Unusual Seebeck and Nernst effects in the mixed state of $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_\delta$

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Measurements of the Nernst and of the Seebeck coefficient in the mixed state on a polycrystalline sample of $Bi_{1.76}Pb_{0.24}Sr_2Ca_2Cu_3O_\delta$ are presented for temperatures between 70 and 120 K and magnetic fields up to 5 T. Nernst and Seebeck voltages are both large. The Seebeck voltage is larger by a factor of about 1000 than expected from the conventional flux creep-flow model. It is shown that our results can be understood if in addition to the flux flow-creep mechanism another important mechanism of dissipation is active. A possible mechanism based on fluctuations of the order parameter over internal weak links is discussed.

Unusual, strong dissipation is observed in the mixed state of the high-T_c superconductors (HTSC). Prominent manifestations of this dissipation are the unusual broadening of the resistive transition in a magnetic field[1] and relaxation phenomena observed in the magnetization.[2,3] In view of these large effects one expects that thermomagnetic effects can be observed in the mixed state also.

In this paper we present measurements of the Seebeck and the Nernst effect on a polycrystalline sample of $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_\delta$ for temperatures between 70 and 120 K and magnetic fields up to 5 T. We find that both effects are large, the ratio of transverse to longitudinal thermal electrical fields being of order 1. This is inconsistent with a conventional flux creep-flow mechanism as the only source of the dissipation, since in that case one expects the Seebeck effect to be smaller than the Nernst effect by a factor of 1000.[4] We show that our results can be explained, if one assumes that an additional important mechanism of dissipation is active in the mixed state. A possible such mechanism, which is closely related to the presence of internal (intergranular and intragranular) weak links,[2,5] is discussed.

The measurements were carried through on one of a series of polycrystalline samples of $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_\delta$ (Bi-Pb-Sr-Ca-Cu-O), which showed reproducible superconducting properties. Sample preparation and characterization as well as a study of the superconducting properties have been presented in Ref. 6. The sample used for the present measurements shows zero resistance in zero magnetic field at $T_{c0}=T_{c(B=0)}\approx 108.5$ K.

The Seebeck coefficient S and the Nernst-coefficient Q are defined as $\nabla T S=\mathbf{E}_l$ and $Q(\nabla T\times\mathbf{B})=\mathbf{E}_t(j_{\mathrm{el}}=0)$, where $\mathbf{E}_l$ and $\mathbf{E}_t$ are the longitudinal and transverse components of the electric field with respect to the temperature gradient ∇T and $\mathbf{B}$ is the magnetic field (e.g., Ref. 7). S and Q were measured as function of magnetic field (swept at about 0.5 T/min) at fixed temperatures stabilized within about 50 mK with an imposed temperature gradient ∇T of about 0.5 to 1 K. The arrangement of the sample allowed measurements for $\nabla T \parallel \mathbf{B}$ and for $\nabla T \perp \mathbf{B}$. The temperature gradient, built by a small heater (manganin; $d\rho/dB\approx 0$) using a constant heating current, was measured via calibrated Pt resistors mounted to the ends of the sample. The magnetic-field dependence of the resistance of Pt in the temperature range of our measurements is reported to be weak [$\Delta R(H,T)/R(0,T)<10^{-3}$, Ref. 8]. While raising the magnetic field, the temperature gradient itself, as well as the heating power, were controlled to be constant. The thermomagnetic voltages were measured using calibrated Cu wires with an accuracy of about 50 nV. The zero-field voltages for temperatures $T<T_{c0}$ (where the thermopower of Bi-Pb-Sr-Ca-Cu-O is zero) revealed the thermopower of Cu, S_{Cu}, within a few percent. The magnetic field dependence of S_{Cu} between 70 and 120 K is reported to be weak ($dS/dB\approx 10$ nV/K T) (Ref. 9) and was neglected when calculating the absolute thermopower of Bi-Pb-Sr-Ca-Cu-O [$S(B,T)=\Delta U(B,T)/\Delta T+S_{\mathrm{Cu}}(T)$].

The adiabatic Nernst and Seebeck effects, which correspond to zero transverse entropy flux in steady state, may differ from the isothermal effects which require the transverse temperature gradient ∇T_t to be zero if the Righi Leduc effect $\nabla T_t=M(\mathbf{B}\times\nabla T)$ is large (M—Righi Leduc coefficient; ∇T—imposed temperature gradient). ∇T_t may itself cause Nernst and Seebeck voltages, which add to the primary voltages due to ∇T (e.g., Ref. 7). We measured the Righi Leduc effect and found that it is negligibly small ($\nabla T_t<10^{-2}\nabla T$). Therefore our results represent the isothermal effects.

Figure 1 shows the experimental results for the Seebeck coefficients, $S_\parallel(\nabla T\parallel\mathbf{B})$, $S_\perp(\nabla T\perp\mathbf{B})$ and the Nernst coefficient, $Q(\nabla T\perp\mathbf{B})$ as a function of temperature for various fixed magnetic fields, between 70 and 120 K and 0 and 5 T, as extracted from the magnetic-field dependence of $S_\parallel$, $S_\perp$, and Q measured at fixed temperatures. The Seebeck voltage is independent of reversal of the sign of B. The transition to the superconducting state is sharp for $B=0$ and the absolute value of S just above the transition agrees with the typical values reported for the normal-state Seebeck coefficient of the HTSC (e.g., Refs. 10 and 11). For finite magnetic fields one observes a striking broadening of the transition.[11] The overall features are very similar to the ones observed for the magnetoresistive transition—whereas T_{c0} is suppressed strongly from 108.5 K at $B=0$ to about 70 K for $B \approx 5$ T, the onset temperature is unaffected by the magnetic field. Above the superconducting transition the magnetic-field dependence of S is weak. Remarkably S is rather independent of the direction of the temperature gradient with respect to the magnetic field; i.e., qualitatively we find that $S_{\perp}/S_{\parallel}$ is of the order 2.

The Nernst voltage changes sign on reversal of the magnetic field and it is small in the normal state as expected. In the superconducting state the Nernst voltage grows to the same order of magnitude as the Seebeck voltage. For example, at 100 K and 1 T both voltages, QB and S, are of the order of 1 μV/K. Note that the Nernst voltage does not become larger than the Seebeck voltage.

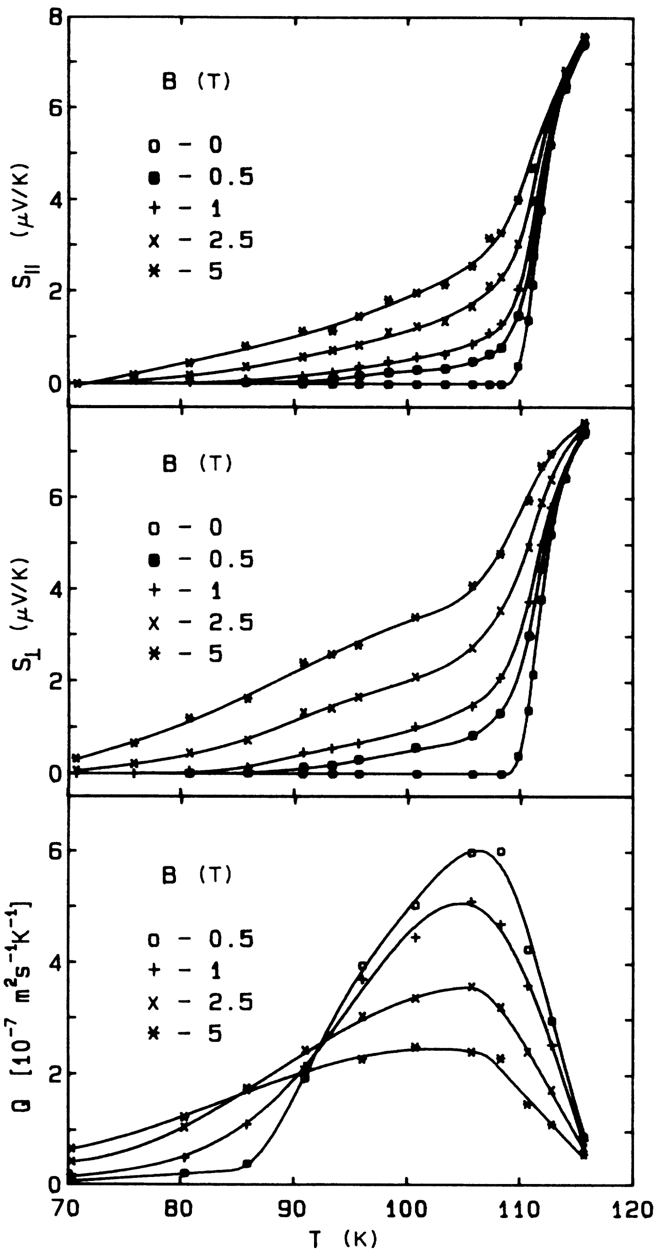

FIG. 1. Seebeck coefficients $S_{\parallel}(\nabla T \parallel \mathbf{B})$, $S_{\perp}(\nabla T \perp \mathbf{B})$ and Nernst coefficient Q of Bi-Pb-Sr-Ca-Cu-O as a function of temperature T at fixed magnetic fields B given in the figure (see text). The solid lines are a guide to the eye.

We mention that our results do not depend on the direction of the magnetic field with respect to the sample, as expected for a polycrystal—i.e., effects due to texture can be excluded.

The "classical" mechanism for the appearance of thermomagnetic voltages in the mixed state of type-II superconductors is the temperature-gradient-driven motion of flux lines (e.g., Ref. 4 and references therein). A moving flux line carries entropy. This flow of entropy is compensated by the appearance of a temperature gradient. Conversely, the presence of a temperature gradient ∇T perpendicular to the magnetic field leads to flux line motion parallel to ∇T and thus to an "induced" voltage perpendicular to this motion—i.e., to a Nernst voltage. A longitudinal Seebeck voltage arises, since a flux line moving with velocity v_f is subject to a "Magnus force" ($\mathbf{F}_m \sim \mathbf{v}_f \times \mathbf{B}$), which leads to a small component of motion perpendicular to the driving force. One usually expects the Seebeck voltage to be by far smaller than the Nernst voltage and this is indeed found in measurements on conventional superconductors (e.g., Ref. 4 and references therein). The ratio $S_{\perp}/QB$ is determined by the Hall angle α—i.e., $S_{\perp}/QB \approx \tan\alpha$ (typically of the order 10^{-3}). Note that this is just opposite to (1) the normal state, where $\tan\alpha = QB/S_{\perp}$, and (2) to the case when an external current is the source of flux-line motion: In that case the Lorentz force between the external current and the flux line gives rise to flux-line motion perpendicular to the current, and thus the longitudinal (resistive) voltage is by a factor $1/\tan\alpha$ larger than the transverse (Hall) voltage (i.e., $\tan\alpha = R_H B/\rho$) as usual, where R_H and ρ are the Hall coefficient and the magnetoresistivity, respectively.

An interpretation of the present results along the lines of a flux-creep/flow model leads to serious inconsistencies.

(1) In the superconducting state the ratio of Nernst to Seebeck voltage, which from our experiment is roughly 1, does not correspond to the Hall angle of order 0.1° found for all of our Bi-Pb-Sr-Ca-Cu-O samples from magnetoresistance and Hall resistance (in agreement with Ref. 12). In other words the Seebeck voltage is by about a factor of 1000 larger than expected. This is visible most clearly in Fig. 2, where we plot $y = \arctan(S_{\perp}/QB)$ versus temperature for $B=0.5$ and 5 T. In the superconducting state for dissipation due to flux-line motion this quantity should be equal to the Hall angle, that is, should be of the order 0.1°. In contrast we find that $y \approx 40-70°$ over a wide temperature range below T_{c0}. Note that also the field and temperature dependence of the Hall angle is completely different to that observed from magnetoresistance and Hall effect.[6,12] We mention that for $T > T_c$ $90° - y = \alpha \approx 0.1°$ as expected, i.e., the Nernst voltage becomes much smaller than the Seebeck voltage.) We emphasize that $\tan\alpha \approx 1$ corresponds to a net directed

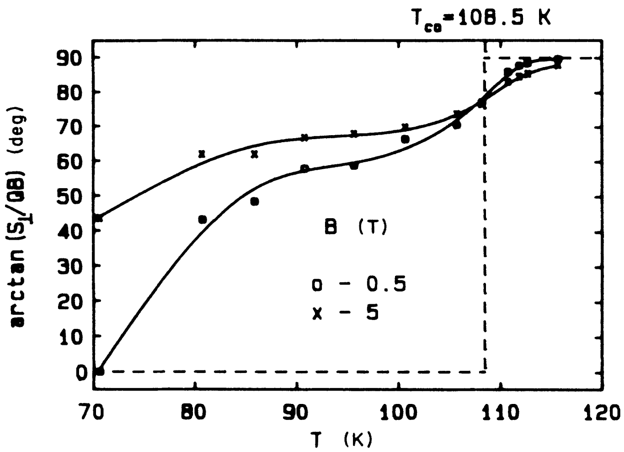

FIG. 2. $y=\arctan(S_\perp/QB)$ as a function of temperature for $B=0.5$ and 5 T. The zero-field transition temperature, T_{c0}, is indicated. Note that for $T>T_c$ $90°-y=\alpha$ and for $T<T_c$ within a flux creep-flow model $y=\alpha$. The expected behavior of y is indicated by the dotted line and corresponds to $\alpha\approx 0.1°$ (see text).

average flux motion at an angle of 45° to ∇T, which obviously is not realistic.

(2) $S_\parallel$ is expected to be zero, since the driving force for flux-line motion is zero for $\nabla T \| \mathbf{B}$. Instead we find that S is nearly independent of the angle between $\mathbf{B}$ and ∇T. Note that this finding agrees well with what is observed for the broadening of the resistive transition in a magnetic field for polycrystals and also for single crystals.[1]

The preceding results in our view seriously question the conventional flux creep-flow models as the only source of dissipation. We mention that even by taking account of a very complex arrangement of flux lines (flux-line liquid, flux-line glass[13]) we do not know how to explain the aforementioned inconsistencies. Therefore, we are led to the conclusion that besides a flux flow-creep mechanism, which of course will determine part of the dissipation, another mechanism of equal importance may be present. The finding $S_\parallel \approx S_\perp$ (together with the corresponding observations on the magnetoresistance[1]) suggests that this mechanism should be independent of the direction between driving force and magnetic field. If we assume an additional such mechanism of dissipation (of for simplicity equal strength) and if we assume that it leads to transport properties proportional to the normal-state transport properties our results can be understood: In this case one must explain

$$\frac{BR_H}{\rho}=B\frac{R_{H,v}+R_{H,n}}{\rho_v+\rho_n}\approx 10^{-3} \text{ and } \frac{BQ}{S}=B\frac{Q_v+Q_n}{S_v+S_n}\approx 1 .$$

Here v and n stand for a vortex and a normal contribution to the voltages. The first equation simply states that the Hall angle is small, as expected (i.e., usually $R_{H,v}$ and $R_{H,n}$ are both much smaller than the corresponding resistive effects, ρ_v and ρ_n). From this, one concludes that because of the small Hall angle the vortex part of the Seebeck voltage S_v is also small and can be ignored. On the other hand Q_v is much larger than Q_n, again due to the small Hall angle, and thus Q_n can be ignored. Therefore one finds $BQ/S\approx BQ_v/S_n\approx 1$.

A possible mechanism known to account for dissipation is (bulk) fluctuations of the order parameter (e.g., Refs. 6 and 14 and references therein). However, the magnetic field would have to enhance these fluctuations by a few orders of magnitude and a mechanism for this is difficult to visualize: A reduction of the characteristic length scale of fluctuations by the magnetic field below that given by the coherence length $\xi(T)$ [$\xi(0)\approx 10-30$ Å (e.g., Ref. 6)] and a corresponding enhancement of the fluctuations as proposed in Ref. 15 will appear for the applied field larger than $\Phi_0/2\pi\xi^2(T)$ only (where Φ_0 is the flux quantum),[14]—i.e., for $(\Phi_0/2\pi B)^{1/2}<\xi(T)$. For $B=1$ T, $(\Phi_0/2\pi B)^{1/2}$ is about 1000 Å. This is large compared to the small coherence lengths $\xi(T)$ except within an interval of a few mK around T_c. Therefore we expect (bulk) fluctuations to be unimportant for temperatures sufficiently far below T_c.

In order to explain our results we shall in the following consider the granular models for the HTSC suggested earlier in Refs. 2 and 5. There is an obvious granularity on the scale of about 1 μm present for a ceramic sample, and an intragrain granularity may also be important.[5] In order to account for such granularity, as an oversimplified model for the HTSC one may use that of a disordered and complicated network of weak links characterized by a coupling energy $E_j=hI_c/2e$ (I_c: critical current) and by a typical spacing, d.[16] Depending on their values these parameters can describe intergranular weak links (grains) or intragranular weak links (due to structural defects[6]).

We note that a naive inspection of the broadening of, for example, the resistivity and Seebeck coefficient in a magnetic field immediately suggests such a model: The regions of strong superconductivity are unaffected by the field and so is the onset temperature of the transition. On the other hand, the magnetic field suppresses superconductivity in the weak links, which leads to a depression of the zero-resistance temperature.

The coupling energy $E_j=hI_c/2e$ will in general depend on temperature and magnetic field. For a granular model, E_j will have a distribution of values. If the absolute value of E_j is sufficiently small so that E_j becomes comparable to k_BT fluctuations of the order parameter over the weak links will serve as a mechanism of dissipation, which leads to transport properties proportional to the normal state properties (see above). This has been discussed in Ref. 17 for current driven dissipation. In the limit of small current one finds $R/R_n=[I_0(E_j/k_BT)]^{-2}$ where I_0 is the modified Bessel function. This result has been shown to account well for the magnetoresistive transition of the HTSC by Tinkham (however with E_j interpreted as a pinning energy).[16,18] The crucial point for such a mechanism of dissipation is certainly the absolute value of E_j and its dependence on the magnetic field, the latter being most important, since large dissipation is observed only for finite magnetic field. For an (random) array of weak links one expects a nonoscillatory falloff of

E_j with the applied field as soon as B becomes larger than the lower critical field B_{c1} of the weak links, typically of the order of 1 G (e.g., Refs. 16 and 19). This is confirmed from experiments and theory on critical currents of three-dimensional granular systems, which show that $E_j(B)/E_j(0) \sim B_{c1}/B$ for $B > B_{c1}$. Additionally, it is found there that I_c does not depend on the angle between the current and the magnetic field, which is attributed to the weak links having random orientation.[19]

Applying these results for the HTSC one arrives at the following picture: The reduction of E_j with B leads to large fluctuations of the phase of the superconducting wave function over internal weak links. The associated dissipation will only very weakly (via a pure geometrical effect) depend on the angle between the magnetic field and the driving forces for randomly oriented weak links and it will add to the flux flow-creep dissipative mechanism.

We finally point out that for the HTSC the experimental findings on dissipation in the presence of a magnetic field are qualitatively the same for polycrystals and for single crystals (except for anisotropy observed for the latter).[1,3] Especially, also for single crystals, inconsistencies with flux creep-flow mechanisms are found, for example, the magnetoresistance is independent of the angle between external current and magnetic field. These findings can be explained by the model presented here, which gives further indication that weak-link behavior may also be important for single crystals as discussed in Refs. 5 and 16. We mention that the dependence of the dissipation on the direction of the magnetic field with respect to the CuO planes as observed for single crystals[1] for a weak link mechanism as described here would be due to the anisotropy of the critical current I_c or, equivalently, to that of the coupling-energy E_j. Therefore it is related to the anisotropic normal state resistivities of the HTSC, since, for example, for a Josephson-type weak link E_j is given as $E_j \sim \Delta/R_n \tanh(\Delta/2k_B T)$, where R_n is the normal-state resistance of the weak link and Δ is the energy gap.[20]

In summary our measurements of the Magneto-Seebeck and the Nernst effect show that in addition to a flux flow-creep mechanism another mechanism of dissipation may be present. A possible such mechanism based on fluctuations of the order parameter over internal (inter and intrangular) weak links has been discussed.

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