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Normal-state Nernst effect in a $Tl_2Ba_2CaCu_2O_{8+\delta}$ epitaxial film

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We report a measurement of the Nernst effect in the normal state of a cuprate superconductor, $Tl_2Ba_2CaCu_2O_{8+\delta}$, over the temperature range from T_c to above room temperature. The signal is small: the Nernst coefficient, Q, is never larger than 3 nV/K T over the entire temperature range. The negative sign of Q suggests that the cyclotron scattering rate, $1/\tau_H$, increases with particle energy in $Tl_2Ba_2CaCu_2O_{8+\delta}$.

One of the most tantalizing features of the high-T_c cuprate superconductors is the strange nature of cyclotron motion in the normal state. Recently, following Anderson[1] and Chien, Wang, and Ong,[2] several groups[3–17] have shown that the temperature dependence of the Hall coefficient, R_H, may be understood as the ratio of two independent relaxation times for cyclotron and longitudinal motion, respectively. Reparametrizing the Hall and resistivity (ρ) data in terms of the cotangent of the Hall angle, $\cot\theta_H=\rho/R_H H$, isolates the cyclotron relaxation rate, $1/\tau_H$, which is found to obey a simple Matthiessen-rule behavior as impurity scattering is introduced. This apparently simple phenomenology fits Hall and resistivity data over a striking range of samples, temperature, carrier density, and impurity density. In nearly all cases the intrinsic cyclotron relaxation rate, $1/\tau_H$, is proportional to the square of the absolute temperature, T. Since the resistivity evolves in a more complicated fashion as the carrier density increases,[18,19] the cyclotron relaxation rate may turn out to be more fundamental than the longitudinal rate, $1/\tau_{tr}$.

The central mystery now is the origin of the ubiquitous T^2 relaxation rate. Studies of the Hall coefficient have been very successful in demonstrating the T^2 law, but as yet provide no indication to the underlying mechanism. For further investigation, additional probes of normal-state cyclotron motion are desirable. The Nernst effect is perhaps the simplest conceptual alternative to the Hall effect—instead of an electric current, a thermal gradient is applied across the sample. The Nernst coefficient, Q, is the ratio of the transverse electric field component to the thermal gradient and magnetic field:

$$Q=\frac{E_y}{H_z\left(\frac{\partial T}{\partial x}\right)}. \quad (1)$$

In the mixed state below T_c, the Nernst effect is large, because motion of vortices down the thermal gradient induces a sizable transverse voltage. The field and temperature dependences of the Nernst effect in the cuprates below T_c have been well studied by several groups[20–26] who have reported excellent agreement with other mixed-state transport properties. We have undertaken to measure the normal-state Nernst effect and report here our results, on a sample of $Tl_2Ba_2CaCu_2O_{8+\delta}$. In our measurement, the thermal gradient, $\partial T/\partial x$ and the Nernst electric field, E_y, were in the a-b plane, and the magnetic field was along the c axis of the material.

We are not aware of any previously reported measurements of the Nernst effect in the cuprate materials over the temperature range from T_c to room temperature, most likely because the experiment is quite difficult. In order to appreciate the difficulty, consider first a measurement of the Hall effect. The resistance signal is larger than the Hall signal by a factor $1/\theta_H$, which is on the order of 10^3 in a 10 T field. Even minute contact misalignment produces resistive voltages which can be orders of magnitude larger than the Hall signal. Since the dominant signal, the resistive offset, is temperature dependent, very high-temperature stability is required during field sweeps. Typically, maintaining temperature fluctuations, ΔT, less than 50 mK is sufficient to resolve the Hall signal, although more accurate control may be necessary for careful study of the Matthiessen-rule behavior of $\cot\theta_H$.

Measurements of the Nernst effect face several new complications not encountered in the Hall effect. The Nernst signal is much smaller than the Hall signal. We expect that the Nernst coefficient, Q, has the order of magnitude, $S\theta_H$, where S is the Seebeck coefficient, typically on the order of 1 μV/K in the optimally doped cuprates. In other words, the Nernst signal is on the scale of nanovolts.

A greatly enhanced noise background further hinders resolution of the small Nernst signal. In the Hall measurement, the resistive signal due to small, but unavoidable, contact misalignment is at least constant at fixed temperature. In the Nernst geometry, the undesired offset signal is a thermal emf from temperature differences between the Nernst voltage contacts. But in any real measurement temperature fluctuates, giving rise to a *fluctuating* background signal, which is, in principle, much larger than the Nernst signal we wish to measure.

In the Hall measurement, the resistive offset can, in principle, be reduced by using a five-probe geometry with external compensation circuitry. Alternatively, a thin-film sample can be lithographically etched into a microbridge geometry which eliminates contact misalignment entirely. Neither approach is possible in the Nernst measurement. Compensation circuitry would introduce spurious thermal emfs far larger than the Nernst signal. Patterning a thin film does not help to reduce thermal offsets in the Nernst measurement because the temperature gradient exists in the substrate.

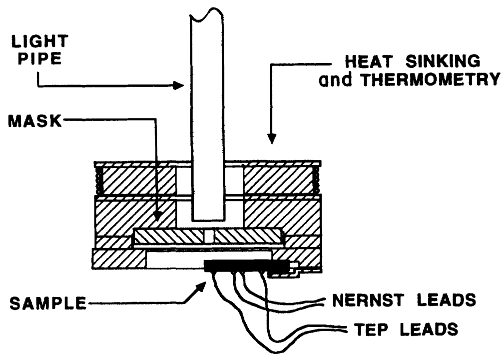

FIG. 1. Simplified drawing of the sample-holding assembly.

Resolution of the nanovolt-scale Nernst signal in a fluctuating background requires an ac measurement capability. In order to have fast thermal response times, thin film samples and single crystals are advantageous. In particular, thin film samples are ideal because the presence of the substrate eliminates contributions to the Nernst voltage from the Righi-Leduc effect.

The Nernst effect measurement needs a spatially homogeneous temperature gradient. Unlike the thermopower measurement which depends only on the total integrated temperature difference between the voltage contacts, the Nernst measurement is sensitive to the *local* temperature gradient, $\partial T/\partial x$. An inhomogeneous gradient would introduce unknown systematic factors into the measurement.

Finally, an important constraint on the Nernst experimental method is the problem of temperature stability. The Nernst effect is a thermally forced measurement, but has the directly competing constraint of high-temperature stability, as in the Hall effect, because θ_H is so small in the normal state. One way to reconcile the competing constraints is to have accurate control of the sample base temperature in the absence of the applied temperature gradient, and to have precise, reproducible temperature gradients. In our apparatus the sample holding assembly has an enormous heat capacity in comparison with the sample and so the base temperature is essentially unaffected by the temperature gradient across the sample. Our temperature gradients, typically about 2 K/cm, are reproducible to one part in 10^3.

Our sample material was a $Tl_2Ba_2CaCu_2O_{8+\delta}$ epitaxial thin film. The Tl-Ba-Ca-Cu-O family of compounds displays a particularly simple systematic behavior of the thermoelectric power (TEP), which should aid eventual interpretations of Nernst results. More importantly, the Tl-based materials have a tetragonal structure and so we do not have to deal with the in-plane, a-b anisotropy of the thermoelectric power as in $YBa_2Cu_3O_7$. We used a commercially available $Tl_2Ba_2CaCu_2O_{8+\delta}$ epitaxial film (Superconductor Technologies Inc., 460 Ward Drive, Santa Barbara, CA 93111-2310).

A simplified diagram of the experimental arrangement is shown in Fig. 1. The sample was held, film-side down, by one edge. The cantilevered end of the sample was heated by light pulses from a tungsten lamp which were brought into the magnetic field region through a light pipe. The sample-holding assembly consisted of copper pieces which had been nickel plated for high albedo. The light pulses had no measurable effect on the base temperature, despite temperature differences of typically 1.0 to 1.5 K across the sample. The temperature gradients were measured by monitoring the thermal emf at the TEP contacts and calibrated by subsequent measurement of the Seebeck coefficient with a standard differential thermocouple. Silver contacts were evaporated following oxygen plasma removal of the film surface layer. The contacts were annealed at 400 °C for 4 hours. Wires were attached with silver epoxy (EPOTEK H20E) which was pumped to 10^{-6} torr before curing at 100 °C for 18 hours.

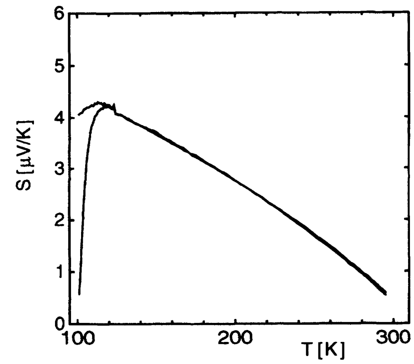

FIG. 2. Temperature dependence of the Seebeck coefficient of the $Tl_2Ba_2CaCu_2O_{8+\delta}$ film used in the Nernst measurement. A 14 T magnetic field along the c axis broadens the superconducting transition with no effect on the normal-state thermopower (Ref. 29).

Results for the Seebeck coefficient in zero magnetic field and at 14 T are shown in Fig. 2. The superconducting transition is broadened in the field but the curves are indistinguishable from each other in the normal state, as expected. Negligible normal-state magnetothermopower is a well-known feature of the hole-doped cuprate superconductors.[29]

The signal from the transverse (Nernst) contacts at three different magnetic-field strengths (0, +14 T, −14 T) is shown in Fig. 3. A large signal from superconducting fluctuations persists up to $T = 130$ K. At higher temperatures, the much smaller normal-state signal is apparent. The inset in Fig. 3

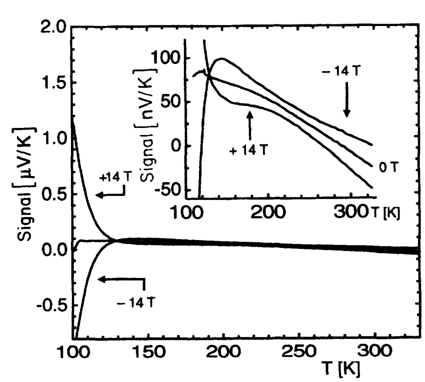

FIG. 3. Signal from the Nernst contacts at three magnetic field strengths (0, 14 T, −14 T), showing the large fluctuation Nernst effect. The inset expands the signal in the normal state. The zero-field curve is a residual thermopower signal from small contact misalignment.

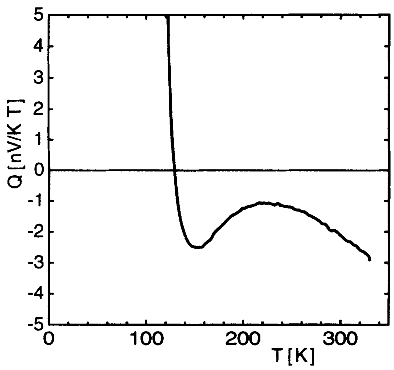

FIG. 4. The measured Nernst coefficient in the normal state of the $Tl_2Ba_2CaCu_2O_{8+\delta}$ film.

expands the normal-state signal, which is superimposed on a small residual thermopower voltage from contact misalignment.

The final result for the Nernst coefficient, Q, is shown in Fig. 4. The Nernst coefficient is negative in the normal state with a local maximum in magnitude near $T=150$ K. After a broad minimum near $T=220$ K, $|Q|$ increases up into our maximum temperature of 330 K.

How do we interpret the results? This is a measurement of the Nernst effect in the normal state of a cuprate superconductor and so a detailed interpretation may be premature, but as a starting point recall that the Nernst coefficient measures the energy dependence of the relaxation time: Since in the Nernst geometry there is no applied current, at any moment equal numbers of charge carriers are drifting up the thermal gradient as down it. The magnetic-field induced transverse velocities cancel, yielding zero Nernst voltage, unless the relaxation time is energy dependent, since the carriers moving down the gradient are more energetic. The result is that Nernst effect is sensitive to the energy derivative of the relaxation time, $d\tau/d\varepsilon$, where ε is the energy. In a free-electron metal with nondivergent $\tau(\varepsilon)$, the Nernst coefficient is given to leading order by

$$Q=\frac{\pi^2 k_B^2 T}{3m^*}\left[\frac{\partial\tau(\varepsilon)}{\partial\varepsilon}\right]_{\varepsilon=\mu}, \qquad (2)$$

where m^* is the effective mass and μ is the chemical potential.[27] In the past, the Nernst effect has not occupied an important place in the study of metals, partly due to experimental difficulty but also because an *a priori* determination of the energy dependence of the electron-phonon coupling in a real material is no easy task. In contrast, the apparent simplicity of the T^2 relaxation rate in the cuprates, which may be purely of electronic origin, suggests that the Nernst effect could be an ideal probe of the underlying physics. In the cuprates where $\tau_H \neq \tau_{tr}$, it is clear that Eq. (2) must be generalized.[28] We have assumed here, for purposes of discussion, that Q in the cuprates is a measure of $\partial\tau_H/\partial\varepsilon$. A more complete theoretical treatment is clearly desirable, and we hope that these measurements will stimulate effort in this area.

If Q is proportional to $\partial\tau_H/\partial\varepsilon$, then the sign of Q suggests that the cyclotron scattering rate for holelike carriers increases with energy, as would be expected if the energetics of the scattering process were dominated by phase-space factors. Nevertheless, such an interpretation may be premature since we do not yet understand, even qualitatively, the details of the nonmonotonic temperature dependence of Q in the normal state of $Tl_2Ba_2CaCu_2O_{8+\delta}$. We anticipate that further Nernst measurements in this and other cuprates will clarify this interesting behavior.

In summary, we have successfully measured the normal-state Nernst coefficient, Q, in one of the cuprate superconductors, $Tl_2Ba_2CaCu_2O_{8+\delta}$, for temperatures ranging from T_c to above room temperature. The signal is small, approximately -2 nV/K T. The negative sign of Q probably indicates a positive energy derivative of the cyclotron scattering rate. The temperature dependence of Q is puzzling and requires further study.

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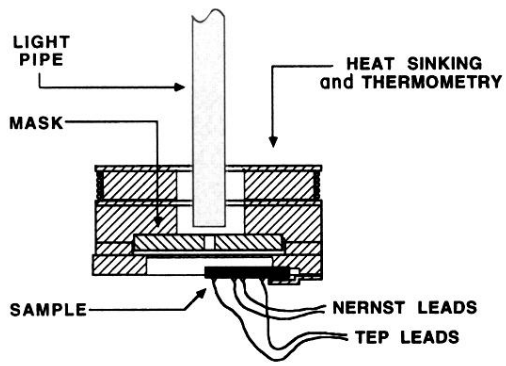

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