Entropy of Vortex Cores Near the Superconductor-Insulator Transition in an Underdoped Cuprate

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We present a study of Nernst effect in underdoped $La_{2-x}Sr_xCuO_4$ in magnetic fields as high as 28 T. At high fields, a sizable Nernst signal was found to persist in the presence of a field-induced nonmetallic resistivity. By simultaneously measuring resistivity and the Nernst coefficient, we extract the entropy of vortex cores in the vicinity of this field-induced superconductor-insulator transition. Moreover, the temperature dependence of the thermoelectric Hall angle provides strong constraints on the possible origins of the finite Nernst signal above T_c, as recently discovered by Xu *et al.* [Nature (London) **406**, 486 (2000)].

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An intriguing case of vicinity between superconducting and insulating ground states occurs in the underdoped cuprates [1]. Various investigations have shown that reducing the density of charge carriers [2], introducing disorder [3], or applying a magnetic field [1] leads to the replacement of a superconductor with an insulator. The latter route (i.e., the field-driven superconductor-insulator transition) raises many unanswered questions, including the possible existence of vortices with insulating cores. The structure of the vortex core in a doped Mott insulator has been a subject of numerous theoretical studies [4–6]. On the experimental side, recent neutron scattering experiments on $La_{2-x}Sr_xCuO_4$ (LSCO) have revealed the existence of a dynamic magnetic order associated with the vortex state which evolves towards a static order in the underdoped regime [7]. Meanwhile, scanning tunneling microscopy (STM) has proved to be a direct source of information on the electronic spectrum inside vortices [8,9]. Until now, however, experimental exploration of field-induced superconductor-to-insulator transition has been limited to resistivity measurements [1,10].

In this Letter, we report on the evolution of the Nernst coefficient in underdoped LSCO in the vicinity of the superconductor-insulator transition. Nernst effect, the generation of a transverse electric field by a longitudinal thermal gradient in a magnetic field, has been an instructive probe of vortex movement in the mixed state of high-T_c superconductors in the early 1990s [11]. Recently, Xu *et al.* [12] reported the existence of a sizable Nernst signal above T_c over a broad temperature range in underdoped LSCO. They argued that the quasiparticle contribution to the Nernst signal should be negligible and interpreted their finding as evidence for the existence of vortexlike excitations above T_c [12]. Our study, concentrated on the Nernst coefficient at high magnetic field, leads to several new findings. First, a large Nernst signal was found to persist in the presence of a field-induced nonmetallic behavior in resistivity. This observation provides new support for the concept of vortices with insulating cores. Moreover, using both Nernst and resistivity data, we calculate the entropy associated with the vortex cores. Finally, we argue that the temperature dependence of the thermoelectric Hall angle puts strong constraints on the origin of the residual Nernst signal above T_c [12].

The preparation and characterization of LSCO single crystals are described in detail elsewhere [13]. Using miniature RuO_2 thermometers, our setup was designed in a way to measure the Seebeck, Nernst, Hall, and resistivity coefficients using the same contacts.

Figure 1 shows the temperature dependence of the Nernst signal and resistivity in a $La_{1.92}Sr_{0.08}CuO_4$ single crystal for various magnetic fields. In the presence of a magnetic field of 12 T, resistivity shows a broad transition ending at $T \sim 5$ K. At the same field, we detect a large Nernst signal which peaks at $T \sim 18$ K. The broad resistive transition is a consequence of dissipation due to the vortex movement. Thus, a concomitant Nernst signal due to the effect of a thermal force on the same vortices is naturally expected and is in agreement with the results on optimally doped cuprates [11]. But the evolution of the Nernst signal at higher fields is surprising. As seen in the upper panel of Fig. 1, a magnetic field of 26 T is large enough to induce a slight nonmetallic behavior in resistivity in the 15–40 K temperature range. However, at this field, the maximum in the Nernst signal occurs at almost the same temperature, broadens, and presents

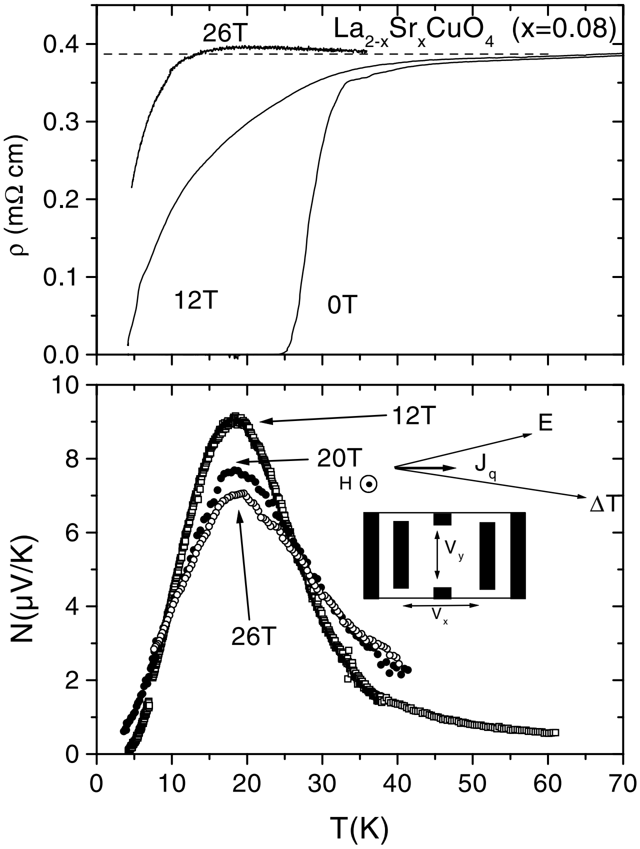

FIG. 1. Resistivity and Nernst effect as a function of temperature in an underdoped LSCO crystal for different magnetic fields. The broken horizontal line represents 0.39 $m\Omega$ cm. Inset shows the contact geometry on the sample and the relevant vectors.

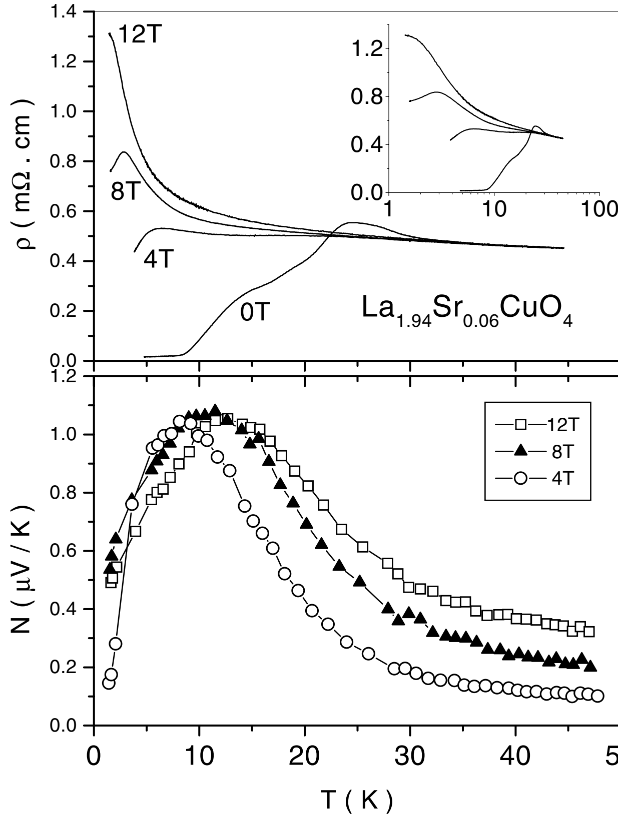

FIG. 2. Resistivity and Nernst effect as a function of temperature in another underdoped LSCO crystal for different magnetic fields. Note the coexistence of a Nernst signal and an apparently insulating behavior at $H = 12$ T. The inset shows the resistivity data in a log-linear scale.

a reduced but still large magnitude. The coexistence of the peak in the Nernst signal and a nonmetallic resistivity is in sharp contrast with what has been reported for all superconductors including optimally doped cuprates [11]. This result is the main new finding of this Letter and was confirmed on two other single crystals at lower doping levels. Figure 2 shows the data on a $La_{1.94}Sr_{0.06}CuO_4$ single crystal. As seen in the figure, the sample shows a very broad resistive transition at zero field. The application of a magnetic field leads to the emergence of an insulating behavior. As seen in the inset, with increasing magnetic field, the resistivity gradually evolves towards a logarithmic temperature dependence as first reported in 60 T studies [1]. However, the magnetic field barely affects the peak in the Nernst signal which, nevertheless, presents a reduced magnitude compared to the $x = 0.08$ case. To reconcile the Nernst and resistivity data, it is tempting to assume that at high field, the system is populated by vortices which can produce a nonmetallic resistivity. This may arise in the context of an insulating normal state, since the flux-flow resistivity ρ_F is a fraction of the normal state resistivity ρ_N [14]. Thus, a nonmetallic $\rho_F(T)$ may reflect the insulating behavior of $\rho_N(T)$ with attenuation.

More insight on the fuzzy phase boundary between the superconducting and the normal states may be achieved by comparing the field dependence of Nernst effect and resistivity of the $x = 0.08$ sample. As seen in the upper panel of Fig. 3, below T_c, the Nernst coefficient is not a linear function of magnetic field. The thermal force on each vortex is proportional to the excess of entropy associated with it. Since the latter would become zero at H_{c2}, the Nernst signal is expected to decrease at a finite field in spite of the increase in the number of vortices. As seen in the lower panel of Fig. 3, the nonvanishing Nernst signal is concomitant with a large magnetoresistance up to the highest explored magnetic fields (28 T). Figure 3 also displays the passage between metallic and nonmetallic behaviors at H $\sim$ 27 T. It is worth noting that the magnitude of resistivity at the boundary between metallicity and nonmetallicity (0.39 mΩ cm) yields a resistance close to $\frac{h}{4e^2}$ per CuO_2 plane, reported as the critical threshold resistance for the superconductor-to-insulator transition in cuprates [2].

Combining the Nernst and resistivity data, one can calculate the entropy associated with these vortices [11]. When vortices move under the influence of a thermal force, $f_{th} = -\frac{\partial T}{\partial x} S_\phi$ ($S_\phi$is the transport entropy per unit length of an individual vortex), they produce a transverse electric field according to the Josephson equation, $E_y = B_z v_x$, where v_x is the average vortex velocity. This velocity is proportional to the force applied on a vortex

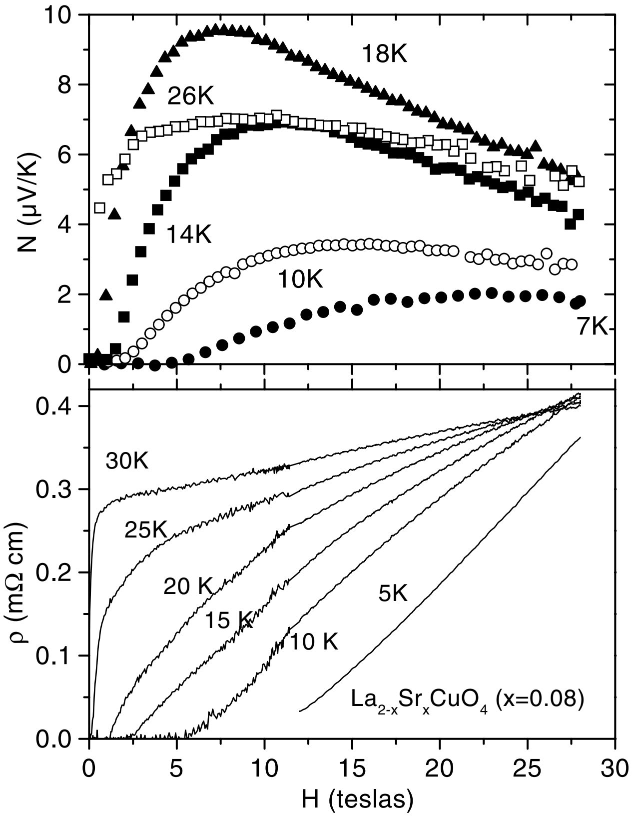

FIG. 3. Resistivity and Nernst effect as a function of field in an underdoped LSCO crystal for different temperatures.

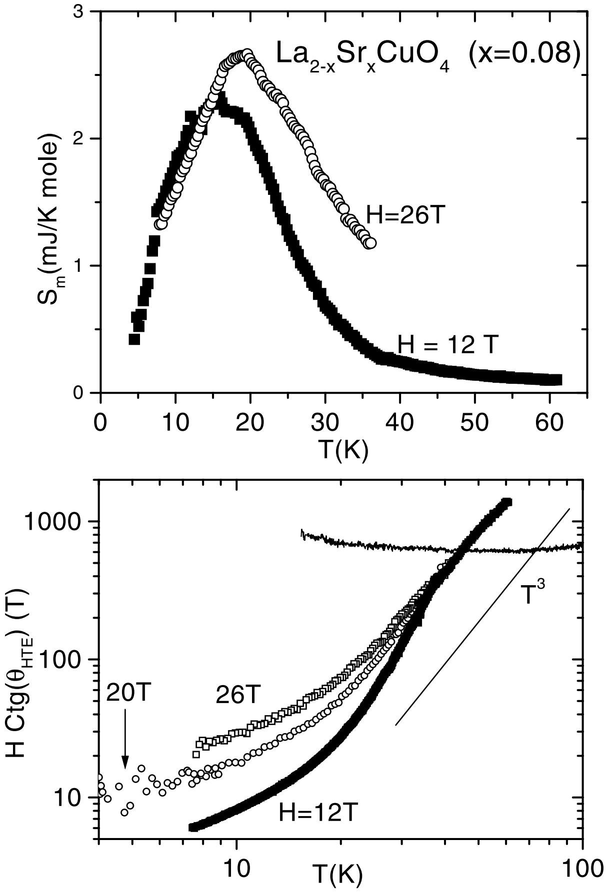

FIG. 4. Upper panel: Entropy carried by vortices as extracted from resistivity and Nernst coefficient for the $x = 0.08$ sample at two different magnetic fields (see text). Lower panel: The temperature dependence of the normalized thermoelectric Hall angle at different magnetic fields. The solid line represents the normalized electric Hall angle (measured at 12 T) for the same sample.

$v_x = \eta f_{th}$ with η being a viscosity coefficient. On the other hand, flux movement in the presence of a Lorentz force on individual vortices, $f_L = J_x \Phi_0$, is at the origin of the longitudinal electric field produced by an electric current, $E_x = B_z v_y$. The same viscosity coefficient relates this velocity to the Lorentz force $v_y = \eta f_L$. Thus

$$\frac{J_x \Phi_0 B_z}{E_x} = \frac{\frac{\partial T}{\partial x} S_\phi B_z}{E_y}. \tag{1}$$

And defining resistivity as $\rho_F = \frac{E_x}{J_x}$ and the Nernst coefficient as $N = \frac{E_y}{\frac{\partial T}{\partial x}}$, one obtains [11]

$$S_\phi = \frac{N \Phi_0}{\rho_F}. \tag{2}$$

Now, the volume entropy at a given magnetic field is obtained by multiplying S_ϕ by $\frac{H}{\Phi_0}$, the density of vortices at a given field H. Hence,

$$S_m = \frac{NH}{\rho_F V_m} \tag{3}$$

is the excess entropy carried by vortices at a given field in molar units. Here V_m is the molar volume (9.5×10^{-29} m^3/mol for LSCO [15]). Figure 4 displays the temperature dependence of S_m obtained in this way for $H =$ 12 T and $H = 26$ T. In the picture sketched above, this plot represents the entropy accumulated inside the vortices.

As seen in the figure for both fields, S_m shows a maximum and remains finite well above T_c ($= 27$ K) in the "normal" state. This is a consequence of the finite value of Nernst coefficient above T_c and up to the highest temperature explored in this study (~63 K; see data in the lower panel of Fig. 1). This latter observation, first reported by Xu *et al.* [12] was interpreted by these authors as evidence for vortexlike excitations in the pseudogap regime. So, the analysis sketched above leads to the existence of substantial S_m persisting up to T^* [12] (the temperature below which the pseudogap opens up). Note that this simple analysis remains valid for any exotic electronic excitation which happens to be a reservoir of entropy (in order to move by a thermal gradient) and either a topological defect in a phase-coherent environment or a fluxoid (in order to produce an electric field by its movement). Now, while below T_c ($H = 0$), the resistivity is entirely generated by the vortex movement and there is no ambiguity about the magnitude of ρ_F; this is not the case in the normal state. Indeed, at this stage, in the absence of any solid evidence for vortex dissipation in charge transport, the magnitude of ρ_F above T_c is a matter of speculation. On the other hand, our estimation of S_m in the superconducting state is straightforward and does not suffer from the current uncertainty on the origin of the residual Nernst signal in the pseudogap regime.

It is interesting to compare the temperature dependence and the magnitude of S_m with the results of an extensive study of specific heat in cuprates by Loram *et al.* [16]. Besides its finite value above T_c, the overall temperature dependence of $S_m(T)$ is reminiscent of the difference between the entropy of the superconducting state and the extrapolated entropy of the normal state obtained by specific heat measurements. However, the magnitude of S_m is almost an order of magnitude smaller than the maximum in the difference in the specific heat data for LSCO at this doping level [16]. The discrepancy is probably due to the important differences between the nature of information obtained by these two probes. First of all, our results are obtained in the presence of a strong magnetic field which is known to diminish and broaden the electronic specific heat jump and consequently reduce the entropy difference between the two states [17]. In the second place, the transport entropy of a vortex is yet to be theoretically clarified in the context of d-wave superconductivity. To the first approximation, the electronic excitation spectrum of a vortex core reflects that of the normal state. There is evidence suggesting that this picture is too naive for the high-T_c cuprates. Notably, STM studies [8,9] have reported a remarkably reduced difference in the low-energy density of states inside and far away from vortex cores compared to what has been observed in a conventional superconductor [18].

Finally, let us consider possible alternative origins of the observed Nernst signal above T_c. Since vortex movement is not the only source of a Nernst signal, it is useful to underline the restrictions that a quasiparticle scenario should face in order to account for such a signal in our context. For this purpose, we used our setup to measure the ratio of the longitudinal to transverse electric fields produced by a fixed thermal current, $\vec{J}_q$, along the sample. This ratio directly determines the field-induced rotation of the electric field produced by a thermal current. The cotangent of the thermoelectric Hall angle, $\cot\theta_{HTE}$, obtained in this way may be compared with the electric Hall angle, $\cot\theta_H$. The lower panel of Fig. 4 compares the evolution of the two angles. As seen in the figure, the thermoelectric Hall angle presents a T^3 behavior in the normal state and becomes even stronger (and field dependent) below 40 K. This reflects a rapid increase in the Nernst signal below T^* and its nonlinearity below 40 K. In the same temperature region, the measured electric Hall angle is almost temperature independent. Now, $\overrightarrow{J_q} = \overline{\alpha}\overrightarrow{E} + \overline{\kappa}\overrightarrow{\nabla}T$ and, since there is no charge current, $\overline{\sigma}\overrightarrow{E} = \frac{\overline{\alpha}}{T}\overrightarrow{\nabla}T$, which yields $\overrightarrow{J_q} = (\overline{\alpha} + \overline{\kappa}\,\overline{\alpha}^{-1}\overline{\sigma}T)\overrightarrow{E}$ (see the inset of Fig. 1). Therefore the angle between $\overrightarrow{J_q}$ and $\overrightarrow{E}$ reflects the successive rotations produced by $\overline{\alpha}$, $\overline{\kappa}$, and $\overline{\sigma}$ which are the thermoelectric, thermal, and electric conductivity tensors. Explaining the rapid temperature dependence of $\cot\theta_{HTE}$ seems to be a major challenge for the standard transport theory. Indeed, it has already been noted that in absence of an energy dependence in the scattering time, $\tau(\varepsilon)$, and even for a highly anisotropic single Fermi surface, the two ratios $\frac{\alpha_{xx}}{\alpha_{xy}}$ and $\frac{\sigma_{xx}}{\sigma_{xy}}$ are expected to be equal [19] which implies an identical rotation due to $\overline{\alpha}$ and $\overline{\sigma}$. More generally, in a Boltzmann picture [20],

$$\overline{\alpha} = \frac{(\pi k_B T)^2}{3} \frac{\partial \overline{\sigma}}{\partial \epsilon}\bigg|_{\varepsilon=\varepsilon_F}, \qquad (4)$$

which establishes an intimate relationship between the two angles even in the case of a highly energy-dependent $\tau(\varepsilon)$. Any alternative scenario on the origin of the finite Nernst signal above T_c implying quasiparticles instead of vortices is expected to explain the contrasting behavior of the two angles. We note that Eq. (4) is valid only when charge and entropy are carried by the same electronic excitations which is not the case in the charge-spin separation scenarios.

In summary, we found that the Nernst signal in underdoped LSCO persists in the presence of a magnetically induced nonmetallic behavior. We extracted the entropy carried by vortices in the vicinity of this superconductor-to-insulator transition and measured the temperature dependence of the thermoelectric Hall angle.

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