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$_4$ ($x = \frac{1}{8}$) 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.

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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 uncovered firmer evidence for TRI-violating states in several cuprates.

The cuprate La$_{2-x}$Ba$_x$CuO$_4$ at doping $x \approx \frac{1}{8}$ undergoes a remarkable series of electronic phase transitions starting at the charge-ordering temperature $T_{co}$ (54 K) and followed by the spin-ordering temperature $T_s$ (40 K) and the Berenzinski-Kosterlitz-Thouless (BKT) transition $T_{BKT}$ (16 K) [7,8,9]. 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 $x = \frac{1}{8}$ (LBCO-$\frac{1}{8}$). 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 $c$ to within $\pm 0.5^\circ$. For each curve of the Nernst signal vs. the applied field $H$, we made dual measurements at two temperature gradients ($-\nabla T = 0.5$ K/mm and 0.7 K/mm) to check for linearity and reproducibility. The field was swept slowly at rates 0.2 T/min to 0.5 T/min. The measured thermal conductivity $\kappa$ has a relatively weak $T$ dependence between 10 and 60 K (varying between 6 and 7.2 W/Km). In our geometry, $-\nabla T$ is applied along $y$ in the LTT phase (with axes $a$, $b$, $c$). With $H||z$, the voltage $V_y$ observed along $y$||$b$ gives the observed Nernst signal, $e_{N,obs}(H,T) \equiv V_y(H,T)/(|\nabla T||d)$ with $d$ the voltage-contact spacing (we use little $“e”$ to denote the Nernst electric-field $E_y$ divided by $|\nabla T|$). In Nernst experiments, $e_{N,obs}$ is often contaminated by unavoidable pickup of the longitudinal thermopower signal caused by slight lead misalignment. We show that the anomalous signal is distinct from this pickup.

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 $e_{N,obs}$ 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 $e_{N,obs}(0,T)$ (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 $S$, $e_{N,obs}(0,T)$ 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.

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

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 $\mu$V/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 $e_y^{\text{obs}}$ vs. $H$ plotted in Fig. 1.

It is also instructive to examine the field-symmetrized form of the observed Nernst signal $e_y^{\text{obs}}(H) = \frac{1}{2}[e_y^{\text{obs}}(H) + e_y^{\text{obs}}(-H)]$ which admixes $e_{N, \text{even}}$ and $S$. At 20 K, $e_y^{\text{obs}}(H, T)$ displays a deep trench centered at $H = 0$ (Fig. 2a). At $T$ 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 $e_y^{\text{obs}}(H)$ 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. 2b). 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 $H_{\rho_{ab}}(T)$ and $H_{\rho_c}(T)$, respectively [2]. In Fig. 3, we compare the $T$ dependences of $H_1$ and $H_2$ (solid symbols) with $H_{\rho_{ab}}(T)$ and $H_{\rho_c}(T)$ (open symbols) (Panel b shows how $H_1$ and $H_2$ are defined). As shown, $H_1$ equals $H_{\rho_{ab}}$ 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...
FIG. 3: (color online) Panel (a): Comparison of the raw, field-symmetrized, Nernst signal $e_{\text{N,even}}^{\text{obs}}(H, T)$ (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 $e_{\text{N,even}}(H, T)$ obtained by subtracting the two sets of curves (see Eq. 1). The oscillatory features are absent in $S(H, T)$. At large $H$, $e_{\text{N,even}}(H, T)$ is suppressed to zero.

$H_1 = H_0 \exp(-T/T_0)$, with $T_0 \sim 6.9$ K. The DW form 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 $H_1 \sim H_{\rho a}$, 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 $e_{N,\text{odd}}(H) = \frac{1}{2}[e_{y}^{\text{obs}}(H) - e_{y}^{\text{obs}}(-H)]$. The Nernst coefficient, $\nu = e_{N,\text{odd}}(H) / (H \to 0)$, 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. 1a). At the onset temperature $T_{\text{onset}} \sim 110$ 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 $T_{\text{co}}$, $\chi_c$ 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 $T_{\text{co}}$, $\Delta \chi$ is complicated by a large local moment response in both $\chi_c$ and $\chi_a$.) The magnetization results verify that, for $T > T_{\text{co}}$, 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 $T_{\text{co}}$, 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 \( H_{irr} \) in the same crystal. As shown in Fig. 2, \( H_{irr} \) has a very different profile from \( e_N(0,T) \); \( H_{irr} \to 0 \) 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 \( S_e \)).

The results in Refs. [7, 8] 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 \( T_{so} \) (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 \( T_{so} \), 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 \( H_{pa} \), 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 \( H_{pa} \), the step increase in \( \rho_{ab} \) 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-\( \frac{1}{4} \) [22]. The Kerr angle \( \theta_K \) at \( H = 0 \) is unresolved from zero above \( T_{co} \), but increases abruptly at \( T_{co} \), reaching a sharp maximum at 41 K.

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