Closing the Pseudogap by Zeeman Splitting in Bi₂Sr₂CaCu₂O_{8+y} at High Magnetic Fields

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Interlayer tunneling resistivity is used to probe the low-energy density-of-states (DOS) depletion due to the pseudogap in the normal state of $Bi_2Sr_2CaCu_2O_{8+y}$. Measurements up to 60 T reveal that a field that restores DOS to its ungapped state shows strikingly different temperature and doping dependencies from the characteristic fields of the superconducting state. The pseudogap closing field and the pseudogap temperature T^* evaluated independently are related through a simple Zeeman energy scaling. These findings indicate a predominant role of spins over the orbital effects in the formation of the pseudogap.

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A central unresolved issue of high temperature superconductivity is the connection of normal state correlations, referred to as the pseudogap [1-5], to the origins of high T_c . At the heart of the debate [6–11] is whether the pseudogap, which manifests itself as a depletion of the quasiparticle density of states (DOS) below a characteristic temperature T^* , originates from spin or charge degrees of freedom and, in particular, whether it derives from some precursor of Cooper pairing [12] that acquires the superconducting coherence at T_c . Energies of the order of the pseudogap have been accessed with elevated temperatures, with applied voltage in tunneling measurements, and with infrared frequencies in optical spectra [1]. But little is known about the effect of magnetic field. The magnetic field response may be unique: e.g., in the case of the superconducting state the upper critical field H_{c2} is determined by the superconducting coherence length, and not directly by the superconducting gap, since magnetic field strongly couples to the orbital motion of Cooper pairs.

Current knowledge about the field dependence of the pseudogap derived from spectroscopic measurements is partly limited by the available dc field range [7-11]. More importantly, there is no systematic doping dependence in a single family of cuprates. Even in optimally doped YBa₂Cu₃O_{7- δ} alone, based on NMR relaxation rate measurements below 27.3 T, the pseudogap was claimed to decrease [7] or be independent of magnetic field [8]. In the underdoped YBa₂Cu₄O₈ no field effect on T^* was reported up to 23.2 T [9], while a recent NMR study indicated a measurable field dependence in slightly overdoped $TlSr_2CaCu_2O_{6.8}$ [10]. In this Letter, we report the interlayer (*c*-axis) resistivity ρ_c measurements in fields up to 60 T in $Bi_2Sr_2CaCu_2O_{8+y}$ (BSCCO) crystals in a wide range of doping, from which we make a first systematic evaluation of the pseudogap closing field H_{pg} that restores DOS to its ungapped state. Our results indicate a pronounced difference between field-temperature (H-T) diagrams of the pseudogap and the superconducting states and a simple Zeeman scaling between $H_{pg}(0)$ and T^{\star} .

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Among various techniques that quantify DOS, the ρ_c measurements are uniquely suited for exploring the highest magnetic field range available only in a pulsed mode. In highly anisotropic materials such as BSCCO where interlayer coupling between CuO₂ layers is sufficiently weak, the *c*-axis transport directly measures Cooper pair or quasiparticle tunneling in both normal and superconducting states [13], providing bulk information about the quasiparticle DOS at the Fermi energy. Thus, ρ_c should be particularly sensitive to the onset of the pseudogap formation, since the DOS depletion is largest at the Fermi energy. Moreover, ρ_c is controlled by the $(\pi, 0)$ points ("hot spots") on the anisotropic Fermi surface [14,15], where the pseudogap first opens up [2]. [This is in contrast to the in-plane resistivity ρ_{ab} mainly determined by carriers with momenta along the (π, π) directions [14].]

Our task here is to map an *H*-*T* diagram of the pseudogap state. To elucidate the field dependence of the pseudogap over a wide doping range, we carefully adjusted hole concentration *p* spanning both underdoped and overdoped regimes in BSCCO crystals grown by the floating-zone method [16]. The doping level was controlled by annealing in O₂ or N₂ at the appropriate pressures [3]. $\rho_c(H)$ was measured using a 33 T dc magnet [17] and a 60 T long pulse (LP) system at the National High Magnetic Field Laboratory (NHMFL) [18].

In slightly underdoped BSCCO at temperatures below T_c , the field dependence of ρ_c exhibits a peak that we have previously demonstrated to arise from a competition between two tunneling conduction channels: of Cooper pairs (at low fields) and quasiparticles (mainly at high fields) [13]. The peak position marks the field (in the superconducting state) where the quasiparticle contribution overtakes the Cooper pair tunneling current. The doping dependence of $\rho_c(H)$ in Fig. 1 clearly shows that the peak field H_{sc} in the highly underdoped crystal, where interlayer (Josephson) coupling is the weakest, is most easily suppressed. Magnetoresistance (MR) above H_{sc} is negative and remains so above T_c , as has been seen at lower fields [19]. An important difference between the underdoped and



FIG. 1 (color). *c*-axis resistivity ρ_c (labeled by temperatures) vs magnetic field H(||c) in an underdoped (UD) BSCCO crystal (a) and two overdoped (OD) crystals (b),(c). In the superconducting state, $\rho_c(H)$ becomes finite above the irreversibility field $H_{\rm irr}$ and exhibits a peak at $H_{\rm sc}$. The core feature in $\rho_c(H)$ that changes with doping is the slope of the high-field negative MR. Dotted lines in (c) are a guide to the eye pointing to the limiting value (<100 T) of $H_{\rm pg}(T)$. Inset: Excess resistivity due to the pseudogap $\Delta \rho_c$ (see Fig. 2) as a function of field for two samples at $T \sim 0.2T^*$. Thin lines are power-law fits and the shades indicate the uncertainties estimated from the leeway in the fitting parameters.

overdoped regimes is in the slope of the negative MR. A gentle slope in the underdoped regime turns steeper in the overdoped regime and, as shown in Fig. 1(c), at the highest fields the low-temperature $\rho_c(H)$ rapidly approaches the normal-state value.

In the overdoped samples, negative MR eventually disappears. This occurs at the same temperature at which the zero-field $\rho_c(T)$ develops a characteristic upturn from the *T*-linear dependence of the metallic state [Fig. 2(a)]. This temperature — at which a gaplike feature also appears in the static susceptibility [3] and in the tunneling spectra [4] of BSCCO—is identified as the pseudogap temperature T^* . In the pseudogap state below T^* , the negative MR is naturally understood by the suppression of the pseudogap by magnetic field [20]. In our most overdoped crystal with $T_c = 67$ K, a magnetic field of ~60 T downshifts the $\rho_c(T)$ upturn and the associated T^* by about 20 K [Fig. 2(b)]. In other words, at this doping level, the 60 T field at ~100 K closes the pseudogap. To track the pseudogap closing field at lower temperatures, we consider the

excess resistivity $\Delta \rho_c$ due to the DOS depletion. It is known from the intrinsic tunneling spectroscopy measurements [5] that the T-linear dependence of the metallic state persists below T^* for bias voltages sufficiently above the pseudogap voltage. Subtracting this metallic contribution gives $\Delta \rho_c$ [21]. The field at which $\Delta \rho_c$ vanishes is the pseudogap closing field $H_{pg}(T)$. To obtain $H_{pg}(T)$, we first note that the *c*-axis MR in the pseudogap state was recently shown to follow a power-law field dependence up to 60 T [13]. A fit to the power-law field dependence of $\Delta \rho_c(H)$ at different temperatures [inset in Fig. 2(b)] allows us to evaluate $H_{pg}(\hat{T})$ beyond 60 T. This evaluation is insensitive to the detailed functional form of the fit, as can be inferred from Fig. 1(c). We tried other extrapolation fits (e.g., polynomial) and they gave the same values of $H_{pg}(T)$ within the error bars in Fig. 3(a).

The entire *H*-*T* diagram of the pseudogap in the overdoped crystal is shown in Fig. 3. At low temperatures H_{pg}



FIG. 2 (color). Determination of the pseudogap temperature T^* and the pseudogap closing field H_{pg} from $\rho_c(T, H)$ in overdoped BSCCO. (a) $\rho_c(T)$ deviates from metallic *T*-linear dependence at the same temperature where negative MR disappears, identified as pseudogap temperature T^* . (b) In our most overdoped BSCCO with $T_c = 67$ K, T^* is shifted by ~20 K by a 58.5 T field. Inset: The excess quasiparticle resistivity $\Delta \rho_c(H)$ (above H_{sc}) is fitted to a power-law field dependence $[\Delta \rho_c(H) - \Delta \rho_c(0)] \propto H^{\alpha}$.



FIG. 3 (color). *H*-*T* diagram showing the pseudogap closing field $H_{pg}(T)$, the peak field $H_{sc}(T)$, and the irreversibility field $H_{irr}(T)$ in the overdoped BSCCO. (a) Up to 60 T, $H_{pg}(T)$ is directly determined from the down-shifting upturn of $\rho_c(T)$ (red squares). At lower temperatures, $H_{pg}(T)$ is obtained by extrapolating $\Delta \rho_c(H)$ to zero [inset in Fig. 2(b)]. The two procedures consistently produce a seamless $H_{pg}(T)$ within the error bars. The starkly different temperature dependencies of $H_{pg}(T)$ and $H_{sc}(T) \leq H_{c2}$ here extrapolate to roughly the same zero-temperature value. The usual estimate $H_{c2}(0) = 0.7(\partial H_{c2}/\partial T)|_{T_c}T_c$ with an initial slope of $\sim 2 \text{ T/K}$ [13] gives $H_{c2}(0) \approx 94 \text{ T}$, very close to the value of $H_{sc}(0)$. (b) At low temperatures, $H_{sc}(T)$ grows nearly exponentially and, in the $T \rightarrow 0$ limit, is only weakly dependent on doping.

is essentially flat with the limiting value of ~90 T. This is in marked contrast with the characteristic fields of the superconducting state: the peak field $H_{sc}(T)$ and the irreversibility field $H_{irr}(T)$. At low temperatures $H_{sc}(T)$ grows exponentially and points to the zero-temperature value of ~100 T, nearly independent of doping [Fig. 3(b)]. This difference, consistent with recent NMR [8,9] and intrinsic tunneling measurements [11], may indicate different origins of the pseudo- and superconducting gaps.

In the overdoped crystals the low-temperature H_{pg} and the zero-field T^* can be obtained independently and the comparison between the two (Fig. 4) leads to a strikingly simple conclusion. The pseudogap closing field scales with T^* as $g\mu_B H_{pg} \approx k_B T^*$. Here g factor $g = 2.0, \mu_B$ is the Bohr magneton, and k_B is the Boltzmann constant. This immediately suggests that magnetic field couples to the pseudogap by the Zeeman energy of the spin degrees of freedom. In the underdoped regime, the appreciable error bars in H_{pg} reflect the fact that the extrapolation extends considerably beyond the maximum laboratory field range of 60 T [inset in Fig. 1(c)]. However, the estimate gives a consistent and physically sensible picture, since in the underdoped regime we find that below 150 K $H_{pg}(T)$ is also flat and $H_{pg}(p)$ is a smooth continuation from the overdoped side. The observed general trend of the high-field slope of $\rho_c(H)$ as a function of doping is unmistakable, and the Zeeman energy scale of H_{pg} is in good agreement with the reported energy scale of the pseudogap and T^{\star} [1,25] (see the shaded band in Fig. 4). Thus, we surmise that the Zeeman scaling found in the overdoped samples holds in the entire doping range.



FIG. 4 (color). Doping dependencies of low-temperature H_{pg} , $H_{sc}(T \rightarrow 0)$, T^* , and T_c . The hole concentration p was obtained from the empirical formula $T_c/T_c^{max} = 1 - 82.6(p - 0.16)^2$ [22] with $T_c^{max} = 92$ K. The right-hand-side field scale directly translates onto the Zeeman energy scale on the left-hand side as $(g\mu_B/k_B)H$. H_{pg} (red squares) and T^* (blue triangles), obtained separately in the same crystals in the overdoped regimes, give a scaling $g\mu_B H_{pg} \approx k_B T^*$ with g = 2.0 (inset). Open and crossed symbols are from our analysis of $\rho_c(H,T)$ in Refs. [23] and [24], respectively. The shaded band covers $T^*(p)$ in cuprates [25] taken from Fig. 26 of Ref. [1].

In contrast to $H_{pg}(p)$, the doping dependence of the peak field H_{sc} is weak and roughly follows a parabolic dependence similar to $T_c(p)$. Note that H_{sc} does not represent an upper critical field H_{c2} . The boundary at H_{c2} in high- T_c superconductors is a fuzzy crossover difficult to estimate [26], but should be higher than H_{sc} [13]. If strong phase fluctuations associated with the precursor superconductivity are responsible for the pseudogap state [12], one would expect $H_{pg} \gg H_{c2} \gtrsim H_{sc}$ in the zero-temperature limit since quantum phase fluctuations should also be significant. In the underdoped regime, the difference between H_{pg} and H_{sc} in the low temperature limit is huge, which in this scenario can be attributed to quantum fluctuations. Surprisingly, in the overdoped regime, while the pseudogap region in the H-T diagram is still nontrivially large, the low temperature values of H_{pg} and H_{sc} are nearly the same, which may raise questions about large quantum fluctuations.

Our finding that Zeeman splitting closes the pseudogap implies that the triplet spin excitation at high fields overcomes the singlet pair correlations responsible for the gap in the spin spectrum and that the orbital contribution is very small. In preformed pair scenarios, our results would require pairing correlations on relatively short length scales with negligible orbital motion of pairs. This may be satisfied in a class of models, where charges (holes) self-organize into microstripes below T^* [27,28]. The mechanism of pairing is the generation of the "spin gap" in spatially confined Mott-insulating regions with local antiferromagnetic correlations in the proximity of the metallic stripes [27]. However, any future theoretical input must reconcile such pairing with a lack of significant quantum fluctuations in the overdoped regime.

A spin gap in a doped Mott insulator also appears in resonating-valence-bond theory, where the spin and charge degrees in the CuO₂ plane are separated into "spinons" and "holons" [29]. Studies based on this idea [30] derive a doping-dependent spin-gap temperature evolving from zero on the overdoped side to a finite value prescribed by the antiferromagnetic exchange $J(\sim 1000 \text{ K})$ as $p \rightarrow 0$. This spin-gap temperature corresponds to the formation of spinon singlet pairs. A gap in the spin excitation spectrum can be seen in the *c*-axis tunneling spectra, since during the interplane tunneling process spinons and holons recombine into conventional carriers with charge and spin. Our empirical linear scaling of $H_{pg}(p)$ and $T^*(p)$ gives an energy scale ~ 930 K in the $p \rightarrow 0$ limit, of the order of J.

Our results up to 60 T point to a predominant role of spins in the formation of the pseudogap consistent with models based on a doped Mott insulator [27–30]. An interesting issue concerns the existence of a quantum critical point at which the pseudogap temperature goes to zero [31]. This has been argued to occur at a critical doping $p \approx 0.19$ [22]. However, our most overdoped crystal with $p \approx 0.22$ has the pseudogap still unmistakably prominent [32], likely reflecting higher sensitivity of the interlayer tunneling at $(\pi, 0)$ points on the Fermi surface [14,15], where a spectral weight depletion onsets at a higher temperature [2].

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Note added.—A recent preprint [33] discusses different sensitivities of T_c and T^* to low fields within a BCS-based approach extended to arbitrary coupling.

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