the anisotropy of the g-factor.

Acknowledgments

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