Unusual Nernst Effect Suggesting Time-Reversal Violation in the Striped Cuprate Superconductor La$_{2-x}$Ba$_x$CuO$_4$

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Unusual Nernst effect suggestive of time-reversal violation in the striped cuprate
La$_{2-x}$Ba$_x$CuO$_4$

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The striped cuprate La$_{2-x}$Ba$_x$CuO$_4$ ($x = \frac{1}{4}$) undergoes several transitions below the charge-ordering temperature $T_{co} = 54$ K. From Nernst experiments, we find that, below $T_{co}$, there exists a large, anomalous Nernst signal $e_{N, even}(H,T)$ that is symmetric in field $H$, and remains finite as $H \rightarrow 0$. The time-reversal violating signal suggests that, below $T_{co}$, vortices of one sign are spontaneously created to relieve interlayer phase frustration.

In the cuprates, there is increasing evidence that time-reversal invariance (TRI) is broken over a large portion of the phase diagram. Following a prediction in cuprates [1], signatures of TRI-violation were obtained in angle-resolved photoemission [2] and polarized neutron scattering experiments [3]. Recently, polar Kerr rotation measurements [4, 5] and polarized neutron scattering experiments [6] have discovered firmer evidence for TRI-violating states in several cuprates.

The cuprate La$_{2-x}$Ba$_x$CuO$_4$ at doping $x \approx \frac{1}{2}$ undergoes a remarkable series of electronic phase transitions starting at the charge-ordering temperature $T_{co}$ (54 K) and followed by the charge-ordering temperature $T_{co}$ (40 K) and the Berenzinski-Kosterlitz-Thouless (BKT) transition $T_{BKT}$ (16 K) [7–10]. Below 5 K, 3D superconductivity is established. We have observed an unusual zero-field Nernst effect signal that appears below $T_{co}$. In principle, such a zero-field Nernst signal is forbidden in a material that has TRI. We discuss the implications of its appearance below the charge ordering temperature $T_{co}$.

Nernst effect measurements were carried out on La$_{2-x}$Ba$_x$CuO$_4$ crystals with $x = \frac{1}{2}$ (LBCO-$\frac{1}{2}$). We cut crystals (2, 0.7, 0.2 mm$^3$ along the crystal axes a, b, c, respectively) from a boule and polished the faces until the normal to the broadest face was aligned with the applied field $H$.

In Fig. 1, we show the observed Nernst signal at selected $T$ from 160 K to 45 K (Panel a) and for $T \leq$35 K (Panel b). Above 35 K, $E_y$ is nominally linear in $H$ with a zero-field intercept that we identify with the zero-$H$ thermopower $S(0)$. The tilt of the curves is the conventional field-antisymmetric Nernst signal. Below the charge ordering at $T_{co} = 54$ K, however, $e_y(H,T)$ displays anomalous features which become prominent below 30 K (Panel b). The sharp, zero-field anomaly visible at 30 K grows steeply in the negative direction (relative to the zero-$H$ value at 35 K) as $T$ falls to 25 K. At 20 K, the anomaly assumes the shape of a narrow $H$-symmetric trench of full-width $\sim 2$ T. As $T$ decreases from 20 to 6 K, the trench width broadens rapidly to 15 T. At low $T$, we observe new structures appearing at lower fields.

Generally, the Nernst electric field $E_y$ is antisymmetric in $H$, vanishing at $H = 0$. Initially, we attributed the zero-$H$ signal in Fig. 1 to pickup of the longitudinal signal $S$. This assumption is valid above 60 K. However, below 54 K, a distinct field-even signal distinct from $S(H,T)$ becomes resolvable. To show this, we have measured the thermopower $S(H,T)$ simultaneously with the Nernst signal. Figure 2(a) displays the $T$ dependence of $e_y^{obs}$ and $S$ measured in zero field. We find that $S$ is positive above $T_{co} \sim 54$ K, decreases rapidly below 54 K, becoming negative below 45 K. At lower $T$, $S$ attains a broad minimum at 30 K before vanishing near $T_c = 5$ K.

First, we compare the zero-$H$ values of the observed Nernst signal $e_y^{obs}(0,T)$ (circles in Fig. 2a) and $S(0,T)$ (solid curve) over a broad interval of $T$. Above 54 K, the two quantities track closely. Multiplying the former by a scaling number $k$, we may superpose the two curves (Fig. 2a). The value of $k$ (-9.8) implies that the voltage contacts were slightly misaligned by $\sim 130 \mu m$ along $x$.

Below $T_{co}$, the two quantities deviate significantly. In contrast to the curve of $S$, $e_y^{obs}(0,T)$ oscillates vs. $T$, changing sign four times. With $k = -9.8$, we may isolate intrinsic Nernst signal $e_N(H,T)$ at finite $H$ by subtracting off the thermopower signal, viz.

$$e_N(H,T) = e_y^{obs}(H,T) - kS(H,T). \quad (1)$$

The quantity $e_N(0,T)$ in zero $H$, plotted in Fig. 2b, is of main interest. In the interval 30-54 K, the magnitude of $|e_N(0,T)|$ equals 0.2 $\mu V/K$, which is easily resolved in...
our experiment. Below 30 K, it rises steeply to a prominent maximum of 2.2 µV/K at 20 K before falling to zero near 5 K. The prominent peak, which is very sensitive to \( H \), is the cause of the trench feature bracketing \( H = 0 \) in the curves of \( e_y^{\mathrm{obs}} \) vs. \( H \) plotted in Fig. 1.

It is also instructive to examine the field-symmetrized form of the observed Nernst signal \( e_y^{\mathrm{obs}, \, \text{even}}(H) = \frac{1}{2}[e_y^{\mathrm{obs}}(H) + e_y^{\mathrm{obs}}(-H)] \) which admixes \( e_N, \text{even} \) and \( S \). At 20 K, \( e_y^{\mathrm{obs}, \, \text{even}}(H,T) \) displays a deep trench centered at \( H = 0 \) (Fig. 3a). As \( T \) decreases to 5 K, the trench broadens rapidly. For comparison, we also plot the curves of \( S(H,T) \) (scaled by the parameter \( k \)). The features in the field profiles are clearly distinct in the two sets of curves. This difference provides strong evidence that the Nernst signal \( e_N(H,T) \) has an intrinsic field-even component that is distinct from \( S(H,T) \).

Subtracting \( kS(H,T) \) from \( e_y^{\mathrm{obs}, \, \text{even}}(H) \) at each temperature, we isolate \( e_N, \text{even}(H,T) \), the field-even part of the intrinsic Nernst signal in Eq. 1. The curves of \( |e_N, \text{even}(H,T)| \) display broad peaks that shift to higher \( H \) as \( T \) decreases (Fig. 3b). The field at which the largest peak occurs is labelled \( H_1(T) \). A smaller shoulder at higher field is labelled \( H_2(T) \). At 20 K, the weight in \( e_N, \text{even}(H,T) \) is concentrated in a narrow trench (\( |H_1| \sim 0.5 \) T). As \( T \) is lowered, the two field scales \( H_1 \) and \( H_2 \) increase rapidly. They correlate with distinct features in the in-plane resistivity \( \rho_{ab} \) and the \( c \)-axis resistivity \( \rho_c \). Below 40 K, the derivatives \( d\rho_{ab}/dT \) and \( d\rho_c/dT \) show maxima at the fields \( H_{\rho_a}(T) \) and \( H_{\rho_c}(T) \), respectively [7]. In Fig. 4a, we compare the \( T \) dependences of \( H_1 \) and \( H_2 \) (solid symbols) with \( H_{\rho_a}(T) \) and \( H_{\rho_c}(T) \) (open symbols) (Panel (b) shows how \( H_1 \) and \( H_2 \) are defined). As shown, \( H_1 \) equals \( H_{\rho_a} \) within the resolution, while \( H_2 \) is roughly of the same scale as \( H_{\rho_c} \). Interestingly, \( H_1(T) \) follows the Debye-Waller (DW) form \( H_1 = H_0 \exp(-T/T_0) \), with \( T_0 \sim 6.9 \) K. The DW form
FIG. 3: (color online) Panel (a): Comparison of the raw, field-symmetrized, Nernst signal $e^{obs}_{y,even}(H,T)$ (solid curves) with the thermopower $S(H,T)$ (scaled by $k = -9.8$, dashed curves) at selected $T \leq 20$ K. Note that $S(H,T)$ is actually negative below 40 K (at all $H$ shown). The two sets of curves have very different field dependences. Panel (b) displays the curves of the intrinsic field-symmetrized Nernst signal $e_{N,even}(H,T)$ obtained by subtracting the two sets of curves (see Eq. 1). The oscillatory features are absent in $S(H,T)$. At large $H$, $e_{N,even}(H,T)$ is suppressed to zero.

implies that thermally induced changes to the vortex system lead to prominent features in the anomalous Nernst signal $e_N(0,T)$. In underdoped La$_{2-x}$Sr$_x$CuO$_4$, the DW form describes the melting field of the vortex solid (with comparable $T_0$) [14]. We also note that the curves of $S$ vs. $H$ (dashed curves in Fig. 3a) display step-like increases when $H$ exceeds $H_1 \sim H_{p,a}$, that match the abrupt increase in $\rho_a$. This pattern suggests that the collapse of the anomalous Nernst signal at $H_1$ leads to an increase in dissipation and entropy flow. We return to this point below.

We field-antisymmetrize the Nernst curves in Fig. 1 to obtain the conventional Nernst signal $e_{N,odd}(H) = \frac{1}{2}[e_y^{obs}(H) - e_y^{obs}(-H)]$. The Nernst coefficient, $\nu = e_{N,odd}/H$ ($H \rightarrow 0$), provides a useful comparison between field-induced vortices and the spontaneous vortices. At high $T$ (120-180 K), $\nu$ is negative, reflecting the quasiparticle contribution to the Nernst signal (dashed line in Fig. 4b). At the onset temperature $T_{onset} \sim 110$ K, $\nu$ deviates from the dashed line and increases rapidly, as observed in La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) [11]. The deviation correlates with an unusual downward deviation in the torque susceptibility $\Delta \chi = \chi_c - \chi_a$ in the torque signal (solid triangles), where $\chi_c$ ($\chi_a$) is the susceptibility with $H||c$ ($||a$). Above $T_{co}$, $\chi_c$ is $\sim 10 \chi_a$ [8], so $\Delta \chi$ is dominated by $\chi_c$. Hence the downward deviation confirms the onset of diamagnetic susceptibility in $\chi_c$ reported in Ref. [8]. (Below $T_{co}$, $\Delta \chi$ is complicated by a large local moment response in both $\chi_c$ and $\chi_a$.) The magnetization results verify that, for $T > T_{co}$, the increase in $\nu$ arises from vortex fluctuations (and not from quasiparticles, as conjectured [15]). A similar agreement between Nernst and torque experiments was obtained for LSCO [11, 12]. At $T_{co}$, the increase in $\nu$ is abruptly interrupted. Below 20 K, however, $\nu$ resumes its steep increase as the condensate establishes long-range phase coherence.

The conventional field-antisymmetric Nernst signal shown in Fig. 4b is generated by vortices introduced
by an external $H$. By contrast, we associate $e_{N,\text{even}}$ with vortices that are present in equilibrium at $H = 0$, as in a 2D superconductor above $T_{BKT}$. However, unlike the BKT problem (in which the net vorticity is zero in $H=0$), here we must have predominantly “up” vortices to produce a finite $e_N(0,T)$. Using torque magnetometry, we have measured the irreversibility field $H_{\text{irr}}$ in the same crystal. As shown in Fig. 2b, $H_{\text{irr}}$ has a very different profile from $e_N(0,T)$; $H_{\text{irr}} \rightarrow 0$ near 20 K, where $\{e_N(0,T)\}$ attains a maximum. Thus $e_N(0,T)$ is not caused by field-induced vortices trapped in a non-equilibrium state. (In the interval $5 < T < 20$ K, the pair condensate rigidity is strongly inhomogeneous. The vortex solid exists in isolated regions detectable by magnetization hysteresis. These regions do not contribute to the observed $e_N$ or $S$.)

The results in Refs. [7, 9] have shown that pronounced superconducting fluctuations extend from $T_{co}$ down to 5 K. The extreme anisotropy of this response indicates that the Josephson coupling between adjacent layers is highly frustrated. To explain this frustration, it has been proposed that pair-density-wave (PDW) superconductivity develops along with the stripe order [17–19]. Because the stripe modulation direction is orthogonal between adjacent layers, Josephson coupling cancels out. The abrupt interruption of the increasing trend in $\nu$ at $T_{co}$ (Fig. 4b) is consistent with a sharp change in the character of the probed phase coherence. Below $T_{so} = 40$ K, previous results [7, 9, 20, 21], imply that competition between the PDW and uniform $d$-wave superconductivity exists. Eventually, at $\sim 5$ K, the latter dominates and true 3D long-range phase coherence prevails. The steep rise of $\nu$ below 20 K is consistent with the eventual development of uniform $d$-wave order.

In the PDW state, small fluctuations in the Josephson phase $\theta(r)$ about the uniform-phase state can lead to a gain in free energy [19]. The present results suggest to us that, below $T_{co}$, the sample spontaneously nucleates an array of 2D vortices in $H=0$, which can provide a large phase-slip of $2\pi$. Having all the vortices be of the same sign (which breaks TRI) entails a cost in the kinetic energy of the supercurrent. However, because the local supercurrent is weak, the cost may be offset by a large gain in condensate energy provided by significant reductions in the interlayer phase frustration. Because $\theta$ is strongly fluctuating, we expect the vortices to flow freely in a gradient $-\nabla T$ and to generate a spontaneous Nernst signal.

The anomalous Nernst signal $e_N(0,T)$ attains its largest amplitude at 20 K close to $T_{BKT}$ (16 K). Below $T_{BKT}$, the small but finite $\rho_{ab}$ implies that phase rigidity extends in the $a$-$b$ plane over sizeable lengths at $H=0$ [7]. However, when $H$ exceeds $H_{\text{so}}$, the collapse of the rigidity produces an increase in $\rho_{ab}$. As mentioned, this coincides with a steep increase in $S$ which measures entropy flow (Fig. 3a), as well as the collapse of $e_{N,\text{even}}$ above $H_1$ (Fig. 3b). This suggests to us that the spontaneous vortices, when present, help to establish a phase-coherent state that has low dissipation and low entropy. At the larger field $H_{pc}$, the step increase in $\rho_\phi$ signals the loss of interlayer coherence. This is also reflected in $e_{N,\text{even}}$ as $H_2$, but as a much weaker feature.

Despite the spontaneous nature of the time-reversal violation, some external influence must nudge the system into selecting one direction in a given experiment. We tried to change the sign by warming the sample to 290 K and then cooling in a different superconducting magnet, but it remained the same. We also tried field-cooling in $H = 14$ T from 290 K, and also swept the field between $+14$ and $-14$ T both above and below $T_{co}$ but could not alter the sign. A. Kapitulnik has suggested to us that a weak magnetic ordering may onset at 360 K. Field-cooling from above 360 K may pre-select the sign; this is left for a future investigation.

Recently, we learned of polar-Kerr rotation TRI violating results in LBCO-1/8 [22]. The Kerr angle $\theta_K$ in $H = 0$ is unresolved from zero above $T_{co}$, but increases abruptly at $T_{co}$, reaching a sharp maximum at 41 K.

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