Unusual magnetic-field dependence of the electrothermal conductivity in the mixed state of cuprate superconductors

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In the mixed state of a superconductor, the electrothermal conductivity, P, which measures the electrical current density produced by a thermal gradient, is supposed to be independent of both the magnetic field and the details of the vortex motion. Measurements of the thermopower and resistivity in $YBa_2Cu_3O_{7-\delta}$, $Bi_2Sr_2CaCu_2O_{8+\delta}$, and $Tl_2Ba_2CaCu_2O_{8+\delta}$ show an unusual, sharp structure in P at low field. In $Bi_2Sr_2CaCu_2O_{8+\delta}$, the extra contribution is negative, causing a sign change of the thermopower versus applied magnetic field. The possible origins of the low-field anomaly are discussed.

I. INTRODUCTION

The electrothermal conductivity, relating electric currents to thermal gradients, is, in principle, the least varied of all transport properties of superconductors in the mixed state. Nearly featureless, it should be independent of the magnetic field,[1,2] independent of the vortex viscosity,[3] and remain unchanged for pinned and unpinned vortices. It should be equal to its value in the normal state.[1,2]

Microscopically, the electrothermal conductivity is also a remarkably direct probe of the mixed state. Whereas most transport quantities represent a balance between a driving force and viscous relaxation, the vortex viscosity plays no role in the electrothermal conductivity. This permits study of the driving force, which, in this case, is ultimately due to the normal carriers in the vortex core. The apparent ''featurelessness'' of the electrothermal conductivity merely expresses the fact that the excitations within the vortex core can be treated as normal, nonsuperconducting electrons.

We have found that the situation is more complicated with the cuprate superconductors. We have measured a pronounced low-field peak or divergence of the electrothermal conductivity in $YBa_2Cu_3O_7$, $Bi_2Sr_2CaCu_2O_{8+\delta}$, and $Tl_2Ba_2CaCu_2O_{8+\delta}$. The peak or divergence narrows and moves to lower magnetic field values as the temperature is increased. In $Bi_2Sr_2CaCu_2O_{8+\delta}$ the contribution is negative in sign, causing a sign change of the mixed-state thermopower.

A. Mixed-state thermopower

The electric current, $\vec{j}_e$, and heat current, $\vec{j}_h$, which give rise to thermoelectric phenomena are given by

$$\vec{j}_e = \hat{\sigma}\vec{E} - \hat{P}\vec{\nabla}T, \quad (1)$$

$$\vec{j}_h = -T\hat{P}\vec{E} + \hat{\kappa}\vec{\nabla}T, \quad (2)$$

where $\hat{\sigma}$ is the electric conductivity, $\hat{\kappa}$ is the thermal conductivity, and $\hat{P}$ is the electrothermal conductivity.[4] Since no electric current flows during the measurement, the thermopower tensor, $\hat{S}$, relating the electric field to the thermal gradient, $\vec{E}=\hat{S}\vec{\nabla}T$, is $\hat{S}=\hat{P}\hat{\sigma}^{-1}$ and the usual, longitudinal thermopower is

$$S_{xx} = P_{xx}\rho_{xx} - P_{xy}\rho_{xy}, \quad (3)$$

where $\hat{\rho}$ is the resistivity tensor. The last term is the Nernst signal times the tangent of the Hall angle, $N\tan\theta_H$, which is negligible for high-T_c materials because of the smallness of the Hall angle and the comparable magnitudes of the thermopower and Nernst voltage. Keeping only the first term gives the usual scalar equation, valid for the cuprates:

$$S = P\rho. \quad (4)$$

The microscopic forces giving rise to the thermopower signal are not the same in a superconductor as in metals or semiconductors.[5] In a metal, the picture is simple—the electric force from charge carriers which have migrated cancels the force from the thermal gradient. In the superconductor, temperature gradient generates a current of normal electrons within each vortex core: $\vec{j}_e = -P_n\vec{\nabla}T$, with the subscript indicating the normal-state value of P, corresponding to normal-core excitations. Charge continuity requires a superconducting backflow current around the vortex, which in turn produces a Lorentz force moving the vortex perpendicular to the thermal gradient.[5] The resulting transverse motion of the vortex creates an electric field parallel to the original current, $\vec{E} = -\vec{v}_L \times \vec{B}$. Consideration of the sign of the vortex thermopower, which has the same sign as the normal-state thermopower, confirms that the superconducting backflow is the source of the Lorentz force on the vortex.

The microscopic origin of the thermopower voltage is very similar to the origin of the mixed-state resistivity. The key difference is that the resistivity is measured with an externally applied current; whereas, in the thermopower measurement, the vortex cores themselves are the source of the current. The comparison is made explicit by expressing the electric field in terms of the Lorentz force, $\vec{F}_L$, and vortex

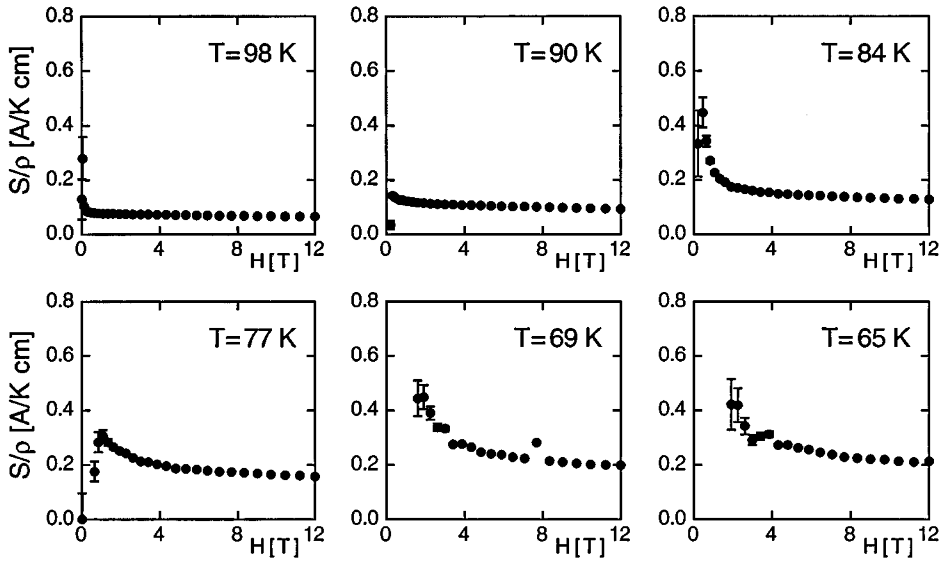

FIG. 1. Magnetic-field dependence of the electrothermal conductivity, P, of the $Tl_2Ba_2CaCu_2O_{8+\delta}$ thin-film sample at various temperatures. The error bars grow at lower magnetic fields as both the thermopower and resistivity approach zero.

viscosity, η: $\vec{E}=\vec{B}\times\vec{F}_L/\eta$. During the measurement of the vortex resistivity, the Lorentz force on the vortex is $\vec{F}_L=\vec{j}_{\mathrm{ext}}\times\vec{\Phi}_0$, where $\vec{j}_{\mathrm{ext}}$ is the externally applied current density and $\vec{\Phi}_0$ is a vector in the direction of the magnetic field with magnitude of the flux quantum. During the thermopower measurement, the Lorentz force from the thermally excited normal core current is $\vec{F}_L=P\vec{\nabla}T\times\vec{\Phi}_0$. The experimental ratio of the thermopower electric field to the resistive electric field is

$$\frac{\vec{E}^T}{\vec{E}^R}=\frac{\vec{B}\times(P\vec{\nabla}T\times\vec{\Phi}_0)/\eta}{\vec{B}\times(\vec{j}_{\mathrm{ext}}\times\vec{\Phi}_0)/\eta}=P\frac{|\vec{\nabla}T|}{|\vec{j}_{\mathrm{ext}}|}\quad\text{with}\quad\vec{\nabla}T\|\vec{j}_{\mathrm{ext}},\qquad(5)$$

where $\vec{j}_{\mathrm{ext}}$ and $\vec{\nabla}T$ are experimentally controlled parameters.

The key point to notice is that the vortex viscosity cancels from the ratio of the measured electric fields. The vortex viscosity does not play a role[3] in the experimentally determined electrothermal conductivity because the vortex velocity which gives rise to the electric field is, in each case, limited by the same vortex viscosity, which cancels when we form the ratio. The electrothermal conductivity is limited by the dissipation of the normal-core excitations, not the vortex viscosity.

B. Magnetic-field independence of the electrothermal conductivity

Equation (5) indicates that the electrothermal conductivity is also independent of the magnetic field. The physical reason for the independence of the magnetic field is that each vortex creates its own local backflow current density. The measured, field-independent value of the mixed-state P_s should be identical to the normal-state value, P_n, because the amount of current is determined by the normal-core excitations.[1] The normal-state value is expressed by the usual Boltzmann integral over occupied states, as in standard textbooks

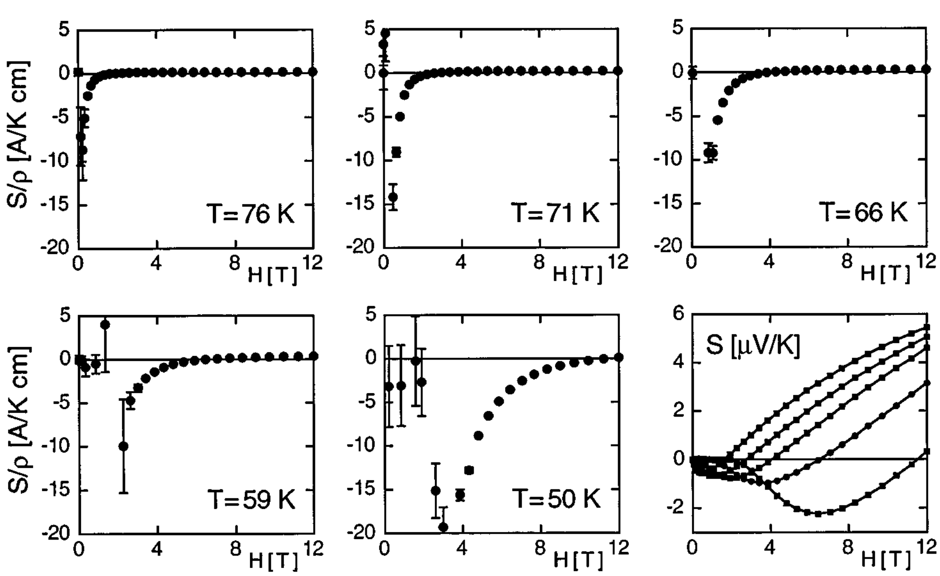

FIG. 2. Magnetic-field dependence of the electrothermal conductivity, P, of the $Bi_2Sr_2CaCu_2O_{8+\delta}$ thin-film sample at various temperatures. The low-field peak or divergence is negative in sign, producing a sign change of the thermopower as a function of magnetic field and temperature, shown in the lower right panel, with decreasing temperature from left to right.

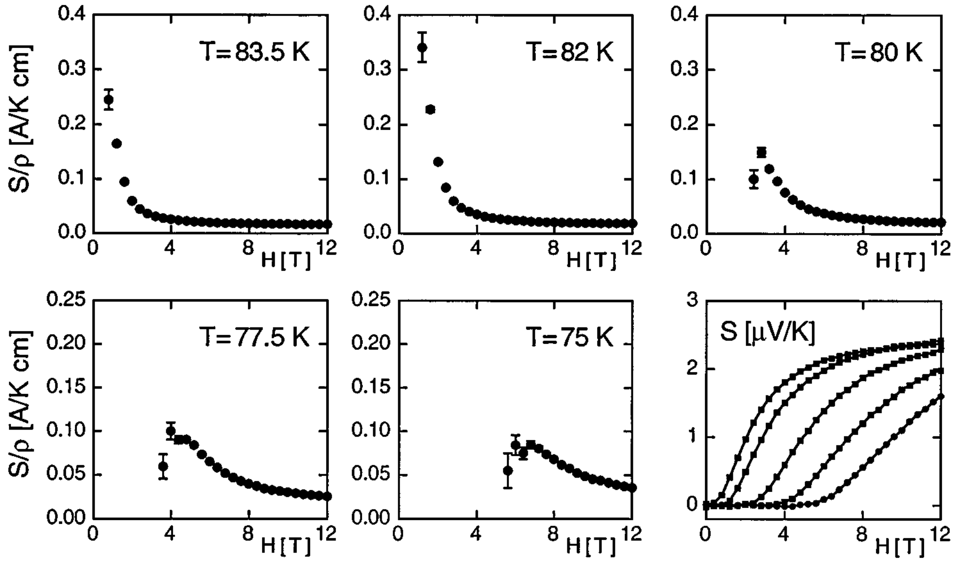

FIG. 3. Magnetic-field dependence of the electrothermal conductivity, P, of the $YBa_2Cu_3O_{7-\delta}$ thin-film sample at various temperatures. At high fields and temperature, P approaches a constant value, but increases sharply at low field. The lower right panel shows the raw thermopower data, with decreasing temperature from left to right.

$$P_s=P_n=\frac{e}{T}\int\frac{d\mathbf{k}}{4\pi^3}\left(-\frac{df}{d\epsilon}\right)(\epsilon-\mu)\mathbf{v}^2\tau(\epsilon), \tag{6}$$

where μ is the chemical potential and τ is the scattering lifetime.

Experimentally, for low-temperature superconductors, it is indeed the case that the electrothermal conductivity below T_c is found to be independent of the magnetic field and equal to its normal-state value. In the case of niobium, the equality of P_s and P_n was established experimentally by Fiory and Serin.[2] Figure 4 of Ref. 2 shows their data on the magnetic-field dependence of P in both superconducting and normal niobium. The value of P was found to be constant, field independent, and continuous across the phase boundary from the superconducting to the normal phase.

II. EXPERIMENT

Previous studies of the mixed-state thermopower of the cuprates have concentrated on comparisons with Eq. (4), indicating the proportionality of the thermopower and resistivity. A study of the superconducting transition widths of thermopower and resistivity curves at various fixed magnetic field strengths confirmed the similarities of these two quantities.[6] For the present work, we wished to examine the detailed magnetic-field dependence of the electrothermal conductivity. Accordingly, for each sample the temperature was first stabilized and the magnetic field subsequently swept to a succession of 61 field values between -12 T and $+$ 12 T, with emphasis on small field strengths. Only after the field stabilized at each value was the thermopower measurement initiated. The field was oriented along the c axis of each compound.

We reproducibly measure thermoelectric voltages with a precision of 1.5 nanovolts. Since the cuprates have thermopower signals of a few microvolts per kelvin, using a ΔT of 0.5 K across the sample means that our signal-to-noise ratio for thermopower measurement typically exceeds a few thousand at the highest magnetic field strengths.

The $Bi_2Sr_2CaCu_2O_{8+\delta}$ thin-film sample, with $T_c=80$ K, was prepared by atomic layer-by-layer molecular beam

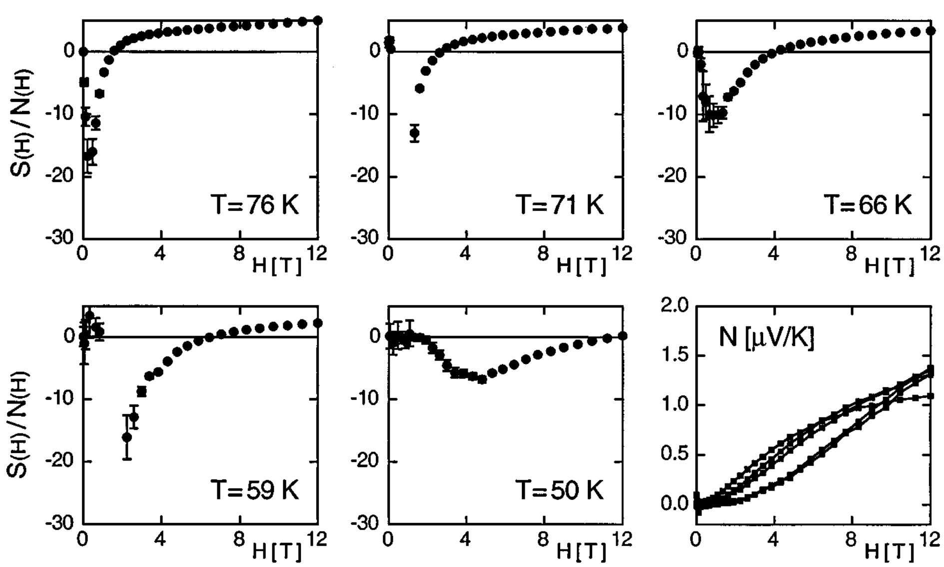

FIG. 4. Ratio of the thermopower to the Nernst voltage in $Bi_2Sr_2CaCu_2O_{8+\delta}$. The two signals are measured simultaneously and the ratio cancels effects of the vortex viscosity. The low-field anomaly remains, confirming the behavior of P. The field dependence of the Nernst signal is shown in the lower right panel, with decreasing temperature from left to right.

epitaxy (ALL-MBE), in which the surface chemistry is controlled layer-by-layer as the film is grown, as has been described in detail elsewhere.[7] The $YBa_2Cu_3O_{7-\delta}$ (Ref. 8) and $Tl_2Ba_2CaCu_2O_{8+\delta}$ (Ref. 9) samples were commercially available epitaxial films with $T_c = 87$ K and 105 K, respectively.

We verified that none of our measured curves were hysteretic. We further verfied through normal-state measurements that P_n was independent of magnetic field, $d\ln P_n/dH < 10^{-3}\ T^{-1}$.

The current used during the resistivity measurements was 100 μA. The high current was used to generate resistive emf's of the same order of magnitude as the thermal emf's from the thermopower measurements. The resistivity results were unchanged when the current was ten times smaller or larger. We verified the linearity of the thermopower voltage with the thermal excitation, varying the applied temperature gradient from 0.03 K/cm to 0.50 K/cm.

Our sample temperature was controlled with a Cernox[10] temperature sensor. Temperature fluctuations were less than 30 mK at all fields and temperatures. Thermal excitation of the sample for the thermopower measurement was achieved with light from a tungsten lamp which was fed into the sample region via a light pipe. This heating method is intrinsically independent of the magnetic field, allowing a calibration of the very slight field dependence of the T-type thermocouples which monitored the sample temperature gradient.

Corrections to the thermopower from the last term in Eq. (3), the Hall rotation of the Nernst signal, which were necessary in the case of niobium,[2] are, as previously mentioned, insignificant in the case of the high-T_c cuprates. In $Tl_2Ba_2CaCu_2O_{8+\delta}$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$ we confirmed this at by measuring all four signals—resistivity, Hall, Nernst, and Seebeck. The correction was less than 0.3% of the total signal at all field values.

We have corrected for the thermopower of the Au voltage leads by subtracting the zero-field thermopower signal. The small positive magnetothermopower of the gold-wire leads introduces a very small systematic underestimate of the sample thermopower because the measurement gives the difference of the sample thermopower and wire thermopower. Accurate absolute thermopower calibrations are not available for our wires in all fields and temperatures. We have, however, estimated the magnitude of the error in our data by remeasuring one sample with Cu wire leads. Since the magnetothermopower of the Cu wires is at least five times larger than for Au wires at $T > 60$ K,[11] comparison of the two results lets us place bounds on the total error. The maximum underestimate of the sample thermopower is probably around 5% at the lowest temperature and near 0.1% at the highest temperature measured. Measurements below the irreversibility line of $YBa_2Cu_3O_{7-\delta}$ provide even stricter upper bounds.

III. RESULTS

A. $Tl_2Ba_2CaCu_2O_{8+\delta}$

Results for the magnetic-field dependence of the electrothermal conductivity, P, at various temperatures in the $Tl_2Ba_2CaCu_2O_{8+\delta}$ sample are shown in Fig. 1. Only at the highest temperature and in the high-field limit does P become independent of the magnetic field, as was found for conventional, low T_c materials.[2] It is tempting to identify the high-field, asymptotic, constant value of P with the the normal-state value, P_n. In this case, the temperature dependence of the constant part reflects the T dependence of the normal-state thermopower continued below T_c.

At low field, contrary to expectation from conventional superconductors, the value of P increases rapidly with decreasing field. The increase becomes more pronounced at lower temperature. It is difficult to determine whether the data indicate a low-field divergence of P, or a narrow maximum, with P continuing to decrease at still lower field values. The increased error at low field complicates the determination. The data at $T = 77$ K seem to indicate a maximum and the data at other temperatures are not inconsistent with that interpretation. Some data from other samples below are also suggestive of a narrow maximum.

B. $Bi_2Sr_2CaCu_2O_{8+\delta}$

Results for $Bi_2Sr_2CaCu_2O_{8+\delta}$ are more striking and are shown in Fig. 2. Here the low-field anomaly is negative in sign. The negative sign of the low-field feature in P actually causes a sign change of the mixed-state thermopower as a function of magnetic field and temperature. The raw thermopower data, showing the sign change, are shown in the lower right panel of Fig. 2.

C. $YBa_2Cu_3O_{7-\delta}$

Results from $YBa_2Cu_3O_{7-\delta}$ are shown in Fig. 3. In high magnetic fields and especially at the highest temperatures, P approaches a constant, field-independent value. At low fields there is a very sharp increase of P, which broadens and moves to higher magnetic fields with decreasing temperature, as in Figs. 1 and 2.

IV. DISCUSSION

A. Comparison with the Nernst effect

As described in Sec. I A, dividing the thermopower by the resistivity eliminates the vortex viscosity and permits us to extract the driving force, in this case, the thermally excited normal-core current. Details of the experimental procedure which allow comparison of the two measurements were described in Sec. II. In order to remove any remaining concerns about the data because the two measurements are not simultaneous, we found another method to cancel the vortex viscosity, which confirms the results.

The Nernst effect, $N(H)$, arises from the entropy associated with the suppression of the condensate at the vortex core.[12] In a temperature gradient, the vortex lowers its free energy by moving to lower temperature, producing a transverse voltage. As Huebener[13] has stressed, the Nernst effect and the thermopower arise from physically distinct forces, so direct comparison of the two signals is often not meaningful. In our case, it is only important that the entropy per vortex, which drives the Nernst effect, is well behaved as the magnetic field is reduced.

Unlike the electrothermal conductivity, the ratio of $S(H)$ to $N(H)$ may not have a direct physical interpretation, but is useful as an experimental verification. In both the thermopower and Nernst measurements, the vortex line velocity is limited by the vortex viscosity. For $Bi_2Sr_2CaCu_2O_{8+\delta}$, it was possible to measure both signals simultaneously, ensuring identical conditions for comparing the two signals. Results for the ratio of $S(H)$ to $N(H)$ in $Bi_2Sr_2CaCu_2O_{8+\delta}$ are shown in Fig. 4. It is apparent from the data of Fig. 4 that the low-field behavior of P is real and is not an artifact of the experiment. In addition, the data in Fig. 4 for $T=76$ K, 66 K, and 50 K suggest strongly that we have measured a local extremum of P, instead of a low-field divergence.

B. Absence of pinning effects with conventional vortices

The data in Figs. 1, 2, and 3 all display sharp features near the magnetic-field strength where the thermopower and resistivity signals approach zero. It would seem natural to ascribe the observed behavior to the onset of strong vortex pinning. The main difficulty is that the microscopic mechanism is missing. Within the conventional understanding, pinning forces should not affect the electrothermal conductivity. Indeed, its insensitivity to pinning contributes much to its attractiveness as a probe of the mixed state. We now mention briefly the reasons why pinning of conventional vortices should not alter P. Later, in Sec. IV D below, we do note a possible way out, using a model much discussed for the cuprate superconductors, in which the pinning forces may, in the end, lead to the peaks such as might be seen in Figs. 1, 2, and 3.

Because P is a measure only of the current density excited by the thermal gradient across the vortex core, a stationary vortex should have a finite electrothermal conductivity just as the moving vortex does. It is the dissipation of the normal excitations within the core which limits P, not the vortex pinning potential. As discussed in Sec. I A, dividing the thermopower by the resistivity to obtain P should remove effects of the vortex viscosity.

The discussion above pertains to linear effects of the vortex viscosity and there remains the possibility that nonlinear forces could, in principle, affect the experimental results. In that case, the electric field would show a nonlinear dependence on the driving force. As described in Sec. II, the measured thermopower voltages did not deviate detectably from linearity in the applied temperature gradient, despite gradients which ranged over $1\frac{1}{2}$ orders of magnitude.

C. Effect of normal quasiparticle transport $T<T_c$

A growing amount of evidence from recent experiments has indicated the strong possibility that many of the cuprate superconductors possess an unconventional superconducting order parameter with zeros at the Fermi surface. It is reasonable to expect that the normal carriers at the zeroes of the gap should also contribute to electric and heat currents below T_c. Indeed, recent measurements in $YBa_2Cu_3O_{7-\delta}$ have shown that mixed-state transport is strongly affected by a high density of uncondensed normal quasiparticles.[14–16] Studies of the Hall effect[15–17] have seen separate additive vortex and quasiparticle contributions to the Hall conductivity, σ_{xy}. The two terms are easily distinguished by their different dependences on the applied magnetic field strength.

Perhaps more relevant for the thermopower are some recent measurements of the Righi-Leduc effect in $YBa_2Cu_3O_7$,[14] which have characterized thermal transport by the quasiparticles below T_c. From these measurements we learn that the entropy carried by the quasiparticles is, as expected, independent of the applied magnetic field, but that their mean free path is not. Vortices are significant scatterers of quasiparticles below T_c.

We account for the quasiparticle contribution to P by requiring that all the currents—normal core, supercurrent, and the uncondensed quasiparticles—sum to zero, as usual in the thermopower measurement

$$\vec{j}=(\sigma_n+\sigma_s+\sigma_{qp})\vec{E}-(P_n+P_{qp})\vec{\nabla}T=0\ . \tag{7}$$

The sum of the electrical conductivities is just the inverse of the measured resistivity, which means that the ratio of the thermopower to the resistivity is

$$S/\rho=P_n+P_{qp}, \tag{8}$$

where the quasiparticle contribution, P_{qp}, presumably would have to account for the sharp peak or divergence of the measured electrothermal conductivity at low field.

The difficulty is that P_{qp}, which follows the Boltzmann form, Eq. (6), is a smooth function of H at low magnetic field. From Ref. 14 we know that P_{qp} from Eq. (6) depends on the magnetic field only through the relaxation time, $\tau(H)$, from scattering by vortices. From the data of Ref. 14 we observe that P_{qp} must follow

$$P_{qp}\propto\frac{1}{a|H|+b}, \tag{9}$$

where the coefficient, a, is proportional to the quasiparticle scattering cross section presented by the vortices, and the coefficient, b, represents scattering from other sources. At these temperatures, even in very pure $YBa_2Cu_3O_7$ single crystals, the coefficient, b, is not small in comparison with the vortex scattering term. The measured relaxation time, $\tau(H)$, and therefore P_{qp} from Eq. (9), is well behaved as H goes to zero. Indeed, P_{qp} from Eq. (9) varies monotonically with H, unlike the measured data. Finally, we determined that the data of Figs. 2 and 3 increase much faster than $1/H$ at low field, which most probably cannot be explained by quasiparticle transport alone.

D. Charge redistribution effects

If the data of Figs. 1, 2, and 3 are related to the onset of strong vortex pinning, then the observed divergent response is reminiscent of the critical behavior of the electric polarization in charge-density-wave materials near the pinning/depinning transition. In charge-density-wave systems below threshold, the electric field polarizes the condensate.[18] The polarization increases in strength with increasing electric field, reaching a maximum at the depinning threshold.[18] The polarization was predicted[19] and measured[18] to show critical behavior, corresponding to a phase transition between the pinned and unpinned states.

The critical polarization is possible in charge-densisty-wave materials because of the internal charge degree of freedom—positive ions moving oppositely to negative electrons. The conventional vortex lattice lacks this freedom. Several authors,[20,21] however, have recently suggested models of vortices in the cuprates in which the charge density at the vortex core, n_0, differs from the asymptotic charge density far from the vortex, n_∞. The charge difference, δn, has been invoked as the origin of the second term of the Hall conductivity.[20,21]

In a lattice of such vortices, where charge neutrality is obeyed on average but not everywhere locally, deformations of the lattice produce local electric polarizations. Strain on the pinned vortex lattice due to a thermal gradient would correspond to the strain on charge density waves below threshold, possibly producing a measurable electric signal. Whether or not this picture works microscopically for a vortex lattice and whether or not it can generate critical behavior of the electric response at the depinning transition, as is the case for charge-density waves, remains an open question and an interesting avenue for further theoretical development.

One important conclusion can be phrased in more general terms. The electrothermal conductivity, P, is odd under the operation of charge conjugation because it depends on the sign of the transported charges. Our data indicate the presence of an additional term in the electrothermal conductivity of the cuprates, a term of either sign in materials which have holelike normal-state thermopowers. The obvious question is what determines the sign of the contribution. Relating the term to vortex dynamics alone would probably require breaking the particle-hole symmetry of the vortex state. As described above, such a scenario is already much discussed by other researchers and may play a central role in understanding other experiments.

ACKNOWLEDGMENTS

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