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Dependence of Upper Critical Field and Pairing Strength on Doping in Cuprates

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We have determined the upper critical field \( H_{c2} \) as a function of hole concentration in bismuth-based cuprates by measuring the voltage induced by vortex flow in a driving temperature gradient (the Nernst effect), in magnetic fields up to 45 tesla. We found that \( H_{c2} \) decreased steeply as doping increased, in both single and bilayer cuprates. This relationship implies that the Cooper pairing potential displays a trend opposite to that of the superfluid density versus doping. The coherence length of the pairs \( \xi_0 \) closely tracks the gap measured by photoemission. We discuss implications for understanding the doping dependence of the critical temperature \( T_{c0} \).

The superconducting state in a metal is completely suppressed if a sufficiently strong magnetic field is applied. In individual type-II superconductors, the field required—defined as the upper critical field \( H_{c2} \)—is an important parameter because it determines the value of the coherence length \( \xi_0 \) (the size of the Cooper pair) as well as the strength of the pairing potential; the higher the field \( H_{c2} \), the stronger is the pairing potential and the smaller the pair size (1). In the phase diagram of the cuprates, superconductivity has been observed in the range of hole concentration 0.05 < \( x \) < 0.25. Many parameters of the superconducting state, notably the superfluid density and superconducting gap, have been measured as a function of \( x \). The conspicuous exception is \( H_{c2} \), which is uncertain for reasons discussed below. Because even the basic trend of \( H_{c2} \) versus \( x \) is unknown, the crucial question of whether the pairing strength, as distinct from the superfluid density, increases or decreases with \( x \) remains unanswered. We report measurements of \( H_{c2} \) versus \( x \) in the Bi-based cuprates using the vortex-Nernst effect. In both single and bilayer systems, it was found that \( H_{c2} \) (and hence the pairing potential) steeply decreased as \( x \) increased. We show that \( \xi_0 \) is intimately related to the gap measured by angle-resolved photoemission spectroscopy (ARPES) (2) and results from scanning tunneling microscopy (STM) (3, 4).

In the Nernst effect (3–11), vortices in the vortex liquid state are driven down an applied temperature gradient \( -\nabla T \). Their velocity \( v \)

E. M. Bowman, V. J. Brown, C. Kertzman, U. Schwarz, and others discussed below. Because even the basic trend of \( H_{c2} \) versus \( x \) is unknown, the crucial question of whether the pairing strength, as distinct from the superfluid density, increases or decreases with \( x \) remains unanswered. We report measurements of \( H_{c2} \) versus \( x \) in the Bi-based cuprates using the vortex-Nernst effect. In both single and bilayer systems, it was found that \( H_{c2} \) (and hence the pairing potential) steeply decreased as \( x \) increased. We show that \( \xi_0 \) is intimately related to the gap measured by angle-resolved photoemission spectroscopy (ARPES) (2) and results from scanning tunneling microscopy (STM) (3, 4).

In the Nernst effect (3–11), vortices in the vortex liquid state are driven down an applied temperature gradient \( -\nabla T \). Their velocity \( v \)
induces an electric field $E_y = B v_y$, which may be detected with high sensitivity (the induction field $B \parallel z$). The total Nernst signal $e^\text{obs}$ is $E_y/|\Psi(T)|$ is the sum of the vortex signal $e_c$ and the carrier contribution $e^\text{c2}$: $e^\text{obs} = e_c + e^\text{c2}$.

Although our focus is on the Bi-based cuprates, we gain perspective and insight by first looking at the electron-doped cuprate Nd$_{2-x}$Ce$_x$CuO$_4$ (NCCO), in which the effects of fluctuations are weak and superconductivity is easily suppressed in fields of $\sim 10$ T. Previous Nernst measurements have revealed a sizable carrier signal $e^\text{c2}$ in NCCO (10, 11). We observed that $e_c$ has a distinctive “tent” profile that distinguishes it from $e^\text{c2}$ (fig. S1). The vortex signal $e_c$ at fixed $T$ was initially zero until $H$ exceeded the solid-to-liquid melting field $H_{c2}(T)$ (Fig. 1A). In the liquid state, the vortex signal rises steeply to a maximum before falling monotonically to zero as $H$ approaches $H_{c2}$, with a similar profile occurring in low-critical $T$ (low-$T_x$) superconductors (7, 8). The steep rise just above $H_{c2}$, which scales accurately with the in-plane resistivity $\rho$ (dashed line), reflects the sharp increase in vortex mobility, whereas the fall at higher fields is caused by the field suppression of the condensate amplitude as $H$ approaches $H_{c2}$. The phase diagram in Fig. 1B, derived from $e_c$, reveals an $H_{c2}$ boundary similar to that observed in conventional type-II superconductors and well described by the Bardeen, Cooper, and Schrieffer (BCS) theory (I). It is weakly $T$-dependent at low $T$ and decreases to zero linearly as $T$ approaches $T_{c0}$, the zero-field transition temperature [the linear field $H_{c2}$ near $T_{c0}$ agrees with that in an earlier Nernst study (11)]. The vortex signal $e_c$ rapidly vanishes above $T_{c0} = 24.5$ K, indicating that the amplitude of the order parameter $\Psi$ vanishes at $T_{c0}$. Hence, the phase diagram of NCCO is similar to the phase diagram of conventional superconductors (I), except that the vortex liquid region is greatly expanded. This example shows that $H_{c2}$ at finite $T$ is reliably determined as the field at which the vortex-Nernst signal reaches zero: The tent profile of $e_c$ versus $H$ (a sharp rise to a peak and a fall to zero at high fields) defines the entire vortex liquid region, in which $|\Psi|$ remains finite. From $H_{c2}(0) \sim 10$ T, we find $\xi_0 \sim 58$ Å in NCCO.

Most attempts to find $H_{c2}$ in cuprates have relied on measuring the field profiles of the resistivity $\rho$ in intense fields. We next show that this method is highly unreliable. The field profile of $\rho$ at 14 K (Fig. 1A, dashed line) reveals that, above $H_{c2}$, it rises steeply toward the normal-state value $\rho_n$. Long before the field reaches $H_{c2}$, $\rho$ in the vortex-liquid state becomes indistinguishable from $\rho_n$. If we had used the “knee” in the profile of $\rho$ to estimate $H_{c2}$, we would have erroneously identified $H_{c2}$ with the “ridge field” $H^p(T)$ defined (7) by the maximum in $\rho$ at each $T$. As shown in Fig. 1B, the ridge field $H^p(T)$ versus $T$ has a positive curvature and remains strongly $T$-dependent as $T$ approaches zero. We note that many $H_{c2}$ curves derived from $\rho$ in cuprates share these features of $H^p(T)$ (12–14). The inset shows that at finite $T$, the true $H_{c2}(T)$ is considerably higher than $H^p(T)$. In the hole-doped cuprates, the difference between $H^p$ and $H_{c2}$ is even greater (7).

With this caveat in mind, we turn to the single-layer cuprate Bi 2201 (Bi$_2$Sr$_2$La$_2$CuO$_8$) and the bilayer Bi 2212 (Bi$_2$Sr$_2$CaCu$_2$O$_8$), which are ideal for exploring the $x$ dependence of $H_{c2}$ because $e^\text{c2}$ is negligibly small, and the anomalously small $H_{c2}(T)$ values allow the scaling studies described below to be extended over the broadest field range. In Bi 2201 (sample A1, $x = 0.16$, $T_{c0} = 28$ K), $e_c$ increases to a broad maximum and then decreases monotonically to zero at just above 45 T (Fig. S2). The tent profile, similar to that in NCCO except for the higher field scales, again reveals how far the vortex liquid extends in the field.

A major difference between NCCO and the hole-doped cuprates arises from strong fluctuations in the phase of $\Psi$ in the latter. Whereas the vortex signal in NCCO rapidly vanishes just above $T_{c0}$, it remains large at $T_{c0}$ in Bi 2201, La$_2$Sr$_2$CuO$_4$ and YBa$_2$Cu$_3$O$_7$ (YBCO) and extends considerably above $T_{c0}$. In all hole-doped cuprates examined to date (5–7, 9), $T_{c0}$ has corresponded to the loss of long-range phase coherence (15–17), rather than the vanishing of $|\Psi|$. Defining the field at which $e_c$ approaches zero as $H_{c2}(T)$ at each $T$, we found that $H_{c2}(T)$ in Bi 2201 is nearly $T$-independent from 5 to 30 K (it goes to zero only at much higher $T$). Despite the non-BCS scenario in hole-doped cuprates, $H_{c2}(T)$ is still reliably obtained from the approach of $e_c$ to zero.

Comparison of $e_c$ versus $H$ in several samples of Bi 2201 of different $x$ revealed a distinctive trend. Figure 2, A and B, compares the $e_c$-$H$ curves in overdoped Bi 2212 (sample B1, $x = 0.22$, $T_{c0} = 65$ K) with those in underdoped Bi 2212 (sample B3, $x = 0.087$, $T_{c0} = 50$ K). The calibration of $x$ is discussed in (18). In sample B1 (Fig. 2A), the vortex Nernst signal closely resembled those in Bi 2201 and anticipated the scaling property to be described. In the underdoped sample (Fig. 2B), however, the curves

Fig. 1. (A) The vortex-Nernst signal $e_c$ versus $H$ in Nd$_{2-x}$Ce$_x$CuO$_4$ ($x = 0.15$, $T_{c0} = 24.5$ K). For example, at 14 K, $e_c$ appears at $H_{c2} = 1.1$ T, rises to a peak at $H^p = 2.8$ T, and decreases, with $H_{c2} = 5.8$ T. The profile of $\rho$ at 14 K (dashed line) matches the initial increase in $e_c$. Above $T_{c0}$, the vortex signal rapidly vanishes (unlike in hole-doped cuprates), $\mu_0$ is the vacuum permeability. (B) The $T$ dependence of $H_{c2}$, $H^p$, and $H_{c2}$ derived from the $e_c$ curves. The $H_{c2}$ curve in NCCO is conventional and terminates close to $T_{c0}$.

Fig. 2. Comparison of the profiles of $e_c$ in (A) overdoped (OD) $T_{c0} = 65$ K) and (B) underdoped (UD) $T_{c0} = 50$ K) crystals of Bi$_2$Sr$_2$CaCu$_2$O$_8$. The curves in (A) peak at relatively low fields (5 to 10 T) and decrease by 50 to 60% when $H$ reaches 30 T. The peaks in (B), however, lie much closer to 30 T.
were more stretched out along the field axis. In the temperature range of 40 to 50 K, $e_x$ was not appreciably diminished from its peak value at the maximum field 30 T, whereas in Fig. 2A $e_x$ had decreased from its peak by more than 60% by 30 T. The trend is summarized (Fig. 3A) for overdoped, optimally doped, and underdoped Bi 2212 (samples B1 to B3), using profiles of $e_x$ measured at their respective $T_{c_0}$'s (65, 90, and 50 K in B1, B2, and B3, respectively). It is apparent that successively higher fields are needed to achieve comparable suppression of $e_x$ as we go from the overdoped to the optimum and to the underdoped sample. The same pattern is observed in Bi 2201 (with lower field data).

We found that, if we used the reduced field $h = H/H_{c2}$' as abscissa (the prime indicates the value at $T_{c0}$), the three traces in Fig. 3A accurately matched the template curve ($18$) of sample A1 (at $T_{c0} = 28$ K) with the right choice of $H_{c2}'$ (Fig. 3B). Using the known $H_{c2}'$ value in A1 (50 T), we found that $H_{c2}' = 50, 67, \text{ and } 144\ T$ in B1, B2, and B3, respectively. A similar scaling was observed in three samples of Bi 2201 (Fig. 3C). The scaling behavior enabled us to accurately track the $x$ dependence of $H_{c2}'$ to field values considerably higher than 45 T.

The scaling behavior continued to hold at $T$ below $T_{c0}'$, if comparisons were made between curves at the same reduced temperature $t = TT_{c0}'$. However, below $t \sim 0.7$, the vortex solid phase below $H_{c2}$ (where $e_x = 0$) expanded considerably and precluded meaningful comparison for scaling behavior. This was not a serious drawback because the $T$ dependence of $H_{c2}'$ was very weak between 0 K and $T_{c0}'$ in the Bi-based cuprates (the prime on $H_{c2}'$ is dropped hereafter).

We carried out the scaling comparison for five samples of Bi 2212 with $0.087 < x < 0.22$, as well as in three samples of Bi 2201, and obtained the variation of $H_{c2}'$ versus $x$ displayed (Fig. 4A). As the doping $x$ decreases, $H_{c2}'$ in Bi 2212 undergoes a steep increase from 50 to 144 T, whereas in Bi 2201 it increases from 42 to 65 T. The gap amplitude $\Delta_0$ in Bi 2212, measured at low $T$ by ARPES (2), also increases as $x$ decreases (Fig. 4A, upper stripe). Most investigators regard this increasing $\Delta_0$ as the normal-state gap in the pseudogap state because it persists unchanged in size ($2$) to temperatures well above $T_{c0}'$.

We next show that our values for $H_{c2}'$ in Bi 2212 are in fact closely related to $\Delta_0$. The two quantities may be compared directly if we convert them to length scales. Expressing $H_{c2}'$ as the coherence length via $\xi_0 = (\hbar v_F/2\pi H_{c2})$, we found that $\xi_0$ decreases from 26 to 15 $\AA$ as $x$ decreases from 0.22 to 0.087 ($v_F$ is the Fermi velocity ($a = \pi$ for $s$-wave superconductors). The plots in Fig. 4B reveal that $\xi_0$ and $\xi_2$ are closely matched if we choose $a = 3/2$, consistent with extreme gap anisotropy. The agreement is strong evidence that the ARPES gap at low $T$ and the Nernst experiments are probing the same length scale over a broad range of $x$. Both experiments uncovered the same trend: The coherence length decreases by a factor of $\sim 2$ as $x$ decreases from 0.22 to 0.087. Moreover, the virtually $T$-independent behavior of $H_{c2}'$ in our experiment is consistent with the $T$-independent behavior in the ARPES gap. The comparison persuaded us that the ARPES $\Delta_0$ (2) represents the gap amplitude of the Cooper pairs.

High-resolution images of vortices in optimally doped Bi 2212 have been obtained by STM. The exponential decay of the quasiparticle state density yields a length scale of 22 Å (3). A longer length scale, $\sim 40$ Å, is defined by the checkerboard pattern imaged outside the core (4). The shorter length of 22 Å is in good agreement with our $\xi_0$ (Fig. 4B, open triangle), suggesting that the exponential fall-off is dictated by $\xi_0$.

Recently, the field scale $H_{pg}$ for suppressing the pseudogap state in Bi 2212 has been estimated from the $c$-axis resistivity $\rho_c$ versus $H$ (19). Insofar as $\rho_c$ probes changes in the single-particle density of states, rather than the pairing amplitude, the inferred $H_{pg}$ values should be distinguished from $H_{c2}'$ (indeed, the estimated $H_{pg}$ values, 100 to 550 T, are much higher than our $H_{c2}'$ values).

To address our initial question, the present experiment establishes that $H_{c2}'$ increases as $x$ decreases in both Bi 2201 and Bi 2212, which...
implies that the pairing potential is strongest in very underdoped samples. This finding may place strong constraints on theories of the pairing mechanism. The trend [opposite to that of the superfluid density \( n_s \) (20, 21), which decreases as \( x \) decreases] implies a scenario for underdoped cuprates that is radically different from that inferred from \( n_s \) and \( T_{c0} \) alone. The physical picture is that, in hole-doped cuprates, the pairing potential \( \Delta_0 \) is maximal in the underdoped regime and falls rapidly with increased hole density, as anticipated in early resonating-valence-bond theories (15, 16). However, the small \( n_s \) at small \( x \) renders the condensate highly susceptible to phase fluctuations (17). At \( T_{c0} \), spontaneous nucleation of highly mobile vortices destroys long-range phase coherence and the Meissner state (7, 22, 23), but vortex excitations remain observable to much higher \( T \) (5, 6). As \( x \) is increased, \( n_s \) and \( T_{c0} \) initially increase, but beyond \( x = 0.17 \), \( T_{c0} \) is suppressed by a steeply falling \( \Delta_0 \). The trade-off between \( n_s \) and \( \Delta_0 \) informs the entire cuprate phase diagram and accounts naturally for the dome shape of the curve of \( T_{c0} \) versus \( x \).

An interesting implication may be inferred in the limit of small \( x \). If the trend in \( \xi_0 \) in Fig. 4B persists, \( \xi_0 \) becomes smaller than the separation \( d \) of Cooper pairs in this limit \((d \sim 25 \, \text{Å} \text{ at } x = 0.05). \) For \( x < 0.05 \), we may treat the carriers as tightly bound pairs (bosons) that are well separated and too dilute to sustain long-range phase coherence. In uncovering the increased pairing strength at small \( x \), our experiment provides clues that bosonic pairs may exist in this limit.

References and Notes

18. Materials and methods are available as supporting material on Science Online.
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Supporting Online Material

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Materials and Methods

Figs. S1 and S2

References

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Three-Dimensional Mapping of Dislocation Avalanches: Clustering and Space/Time Coupling

Jérôme Weiss1 and David Marsan2

There is growing evidence for the complex, intermittent, and heterogeneous character of plastic flow. Here we report a three-dimensional mapping of dislocation avalanches during creep deformation of an ice crystal, from a multiple-transducers acoustic emission analysis. Correlation analysis shows that dislocation avalanches are spatially clustered according to a fractal pattern and that the closer in time two avalanches are, the larger the probability is that they will be closer in space. Such a space/time coupling may contribute to the self-organization of the avalanches into a clustered pattern.

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