High Field Phase Diagram of Cuprates Derived from the Nernst Effect

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Measurements of the Nernst signal in the vortex-liquid state of the cuprates to high fields (33 T) reveal that vorticity extends to very high fields even close to the zero-field critical temperature T_{c0}. In overdoped $La_{2-x}Sr_xCuO_4$, we show that the upper critical field $H_{c2}(T)$ curve does not end at T_{c0}, but at a much higher temperature. These results imply that T_{c0} corresponds to a loss in phase rigidity rather than a vanishing of the pairing amplitude. An intermediate field $H^*(T) \ll H_{c2}(T)$ is shown to be the field scale for the flux-flow resistivity.

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In the cuprate superconductors, strong fluctuations of the order parameter [1,2] and the extreme field scales make the task of constructing the experimental phase diagram a formidable challenge. While the important vortex-solid melting line $H_m(T)$ is now well known [3–5], there is great uncertainty in the region above H_m. A key difficulty is the *loss of long-range phase rigidity* in the transition to the vortex-liquid state. In the vortex-liquid state, the flux-flow resistivity ρ rises very rapidly to the (extrapolated) normal-state value [6] even though substantial condensate strength and pairing amplitude remain. This makes ρ an unreliable "diagnostic" of the field suppression of the pairing amplitude. By contrast, the Nernst and Ettinghausen effects can probe the existence of vorticity in the superfluid with undiminished sensitivity even in intense fields. From Nernst measurements in $La_{2-x}Sr_xCuO_4$ (LSCO) and $YBa_2Cu_3O_y$ (YBCO) in fields up to 33 T, we have uncovered an anomalous property of $H_{c2}(T)$ near their zero-field critical temperature T_{c0}. This anomaly is related to the existence of vortexlike excitations high above T_{c0} [7,8], and is central to the key question of whether the Meissner transition in zero field is caused by the collapse of long-range phase coherence or the vanishing of the pairing amplitude (see also Corson *et al.* [9]). In YBCO, previous Ettinghausen [10] and Nernst [11,12] measurements were performed in lower fields. Measurements on LSCO have been extended recently to high fields [13].

In the Nernst experiment, vortices moving with velocity $\mathbf{v}$ down a thermal gradient $-\nabla T \parallel \hat{\mathbf{x}}$ generate a Josephson voltage which is observed as a transverse E-field $E_y = B v_x$, with $\mathbf{B}$ the mean flux density. The vortex-Nernst effect is well explored in low-T_c superconductors. Figure 1b shows the Ettinghausen-Nernst effect in PbIn [14]. The depinning of the vortex lattice by a supercurrent ($\mathbf{J} \parallel \hat{\mathbf{x}}$) leads to a large transverse heat current $J_y^h \parallel \hat{\mathbf{y}}$ that rises to a maximum and then decreases to zero at H_{c2} (the weak fluctuation tail above H_{c2} is not relevant here). By Onsager reciprocity, the Nernst signal $e_y \equiv E_y/|\nabla T|$ shares the same profile as J_y^h in Fig. 1b [15]. To compare with theory, e_y is divided by $\rho(H)$ to obtain the "transport line-entropy" $s_\phi = \phi_0 e_y/\rho$ (Ts_ϕ is the heat energy carried by a unit-length vortex line). In PbIn, the linear decrease in s_ϕ near H_{c2} matches that of the magnetization $M(T,H) = -[H_{c2}(T) - H]/1.16[2\kappa^2 - 1]$ (κ is the Ginzburg-Landau parameter), as given by the Caroli-Maki expression [14,16],

$$s_\phi(T,H) = \frac{\phi_0}{T}\,|M(T,H)|L_D(T) \qquad (1)$$

[here $L_D(T)$ decreases gradually with T from 1 at T_{c0}].

We start with the overdoped regime, where the field profiles of e_y most resemble those in PbIn. In Fig. 1a, we display curves of e_y vs field H in sample 1 ($La_{2-x}Sr_xCuO_4$ with $x = 0.20$ and $T_{c0} = 28$ K) (for experimental details, see Ref. [8]). At each temperature T, the signal rises steeply above H_m, attaining a prominent maximum before decreasing. The total data set defines experimentally the region in H and T where vorticity is strongly present. As in PbIn, the monotonic decrease at high fields reflects the field suppression of the condensate strength. At high fields, all the curves below 14 K are observed to follow a common curve towards zero (dashed line). The trend implies that the low-T traces are all wedged between the field axis and the broken line. Hence all the low-T curves vanish at the intercept of the common curve with the field axis (45–50 T), which corresponds to $H_{c2}(0)$.

Going to higher T, we immediately encounter an anomaly. Conventionally, the H_{c2} line goes linearly to zero at T_{c0}. Hence, e_y ought to be finite in a field interval that $\to 0$ as $T \to T_{c0}^-$. In sharp contrast, we find that, close to T_{c0}, the magnitude of e_y remains large and nearly unchanged up to intense fields. There is no evidence for a field scale (flagged by $e_y \to 0$) that decreases to zero regardless of how close we are to T_{c0}. To make this point quantitative, we convert e_y to s_ϕ using $\rho(T,H)$ measured in the same sample. The line entropy rises steeply to a sharp maximum before decreasing monotonically (Fig. 1c). The linear decrease at high fields, strikingly similar to that in PbIn, allows $H_{c2}(T)$ to be determined [17]. These results

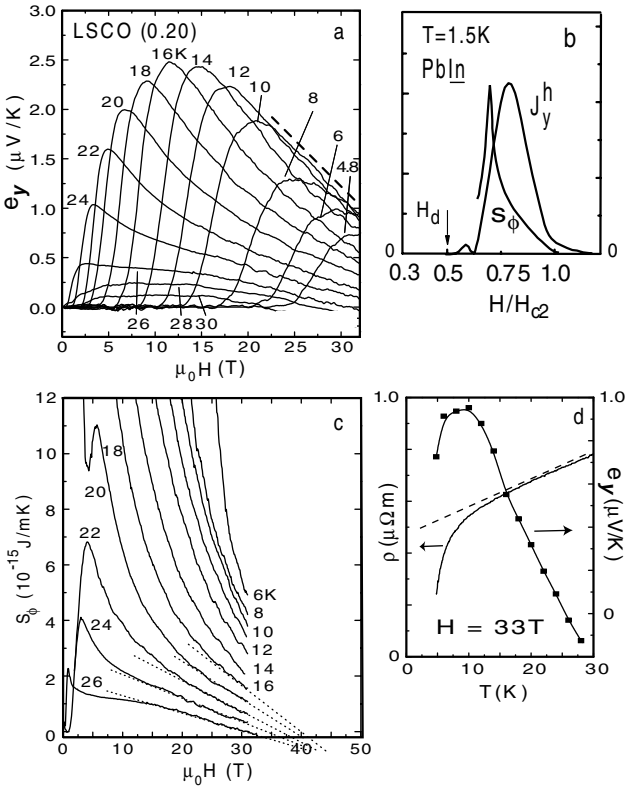

FIG. 1. (a) The field dependence of $e_y \equiv E_y/|\nabla T|$ at indicated T in sample 1 (LSCO with $x = 0.20$, $T_{c0} = 28$ K). Below 25 K, e_y rises to a sharp peak before decreasing monotonically. The dashed line indicates asymptotic behavior at large-H and low-T. (b) Plots of the transverse heat flux J_y^h and s_ϕ vs H in an Ettinghausen experiment in PbIn at 1.5 K (by reciprocity, $J_y^h = e_y/JT$, with J the applied supercurrent). H_d is the depinning field (modified from Vidal [14(a)]). (c) The field dependence of the transport line entropy s_ϕ obtained from e_y and ρ in sample 1 (LSCO, $x = 0.20$). The high-field linear extrapolations (dotted lines) provide estimates of $H_{c2}(T)$. Fits to Eq. (1) give $\kappa = 670$ and 500 at 26 and 18 K, respectively. (d) The temperature dependence of ρ and e_y in sample 1 in a fixed field (33 T). The dashed line is the extrapolation of ρ_N. Note that e_y (measured at 33 T) goes to zero at ~26 K instead of ~9 K (the "knee" in ρ).

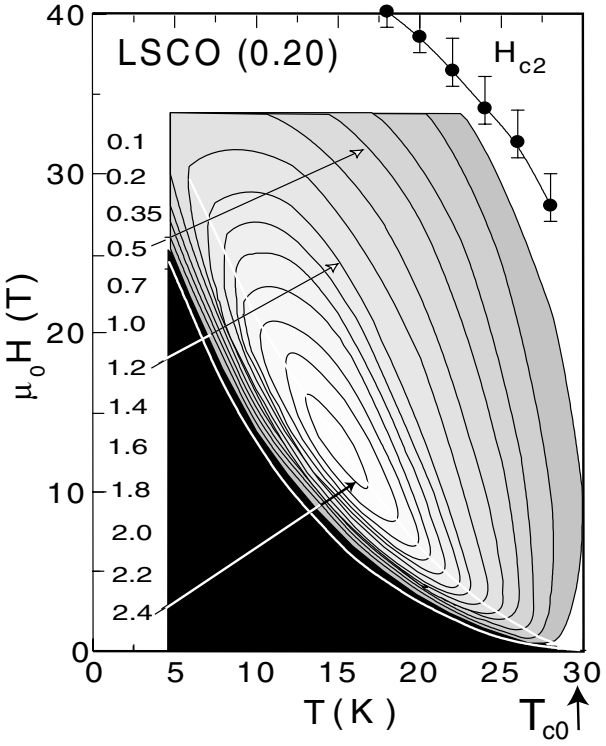

FIG. 2. The measured values of $e_y(T, H)$ in sample 1 (LSCO, $x = 0.20$) represented as a contour plot in the T-H plane. Values of e_y (in μV/K) at successive contour lines are shown on the left column (white arrows). The lower and upper white lines are the melting field H_m and "ridge" field H^*, respectively. $H_{c2}(T)$ values estimated from Fig. 1c are shown as solid circles. T_{c0} is indicated in the lower-right corner.

show that $H_{c2}(T)$ decreases with rising T, *but remains at extraordinarily high values* as $T \to T_{c0}$ (see Fig. 2). [We neglect a small correction from the negative Nernst coefficient $\nu_N \simeq -40$ nV/KT of the holes in overdoped LSCO (Fig. 3 of Ref. [7]). Accounting for the background increases s_ϕ and $H_{c2}(T)$ by ~10%.]

A powerful way to summarize the anomalous trends described is to represent $e_y(T, H)$ as a contour plot in the T-H plane (Fig. 2). In the vortex-solid phase below the melting line H_m (lower white curve), $e_y = 0$ because the vortices are pinned. As the vortex solid melts across H_m, e_y increases steeply up to the "ridge" which represents a field scale that we call $H^*(T)$ (upper white curve). To the right of the ridge, e_y falls monotonically. We focus on the two interesting regimes: the low-T limit and the region near T_{c0}.

As $T \to 0$, the two fields $H_m(T)$ and $H^*(T)$ increase rapidly towards values approaching $H_{c2}(0)$ [18]. In this large-H, low-T region, the contours are nearly horizontal, as required by the asymptotic high-field behavior (broken curve in Fig. 1a). This implies that the H_{c2} line is nearly horizontal below ~12 K, as anticipated above. At temperatures $T \leq T_{c0}$, the weak attenuation of e_y to high fields corresponds to the nearly vertical orientation of the contours. Clearly, there is no evidence for a putative H_{c2} line that terminates at T_{c0}. The values of $H_{c2}(T)$ derived from s_ϕ are plotted in Fig. 2 as solid circles.

The anomalous features of the Nernst signal become more pronounced when we go to the underdoped regime. A major change from the overdoped case is apparent in the field profiles of e_y. We show in Fig. 3 results in underdoped YBCO (sample 2, with $y = 6.50$ and $T_{c0} = 50$ K). As H increases above H_m, e_y rises rapidly, but attains a very broad maximum that extends undiminished to 30 T. In comparison with the curves in Fig. 1a, we see that e_y in sample 2 approaches zero only at fields considerably higher than 30 T. If we now convert e_y to s_ϕ using the measured ρ, the broad profile translates to a line entropy that extrapolates to zero at fields much higher than our maximum field. At 40 K, we may estimate the lower

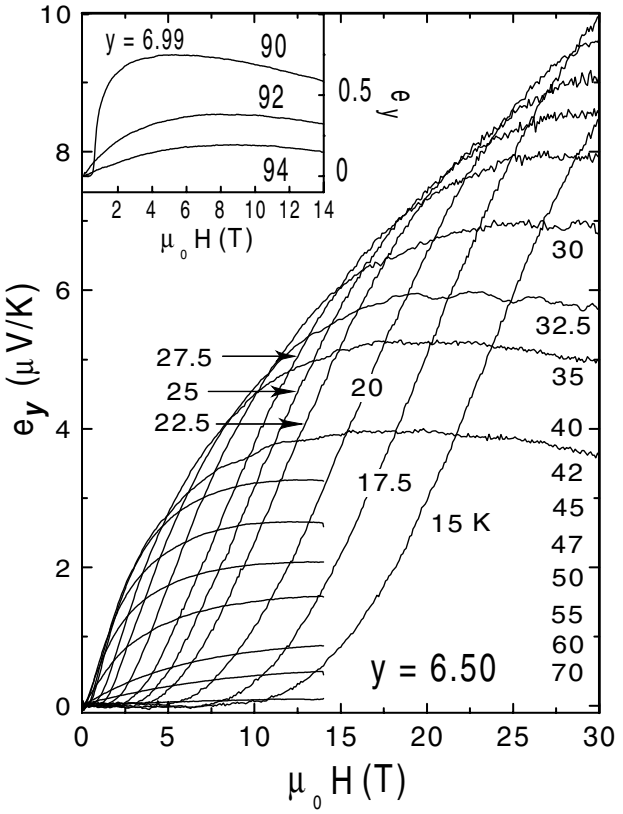

FIG. 3. Variation of the Nernst signal $e_y \equiv E_y/|\nabla T|$ with field H at fixed T in sample 2 (YBCO with $y = 6.50$, $T_{c0} = 50$ K). Above 40 K, the measurements were performed up to 14 T only. The inset shows e_y vs H in sample 3 (YBCO, $y = 6.99$) close to its $T_{c0} = 93$ K. Both are detwinned crystals grown in barium zirconate crucibles.

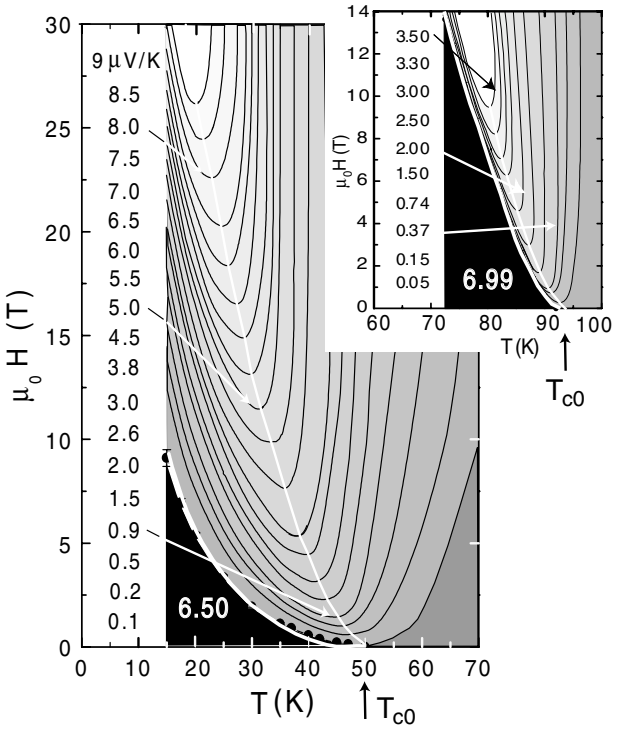

FIG. 4. (Main panel) Contour plot of e_y in the T-H plane in sample 2 (YBCO, $y = 6.50$). The upper (lower) white curve represents the "ridge" field H^* (melting field H_m). Inset shows the corresponding contour plot in sample 3 ($y = 6.99$). Note that the region between H_m and H^* is compressed to a thin sliver.

bound $H_{c2}(T) \geq 60$ T, which is very high for a temperature so close to $T_{c0} = 50$ K. These features are common to all the underdoped cuprates we have investigated until now (for results in LSCO and Bi 2201, see Ref. [8]).

The contour plot of e_y for sample 2 is displayed in the main panel of Fig. 4. In comparison with Fig. 2, the melting line is now considerably lower relative to H^*. Further, the contour lines in the region around T_{c0} are more spread out. The Nernst signal retains considerable strength at 70 K, indicating that vorticity strongly persists up to 20 K above T_{c0}. The contour plots show rather clearly the *continuity* between the high-temperature fluctuation phase observed by Xu *et al.* [7,8] and the vortex-liquid state below T_{c0}. The two regimes smoothly merge together, and no phase boundary or "crossover" line separating them is apparent in the T-H plane. As mentioned above, the upper critical field is estimated to be above 60 T even at 40 K. The much higher field scale in underdoped YBCO is also apparent if we compare the contour plots in Figs. 2 and 4. In overdoped LSCO, the contours close at fields above 13 T (the field scale of the "island"), whereas, in underdoped YBCO, they do not close until H exceeds 30 T. For comparison, we also display the contours in near-optimal YBCO (sample 3 with $y = 6.99$ and $T_{c0} = 93$ K). As shown by the vertical orientation of the contour lines from 85 to 95 K, as well as the field profiles in the inset in Fig. 3, e_y is virtually field independent up to 14 T. Even for near-optimal YBCO, there is no evidence for an H_{c2} line that decreases to zero at T_{c0} (this calls into question the interpretation of "H_{c2}" values inferred from bulk magnetization results [19]).

The observation of vortexlike excitations high above T_{c0} by Xu *et al.* [7,8] raised the question of whether the cuprate superconducting transition at T_{c0} actually corresponds to the loss of long-range phase rigidity, as opposed to the vanishing of the Gorkov pairing amplitude $\mathcal{F}(\mathbf{x}, \mathbf{x}')$ [20]. The present results provide strong support for the former. A prominent anomaly at temperatures near T_{c0} is the remarkably weak field dependence of e_y above the field scale $H^*(T)$, which implies that the condensate strength extends undiminished to very high fields. In underdoped cuprates, this anomalous behavior persists to our lowest temperatures (Fig. 3). However, in overdoped samples, the profiles sharpen up below 25 K to reveal pronounced field suppression at high fields (Fig. 1a). Values of $H_{c2}(T)$ inferred from the line-entropy profiles show that the reason for the anomaly is that the H_{c2} vs T line does not terminate at T_{c0}, but at a much higher temperature.

This seems to us to be compelling evidence that, at T_{c0}, the steep increase of ρ and the collapse of the Meissner effect correspond to a preemptive loss of phase rigidity (as has been advocated by Emery and Kivelson, and the early resonating valence bond theory [20]) rather than the vanishing of $\mathcal{F}(\mathbf{x}, x')$. This scenario is qualitatively distinct from that in bulk low-T_c superconductors. Although ρ saturates rapidly to ρ_N above T_{c0} (in zero field), there remains sufficient condensate density to support vorticity. In sample 1, the measured H_{c2} curve implies that we need to apply fields above 30 T to suppress all vorticity near T_{c0}. The field scale is even higher in underdoped YBCO.

A hallmark of the vortex-liquid state in cuprates is that ρ rapidly increases and "saturates" to the extrapolated normal-state value (ρ_N) even when $H \ll H_{c2}(T)$. To illustrate this, we plot the T dependence of ρ and e_y in sample 1 with H fixed at 33 T in Fig. 1d (inset). Clearly, the large Nernst signal indicates that the region from 5 to 25 K is in the vortex-liquid state (see Fig. 2). However, between 15 and 30 K, ρ virtually lies on the extrapolated ρ_N curve. Hence, reliance on ρ alone would lead to a serious underestimate of H_{c2}.

In all cuprates studied, the ridge field $H^*(T)$ line is ubiquitous. Over a broad range of T, the ratio ρ/ρ_N has the value 0.3–0.4 on the curve H^* vs T. $H^*(T)$ serves as the boundary separating the low-field regime in which ρ rapidly increases with H, from the high-field regime in which ρ slowly asymptotes to the extrapolated ρ_N. Hence, H^* represents an intrinsic field scale that controls dissipation in the vortex liquid state (attempts to find H_{c2} using ρ alone invariably turn up a curve akin to H^* rather than the higher H_{c2}). If we identify a length scale $\xi^*(T)$ by the ratio $\xi^*(T)/\xi_0(T) \sim \sqrt{H_{c2}(T)/H^*(T)}$, we find that, at low T, $\xi^*/\xi_0 \approx 1$. As T increases, however, the ratio rapidly increases (to 1.8 and 3.7 at 15 and 25 K, respectively). Since H^* determines the onset of strong flux-flow dissipation, ξ^* represents a radius larger than the vortex core radius ξ_0 ($\simeq 2.6$ nm). Packing vortices closer than the outer radius ξ^* leads to a crossover to the high-dissipation state in which ρ asymptotes to ρ_N. However, as superfluidity and its associated 2π phase winding survive right up to the inner radius ξ_0 [21], the Nernst signal is observable to the higher field scale H_{c2} (s_ϕ is strongly attenuated in magnitude because of the reduced supercurrent for $\xi^* < r < \xi_0$) [17]. A length scale extending outside the vortex core has been observed in tunneling experiments [22].

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