Strongly nonlinear magnetization above $T_c$ in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$.

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Using high-resolution magnetometry we have investigated in detail the magnetization $M$ above the critical temperature $T_c$ in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$. In a broad range of temperature $T$ above $T_c$, we find that $M(T, H)$ is strongly non-linear in the field $H$. We show that as $T \rightarrow T_c$, the susceptibility $\chi(T, H)$ diverges to very large values ($\chi \rightarrow -1$) if measured in weak $H$. In addition, $M(H)$ displays an anomalous non-analytic form $M \sim H^{1/3}$ in weak fields with a strongly $T$-dependent exponent $\delta(T)$. These features strongly support the proposal that, above $T_c$, the pair condensate survives to support significant London rigidity.

I. INTRODUCTION

The notion that superconductivity in the cuprates is destroyed by thermally created vortices has gained substantial experimental support. In single crystals, evidence for vortex excitations that persist above the critical temperature $T_c$ to a temperature $T_{\text{onset}} \sim 120$ K has been obtained from the Nernst effect [1]. An enhanced diamagnetic signal that scales accurately as the Nernst signal has also been observed using high-field magnetometry [2]. These results reveal that, above $T_c$, significant pair-condensate amplitude survives up to intense magnetic fields $H$. Further, in ultra-thin films, the kinetic inductance has been observed to persist above $T_c$ [3]. The sensitivity of superconductivity to phase fluctuations in cuprates has been investigated theoretically by several groups [4, 5].

In low-$T_c$ superconductors, the condensate vanishes above $T_c$, except inside evanescent droplets created by amplitude (or Gaussian) fluctuations. The diamagnetic susceptibility $\chi$ from Gaussian fluctuations is very small ($\chi \sim 10^{-5}$) [6]. Given the vortex scenario in cuprates, the magnetization $M$ measured above $T_c$ ought to be qualitatively different from the Gaussian picture. However, resolving the signal in tiny crystals is very challenging, and the available results are confusing [7, 8, 9, 10]. An experiment performed on aligned grains [7] claims agreement with the mean-field Gaussian theory. Other experiments are seemingly consistent with the vortex scenario, but uncover anomalies that are hard to interpret [8]. Here we report detailed measurements in Bi$_2$Sr$_2$CaCu$_2$O$_8$ (Bi 2212) showing that the weak-field susceptibility $\chi$ diverges exponentially consistent with Kosterlitz-Thouless (KT) behavior. The strong nonlinearity observed in $\chi$ suggests the existence of a subtle rigidity in the pair wave function $\Psi$ which persists above $T_c$.

II. EXPERIMENTAL DETAILS

To cover a broad range of fields, we have combined torque magnetometry with SQUID magnetometry. In the torque experiment, the crystal is glued at the tip of the Si cantilever beam with its $c$-axis at a tilt-angle $\varphi \approx 15^\circ$ to $H$. In our experiment, the torque $\tau$ is the sum of a paramagnetic term from the spin moment $m_p$ and a diamagnetic term from the magnetization $M$ of interest here, viz. $\tau = |m_p + VM| \times \mu_0 H$, with $V$ the sample volume and $\mu_0$ the permeability. For temperature $T > T_c$, $M$ is strictly along the $z$-axis (we take $\hat{z} = \hat{\varepsilon}$). An in-plane component $M_x$ is resolvable only below the irreversibility line [11] (from hereon, $M_x = M$). We define the effective magnetization $M_{\text{eff}} \equiv \tau/(\mu_0 H_x V)$ where $H_x = H \sin \varphi$. 

FIG. 1: Magnetization $M$ vs. $H$ measured in Sample 1 in fields $|H| < 1.000$ Oe (Panel a) and in fields $|H| < 40$ Oe (b). $M$ is measured in the SQUID magnetometer in the long-time averaging mode ($\sim 12$ h per curve). In Panel (a), the sharp notch near $H = 0$ for $T < T_c (= 86$ K) is $H_{c1}$. These features are shown more clearly in Panel (b) with a 25-fold expansion in the field scale. The dotted line is $M/H = \chi/\langle 1 + N_c \chi \rangle = 2.55$ with $\chi = -0.95$ and $N_c = 0.64$ for Sample 1 with $H||\hat{\varepsilon}$. The peaks locate $H_{c1}$. 

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For \( \varphi \ll 1 \), we have

\[
M_{eff}(T, H) = \Delta \chi_p H_z + M(T, H_z),
\]

where the anisotropy \( \Delta \chi_p = \chi_c - \chi_a \) is the difference between the \( c \)-axis and in-plane spin susceptibilities. As explained in Ref. \[2\], the very weak \( T \) dependence of \( \Delta \chi_p \) in cuprates allows the strongly \( T \)-dependent \( M(T, H) \) to be extracted reliably from \( M_{eff} \). All curves reported here are fully reversible (hystereses caused by pinning of vortices are only seen below \( \sim 50 \) K). We studied 2 slightly underdoped Bi 2212 crystals with \( T_c = 86 \) and 85 K (Samples I and II) with very similar results.

If \( M \) is nearly linear in \( H \) (valid if \( T \geq 1.1 T_c \) and \( H > 0.5 \) T), Eq. \( 1 \) implies that the torque varies sinusoidally, viz. \( \tau/V = \mu_0(\Delta \chi_p - \chi)H_zH_z \equiv A_1 \sin 2\varphi \), where \( A_1(T, H) \sim H^2 \) and changes sign above \( T_c \), as observed in cuprates \( [1] \) (see below).

III. MAGNETIZATION AND SUSCEPTIBILITY

First, we discuss the curves of \( M(H) \) below \( T_c \). The curves of \( M(H) \) in Fig. \( 1 \) were measured by SQUID magnetometry. Much analysis have focused on the curve of \( M(H) \) at the “crossing temperature” \( T_s (= 84 \) K) at which \( M(H) \) is very nearly \( H \) independent. By detecting the flux-exclusion in very weak fields, we have determined that the 3D transition at \( T_c \) (86 K) actually lies 2 K above \( T_s \). In the interval \( T_s-T_c \), a prominent anomaly of the curves of \( M(H) \) is that they curve steeply towards zero as \( H \to 0 \) even though full flux exclusion obtains when \( H < H_{c1} \) (the lower critical field). This feature may be seen in earlier reports on \( M \) \[12\], but has not received comment. Closer inspection reveals that \( M(H) \) displays a notch feature which we show in expanded scale in Fig. \( 3b \). The notch corresponds to the initial entry of vortices at \( H_{c1} \). Below \( H_{c1} \), flux exclusion gives a linear \( M-H \) variation with a steep slope given by \( M/H = \chi/(1 + N_c \chi) \), where \( N_c \simeq 0.64 \) is the demagnetization factor. Taking \( \chi = -0.95 \), we have \( M/H = -2.55 \) which is plotted as the broken line in Fig. \( 3b \). Both the full flux-exclusion implied by \( \chi \sim 1 \) and the sharp onset are evidence for a high degree of electronic homogeneity and uniformity of the condensate.

In low-\( T_c \) superconductors, \( |M| \) falls steeply above \( H_{c1} \), and continues its monotonic decrease until \( H \) attains the upper critical field \( H_{c2} \). Here, the behavior is qualitatively different. Just above \( H_{c1} \), \( |M(H)| \) falls to a minimum, but then slowly increases over several decades in \( H \). Following the initial entry of vortices, the sample steadily becomes more diamagnetic with increasing \( B \). As we show, this paradoxical pattern reflects the phase-disordering nature of the transition at \( T_c \) in hole-doped Bi 2212.
FIG. 4: The temperature and field dependence of the susceptibility \( \chi(T, H) = M(T, H)/H \). In weak \( H \), \(|\chi|\) increases by over 5 decades from 100 K to \( T_c \), reaching the full flux-expulsion value (enhanced by demagnetization) shown by the arrow. The dashed curve is a fit to Eq. 3 with \( b = 1 \), \( d = 12 \, \text{Å} \), \( a = 10 \, \text{Å} \), and \( T_{KT} = 84 \, \text{K} \). Open circles represent the KT correlation length \( \xi_{KT} \) obtained from the fit.

We turn next to temperatures above \( T_c \), where the resolution of the torque cantilever (\( \sim 10^{-9} \, \text{emu at 10 T} \)) allows \( M \) to be studied in detail. Figure 2 displays the \( H \) dependence of \( M \) at temperatures increasing from near \( T_c \) to 100 K. Over this broad interval, \( M \) increases with \( H \) with a curvature that becomes increasingly pronounced near \( T_c \). These curves are strikingly similar to those in the interval \( T_s - T_c \), except for the conspicuous absence of the notch feature associated with a finite \( H_{c1} \).

In Fig. 4, the susceptibility \( \chi(T, H) = M(T, H)/H \) is displayed versus \( T \), with \( H \) as the parameter. Evidently, \( \chi(T, H) \) is highly sensitive to \( H \) especially near \( T_c \). If \( \chi \) is measured at a fixed \( H \), say 0.5 T, it stays non-divergent across \( T_c \). However, this is not intrinsic. Instead, from 105 to 86 K, \(|\chi|\) measured in a very weak field increases by 5 decades to attain values 0.1–1 just above \( T_c \). As discussed later, this steep increase reflects a rapidly changing phase correlation length.

Previous experiments on “fluctuation diamagnetism” in cuprates failed to observe a divergent \( \chi \) because the \( H \) applied was too large \( \cite{2, 7, 8, 10} \). The strong \( H \) dependence in \( \chi(T, H) \) now call their conclusions into question. Further, vortex pinning effects, especially strong in polycrystalline samples of La\(_{2-x}\)Sr\(_x\)CuO\(_4\) and YBa\(_2\)Cu\(_3\)O\(_{7-x}\), introduce extrinsic features in \( M \) which render analyses problematic \( \cite{13} \).

IV. KT CORRELATION LENGTH

There are 2 major aspects of the non-linearity in \( M(T, H) \) above \( T_c \). The first reflects the divergent growth of the phase correlation length \( \xi(T) \) while the second is the singular behavior in Eq. 2. The 2D phase correlation length measures the region within which phase coherence prevails in each layer. In KT theory long-range phase coherence is destroyed by the appearance of (anti) vortices...
at the KT transition (at $T_{KT}$). Hence the KT correlation length $\xi_{KT}$ is the average spacing between these thermally created vortices. Above $T_{KT}$, $M(T, H)$ is linear in $H$ in low fields. The weak-field susceptibility is

$$\chi_{KT} = -\frac{\mu_0 k_B T}{2d \phi_0} \xi_{KT}, \quad (T > T_{KT}, H \to 0),$$

(3)

where $k_B$ is Boltzmann’s constant, $d$ the bilayer spacing, and $\phi_0$ the superconducting flux quantum. The KT correlation length is exponential in the reduced temperature $t' = T/T_{KT} - 1$, viz. $\xi_{KT} = a e^{b/\sqrt{T}}$, with $b \sim 1$ a non-universal constant and $a$ a cut-off length $\sim$ the vortex core size.

Clearly, an interlayer coupling $J_L$ needs to be included to describe the actual 3D transition at $T_c$ (see below). Nonetheless, the KT theory describes the essential features of a divergent $\xi(T)$ and the roles of thermally-induced vs. field-induced vortices in each layer. In Fig. 4 the dashed curve is a fit of Eq. 3 with $b = 1.0$ and $a = 10 \, \AA$. The fit captures quantitatively the broad features of the 5-decade rise in $\chi(T, H)$ in weak $H$, matching its curvature both near $T_c$ and near 100 K.

The KT correlation length $\xi_{KT}$ inferred from the fit increases from $\sim 5$ nm at 105 K to 700 nm at 86 K. A second length scale is the spacing $a_B = \sqrt{\phi_0 / B}$ of vortices inserted by $H$. To observe the intrinsic divergence in $\xi_{KT}$, we need $a_B \gg \xi_{KT}$ or $B \ll B_{th}$, where $B_{th} = \phi_0 / \xi_{KT}^2$ is a field-scale set by the density of thermally created (anti)vortices. As $T \to T_c$ in a fixed field $B \ll B_{th}$, $\chi(T)$ initially increases rapidly (and $B_{th}$ decreases). However, $\chi$ saturates to a constant when $B_{th}$ approaches $B$, as evident for the curves in Fig. 4.

V. SINGULAR RESPONSE AS $H \to 0$

Despite the reasonable fit to $\chi(T)$, Eq. 3 does not account for the non-analytic behavior expressed in Eq. 2. As mentioned, the deviation between $M(H)$ at 87 K and the dashed line in Fig. 3 grows as $H$ decreases. We now discuss in more detail the $T$ dependence of this non-analyticity. From 105 K to 84 K, the weak-field slopes in the log-log plots in Fig. 3 decrease smoothly from $\sim 1$ to zero, i.e. $\delta(T)$ diverges to values exceeding 20. The steepness of this increase is evident in the plots in Fig. 5 for Samples I and II (solid and open circles, respectively). The continuous nature of the increase in $\delta(T)$ is made clearer by plotting its reciprocal, which falls below 1 and smoothly approaches 0 as $T \to T_c^+$ (triangles). In Fig. 6 the shading highlights the interval from $T_s$ to $\sim 105$ K in which $\delta$ exceeds 1. In the shaded interval above $T_c$, $\chi$ diverges without limit in the limit $H \to 0$ assuming Eq. 2 remains valid. Hence, despite the absence of a true Meissner state, a feebly $H$ induces a very large diamagnetic screening current. Below $T_c$, however, this limit is interrupted at $H_s$ by the appearance of the Meissner state, as discussed in Fig. 4.

Generally, the Meissner state originates from the rigidity of the pair wave function $\Psi$. In a field, this “London rigidity” suppresses the paramagnetic current response, leaving only the full diamagnetic current $\Psi$. Here, our magnetization results imply that, despite the loss of the Meissner effect, $\Psi$ retains some rigidity above $T_c$. However, the rigidity is very fragile and observable only in a feeble field. The existence of a singular diamagnetic response above $T_c$ has not been predicted (for e.g., $\delta$ is strictly 1 above $T_{KT}$ in KT theory). Singular behavior in the weak-field vortex-liquid response is discussed in Ref. 20, and in Ref. 21.

VI. DISCUSSION

We now return to the anomalous behavior of $M(H)$ in the interval $T_s - T_c$ where full flux-exclusion prevails for $H < H_{c1}$. As mentioned, the curves below $T_c$ closely resemble those above (compare Figs. 1a and 2). Aside from fields below $H_{c1}$, the curves of $M$ in the log-log plot in Fig. 3 assume a fan-like dispersion that smoothly extends the behavior in this interval to above $T_c$. The exponent $1/\delta(T)$, which measures the slopes, continuously decreases across $T_c$ (Fig. 5). This continuity strongly implies that the anomalous magnetization in the 2-K interval below $T_c$ and above $T_c$ share the same origin; both represent the diamagnetic response of a strongly phase-disordered 2D superconducting state with a very high depairing field $H_{c2} > 80$ T. Between $T_s$ and $T_c$, the magnetization is comprised of a weak-field 3D Meissner state that pre-empts the phase-ordering transition of the 2D condensate at $T_s$. Application of $H > H_{c1}$ destroys the Meissner state and uncovers the underlying 2D state whose magnetization continues to grow with field. This accounts for the unusual “minimum” in $|M|$ just above $H_{c1}$ mentioned in Fig. 4. The steep fall of $|M|$ just above $H_{c2}$ reflects the destruction of the 3D Meissner state, while the subsequent power-law increase reflects the behavior of $M$ in the underlying 2D condensate which is robust to intense fields.

A hint of our results may be seen in previous torque experiments 2 11. In crystals of Tl2Ba2CuO6+x with $T_c = 25$ and 15 K, the measured curve of $\tau$ vs. $\varphi$ displays a puzzling, unexpected term $A_1 \sin 2\varphi$ which is actually dominant above $T_c$. Using Eq. 2 and our analysis, we now understand this term to be the robust 2D magnetization which persists to intense $H$ and high $T$ as discussed above. As noted, the rapid decrease in $|M|$ above $T_c$, compared with the mild change in $\Delta \chi_p$, engenders a sign change in $A_1(T, H)$ as seen (Fig. 5 in Ref. 11).

The transition in cuprates has been frequently compared to the 3DXY transition with large anisotropy $\alpha^{-1}$, where $\alpha = J_L / J$ with $J$ the intralayer coupling and $J_L$ the interlayer coupling 12 12 16. Our findings that the 2DKT theory gives a broadly accurate description of $\chi(T, H)$ and that $T_s - T_{KT} \sim 2K \ll T_c$ confirm that Bi 2212 falls in the extreme limit $\alpha \ll 1$. [By way of com-
parison, in the planar ferromagnet $K_{2}CuF_{4}$ with $\alpha \ll 1$, the KT theory also describes well $M$ vs. $H$ above its Curie temperature $T_{C}$ \[14\]. However, the 3D transition $T_{C} = 6.25$ K pre-empts the 2D KT transition ($T_{KT} \sim 5.6$ K) \[17\]. Below $T_{C}$, the destruction of the 3D Meissner state here obviously has no parallel in the planar magnet.

Unfortunately, the limit $\alpha \ll 1$ is intractable analytically. Currently, numerical simulations of $M$ on lattices \[15, 16\] are not accurate enough to compare with our measurements, but we hope the detailed measurements here will motivate simulations on larger lattices.

As we cool from 300 K, the first signs of the pair condensate gradually appear at $T_{onset} \sim 120$ K as a vortex-Nernst signal \[1\] and an enhanced diamagnetic susceptibility \[2\]. The phase of $\Psi$ is strongly disordered by vortex motion. However, the continued growth of these signals to intense fields (30-45 T) implies that the depairing field $H_{c2}$ lies much higher (80-100 T). Below $\sim 105$ K, the non-linear behavior of $\chi$ becomes increasingly apparent. In weak $H$, $\chi(T, H)$ undergoes a 5-decade increase as $T_{c}$ is approached, whose broad features can be understood in terms of a diverging phase-correlation length $\xi$, as prescribed in KT theory (Fig. 4). In the shaded interval in Fig. 5 $\chi$ is further enhanced by a singular power-law dependence suggestive of increasing London rigidity.

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