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Seebeck effect in the mixed state of epitaxial $YBa_2Cu_3O_7$

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The temperature dependence of the Seebeck coefficient in the mixed state of a superconductor is closely related to the resistivity due to flux motion. This is a result of the counterflow of normal current and supercurrent in the presence of a temperature gradient, as discussed by Ginzburg. Measurements of the Seebeck coefficient performed with epitaxial c-axis-oriented $YBa_2Cu_3O_7$ films show excellent agreement with the theory.

In addition to the high critical temperature of the copper-oxide-based superconductors, another unique property of these materials is the broadening of the resistive transition in an applied magnetic field and the existence of the irreversibility line.[1-3] The latter represents a line in the phase space of temperature and magnetic field above which the magnetization becomes perfectly reversible and magnetic-flux pinning appears to be absent. Recently, for explaining this resistive and magnetic behavior, the mechanism of thermally assisted flux flow (TAFF) has been proposed.[4,5] This mechanism results from the small values of the pinning potential in combination with the high operating temperatures.

Similar to the resistive transition, in the mixed state of the high-T_c superconductors a broadening of the transition from the normal-state value to (almost) zero is also expected for the thermoelectric effects.[6,7] Again, in the broadening regime of these effects TAFF and magnetic-flux diffusion play a central role. In this paper, we deal specifically with this broadening of the transition for the Seebeck effect.

Recently, measurements of the Seebeck effect have been reported for the mixed state in polycrystalline[8] and single-crystalline[9] Y-Ba-Cu-O-based and in polycrystalline Bi-Sr-Ca-Cu-O-based materials,[10-12] showing the broadening effect mentioned above. The appearance of the Seebeck effect in the mixed state can be qualitatively explained in terms of the counterflow of a quasiparticle current and a supercurrent in the presence of a temperature gradient. As first pointed out by Ginzburg,[13] in a superconductor the diffusion of quasiparticles in a temperature gradient is not compensated by the electric field generated by the resulting space charges as in a normal conductor. Instead, the compensation is due to a supercurrent flowing in the opposite direction as the diffusive quasiparticle current. As discussed in more detail elsewhere,[14] in the mixed state it is only the supercurrent which interacts with the spatially varying phase of the superconducting wave function, causing a temporal phase change and a voltage of quantum-mechanical origin governed by the Josephson relation. Based on this counterflow model,[13,14] the following expression for the temperature-dependent Seebeck coefficient $S(T)$ in the mixed state has been derived[6]

$$S(T)=\frac{\rho_{\mathrm{fl}}(T)}{\rho_n(T)}S_n(T)\,. \tag{1}$$

Here, ρ_{fl} and ρ_n denote the electric resistivity in the flux-motion state and in the normal state, respectively, and S_n the Seebeck coefficient in the normal state. From Eq. (1) we see that the broadening of the transition regime of the Seebeck effect in a magnetic field is closely related to the broadening of the resistive transition. We recall that the Seebeck coefficient is defined as the ratio of the electric field E in the direction of the temperature gradient ∇T and ∇T: $S=E/\nabla T$. In this paper, we present experimental results which quantitatively confirm the prediction of Eq. (1).

Before turning to the experiments, we point out that Eq. (1) has been derived by Caroli and Maki[15] from the time-dependent Ginzburg-Landau theory. It can also be obtained from the following simple heuristic argument. We start from the kinetic equation for the quasiparticle current density j_n,[16]

$$-j_n=\frac{1}{\rho_n e}\nabla\mu+\frac{S_n}{\rho_n}\nabla T\,, \tag{2}$$

where e is the elementary charge and μ the electrochemical potential. For a superconductor in the configuration for the Seebeck effect with zero total electric transport current, according to the counterflow concept of Ginzburg,[13] in a temperature gradient the supercurrent density $j_s=-j_n$ is generated. Taking the chemical part of the electrochemical potential as constant, the electric field E is given by $E=-\nabla\mu/e$, and we obtain

$$j_s=-j_n=-(1/\rho_n)E+(S_n/\rho_n)\nabla T\,. \tag{3}$$

Due to its interaction with the vortex structure in the mixed state, the supercurrent density j_s results in the electric field E due to flux motion, and we can write $E=\rho_s\cdot j_s$. We finally obtain

$$(1/\rho_s+1/\rho_n)E=(S_n/\rho_n)\nabla T\,. \tag{4}$$

Noting that $(1/\rho_s + 1/\rho_n) = 1/\rho_{fl}$, we find the Seebeck coefficient in the mixed state $S = E/\nabla T = (\rho_{fl}/\rho_n) S_n$, i.e., the result of Eq. (1).

The data reported in the following were obtained using a c-axis-oriented epitaxial film of $YBa_2Cu_3O_7$ prepared on a (100) $SrTiO_3$ substrate by laser ablation deposition.[17] The substrate size was 10 mm × 10 mm × 0.5 mm, and the film thickness was 400 nm. Since the thickness of the substrate is about one thousand times larger than that of the film, the temperature gradient in the superconducting film is predominantly determined by the temperature gradient in the substrate. The film originally covering the whole substrate was patterned by standard photolithography, yielding the geometry shown in Fig. 1. The horizontal strip A running along the direction of the applied temperature gradient served for measuring the Seebeck effect. The two vertical strips B and C (connected with strip A) were used for studying the Nernst effect, and the results of the Nernst experiments will be published elsewhere.[18] For the present studies of the Seebeck effect, the vertical strips B and C served for accurately determining the temperature gradient along strip A using the temperature-dependent resistivities of strips B and C for thermometry.

The width of strips A, B, and C was 100 μm. The distance between strips B and C was 3.0 mm. The contact pads indicated in Fig. 1 by the black areas also consisted of the $YBa_2Cu_3O_7$ film upon which a Ag film of 50-nm thickness has been deposited. All leads were 0.08-mm-diam copper wire attached to the contact pads for all three strips in a four-point configuration using indium press contacts. For applying the temperature gradient along strip A, the free surface of the substrate was glued to two gold-plated copper blocks at both ends on the left and the right using high thermal conductivity Stycast. The overlap between both ends of the substrate and the copper blocks was 2.5 mm each, such that the gap between the copper blocks was 5.0 mm wide. For applying the temperature gradient, the temperature of both blocks could be

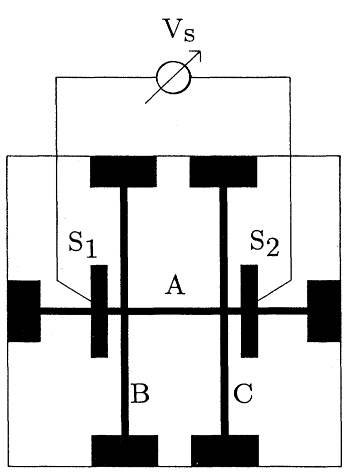

FIG. 1. Schematic diagram of the Y-Ba-Cu-O sample film on the $SrTiO_3$ substrate: Strips B and C are used to determine the temperature gradient. The measurements of Seebeck coefficient and resistivity have been performed with strip A.

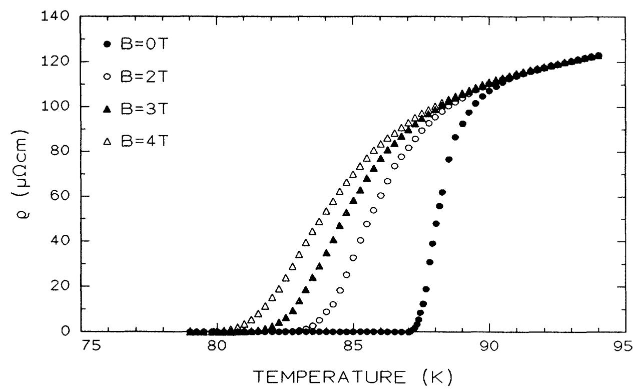

FIG. 2. Resistivity vs temperature of strip A with a zero-temperature gradient for various magnetic fields.

individually controlled within 5 mK using an arrangement of bifilarly wound heaters, temperature sensors, and electronic regulators. The copper leads for measuring the Seebeck effect in strip A were attached to the contact pads marked S_1 and S_2 in Fig. 1. These pads were 4 mm apart, each being placed 0.5 mm away from the corresponding crossing point between strip A and strips B or C. In this way, the temperature at the hot and cold ends of the $Cu/YBa_2Cu_3O_7$ thermocouple could be obtained with reasonable accuracy by linearly extrapolating the temperature gradient derived from the resistive measurements and the locations of strips B and C. A more detailed description of the thermometry technique utilizing the temperature-dependent resistivities in strips B and C will be given elsewhere.[18]

During the experiments, the sample holder was surrounded by a stainless-steel vacuum can and inserted into liquid helium. A superconducting magnet served for generating the applied magnetic field in the direction perpendicular to the film plane.

The temperature-dependent resistivity of strip A is shown in Fig. 2 for different magnetic fields up to 4 T. Figure 3 shows the thermopower S_{TC} of the $Cu/YBa_2Cu_3O_7$ thermocouple as a function of temperature at zero magnetic field and at 4 T and the difference between both curves. The thermopower S_{TC} is given by

$$S_{TC} = S_{Cu} - S_{Y\text{-}Ba\text{-}Cu\text{-}O}, \tag{5}$$

where the quantities on the right-hand side are the Seebeck coefficients of copper and Y-Ba-Cu-O, respectively. In the temperature range where Y-Ba-Cu-O becomes superconducting (see Fig. 2), $S_{Y\text{-}Ba\text{-}Cu\text{-}O}$ vanishes and we have $S_{TC} = S_{Cu}$. From Fig. 3, the broadening of the transition of $S_{Y\text{-}Ba\text{-}Cu\text{-}O}$ from its normal-state value to zero due to the applied magnetic field can clearly be seen. This broadening effect and its quantitative comparison with Eq. (1) can best be seen by looking at the difference $S_{TC}(T,B) - S_{TC}(T,0)$, also plotted in Fig. 3. Since the magnetic-field dependence of S_{Cu} is negligible in the range investigated, we have

$$S_{TC}(T,B) - S_{TC}(T,0) = S_{Y\text{-}Ba\text{-}Cu\text{-}O}(T,0) - S_{Y\text{-}Ba\text{-}Cu\text{-}O}(T,B). \tag{6}$$

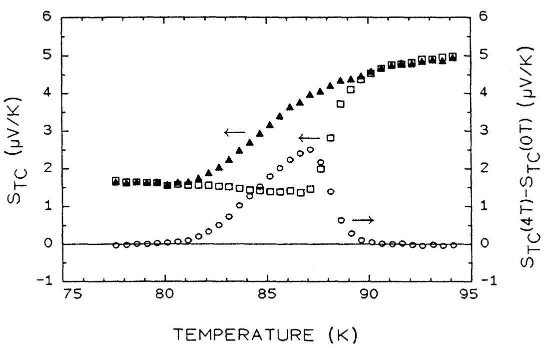

FIG. 3. Thermopower $S_{TC} = S_{\mathrm{Cu}} - S_{\mathrm{Y\text{-}Ba\text{-}Cu\text{-}O}}$ of the Cu/$YBa_2Cu_3O_7$/Cu thermocouple vs temperature for strip A for zero magnetic field (□) and for $B = 4$ T (▲) and the difference between both curves (○). The finite value in the superconducting state reflects the value of copper. All data were taken with an absolute temperature difference of 0.5 K between the contacts S_1 and S_2.

Inserting Eq. (1), we find

$$S_{TC}(T,B) - S_{TC}(T,0) = S_{n,\mathrm{Y\text{-}Ba\text{-}Cu\text{-}O}}(T)/\rho_n(T) \times [\rho_{\mathrm{fl}}(T,0) - \rho_{\mathrm{fl}}(T,B)] , \qquad (7)$$

where $S_{n,\mathrm{Y\text{-}Ba\text{-}Cu\text{-}O}}$ denotes the normal-state value of $S_{\mathrm{Y\text{-}Ba\text{-}Cu\text{-}O}}$. Again, in Eq. (7) we have neglected the magnetic-field dependence of the normal-state quantities $S_{n,\mathrm{Y\text{-}Ba\text{-}Cu\text{-}O}}(T)$ and $\rho_n(T)$. From (7) we see that the difference $S_{TC}(T,B) - S_{TC}(T,0)$ is essentially given by the difference in the corresponding resistive transition curves such as shown in Fig. 2.

In Fig. 4 we have plotted the experimental and theoretical values for the difference $S_{TC}(T,B) - S_{TC}(T,0)$ as a function of temperature for the magnetic fields of 2, 3, and 4 T. The theoretical values were calculated from Eq. (7) using the experimental resistive transition curves of Fig. 2. In this calculation we have taken the values of $\rho_n(90\ \mathrm{K})$ and $S_{n,\mathrm{Y\text{-}Ba\text{-}Cu\text{-}O}}(90\ \mathrm{K})$ from our experimental data in the following way: extrapolating the linear part of the $\rho(T)$ curve down to 90 K results in $\rho(90\ \mathrm{K}) = 120.5$ $\mu\Omega$ cm, and extrapolating the measured data of $S_{TC}(T,0)$ between 80 and 86 K up to 90 K yields the value $S_{\mathrm{Cu}}(90\ \mathrm{K}) = +1.25$ μV/K and with Eq. (5) $S_{n,\mathrm{Y\text{-}Ba\text{-}Cu\text{-}O}}(90\ \mathrm{K}) = -3.29$ μV/K. Furthermore, the temperature dependence of these two quantities has been neglected over the relatively small temperature range. As seen from Fig. 4, the experimental and theoretical values are in excellent agreement, confirming the validity of Eqs. (1) and (7).

Our measured value $S_{n,\mathrm{Y\text{-}Ba\text{-}Cu\text{-}O}}(90\ \mathrm{K}) = -3.29$ μV/K appears to be in good agreement with the published data.[9,19,20] Finally, we note that in our discussion we have concentrated only on the contribution to the Seebeck effect caused by flux motion and we have left out all other much smaller and more subtle effects. A discussion of these additional effects can be found in a review by Van Harlingen.[21]

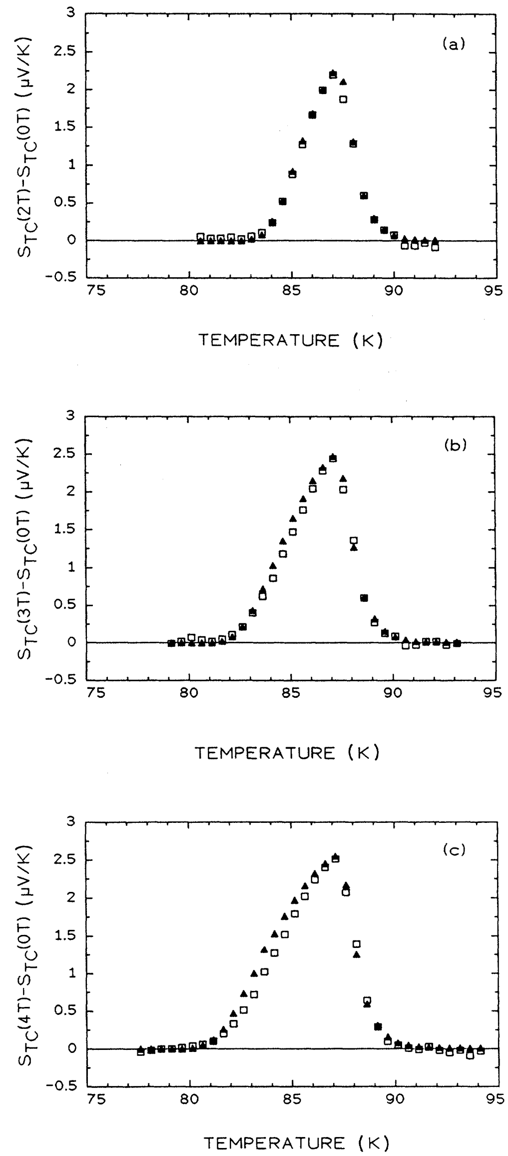

FIG. 4. Difference $S_{TC}(T,B) - S_{TC}(T,0)$ of the thermopower of the Cu/$YBa_2Cu_3O_7$/Cu thermocouple (where $S_{TC} = S_{\mathrm{Cu}} - S_{\mathrm{Y\text{-}Ba\text{-}Cu\text{-}O}}$) for magnetic field B and for $B = 0$ vs temperature (□) and the theoretical values calculated from Eq. (7) (▲) for three magnetic fields. (a) $B = 2$ T, (b) $B = 3$ T, (c) $B = 4$ T.

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[1]K. A. Müller, M. Takashige, and J. G. Bednorz, Phys. Rev. Lett. **58**, 1143 (1987).
[2]Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).
[3]M. Tinkham, Phys. Rev. Lett. **61**, 1658 (1988).
[4]D. Dew-Hughes, Cryogenics **28**, 674 (1988).
[5]P. H. Kes, J. van den Berg, C. J. van der Beck, and J. A. Mydosh, Supercond. Sci. Technol. **1**, 242 (1989).
[6]R. P. Huebener, Physica C **168**, 605 (1990).
[7]R. P. Huebener (unpublished).
[8]Ch. Laurent, S. K. Patapis, M. Laguesse, H. W. Vanderschueren, A. Rulmont, P. Tarte, and M. Ausloos, Solid State Commun. **66**, 4454 (1988).
[9]M. A. Howson, M. B. Salamon, T. A. Friedmann, J. P. Rice, and D. Ginsberg, Phys. Rev. B **41**, 300 (1990).
[10]V. V. Gridin, P. Pernambuco-Wise, C. G. Trendall, W. R. Datars, and J. D. Garrett, Phys. Rev. B **40**, 8814 (1989).
[11]V. V. Gridin, P. Pernambuco-Wise, C. G. Trendall, W. R. Datars, and J. D. Garrett, Solid State Commun. **74**, 187 (1990).
[12]M. Galffy, A. Freimuth, and U. Murek, Phys. Rev. B **41**, 11 029 (1990).
[13]V. L. Ginzburg, Zh. Eksp. Teor. Fiz. **14**, 177 (1944) [J. Phys. USSR **8**, 148 (1944)].
[14]R. P. Huebener, A. V. Ustinov, and V. K. Kaplunenko, Phys. Rev. B **42**, 4831 (1990).
[15]C. Caroli and K. Maki, Phys. Rev. **164**, 591 (1967).
[16]H. B. Callen, *Thermodynamics* (Wiley, New York, 1960).
[17]G. Koren, A. Gupta, E. A. Giess, A. Segmüller, and R. B. Laibowitz, Appl. Phys. Lett. **54**, 1054 (1988).
[18]F. Kober, H.-C. Ri, R. Gross, R. P. Huebener, and A. Gupta (unpublished).
[19]P. J. Ouseph and M. Ray O'Bryan, Phys. Rev. B **41**, 4123 (1990).
[20]G. Uher and W.-N. Huang, Phys. Rev. B **40**, 2694 (1989).
[21]D. J. Van Harlingen, Physica B **109 & 110**, 1710 (1982).