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Magneto-Seebeck and Nernst-effect measurements on $YBa_2Cu_3O_{7-x}$ single crystals

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We report on a detailed investigation of the magneto-Seebeck and Nernst effects in $YBa_2Cu_3O_{7-x}$ single crystals. Measurements were performed by means of a sensitive ac technique for fields up to 5 T and temperatures varying from 70 to 100 K. Our experimental results are consistent with those obtained in epitaxial films. The data are interpreted using Caroli and Maki's microscopic theory. The transport line energy U_ϕ carried by each vortex is possibly dependent upon oxygen concentration.

Voltages observed in type-II superconductors in the mixed state are usually attributed to flux flow. In fact, any physical perturbation that drives vortices into movement induces an electric field inside the bulk of the material.[1,2] For example, a temperature gradient applied to a superconducting sample in the mixed state produces a thermal force exerted on each vortex and directed from the hot side to the cold one.[3,4] If the flux lines are not pinned, this thermal force gives rise to flux flow. The high T_c superconducting oxides present a wide range of magnetic fields and temperatures in which the vortices are not pinned[5-7] (at high magnetic fields and for temperatures close to T_c, the magnetization becomes reversible). This situation has allowed many experimentalists to exploit the magnetothermal properties of these materials to characterize flux flow.[8-16]

This paper involves a study of flux flow through a detailed investigation of the magneto-Seebeck and Nernst effects in an $YBa_2Cu_3O_{7-x}$ single crystal. Such studies have been reported for films and sintered material where conventional dc techniques are suitable.[12-16] The thermoelectric power of $YBa_2Cu_3O_{7-x}$ is only approximately 1 $\mu V/K$. In addition, good-quality single crystals are small in size and can only generate small signals for reasonable temperature gradients. Thus a more sensitive ac technique had to be developed before proceeding with the measurements.

In this investigation we used a *microtwinned* single crystal of $YBa_2Cu_3O_{7-x}$ with dimensions $2\times2\times0.5$ mm^3. It had been obtained by an improved flux method in which a temperature gradient (4 °C/cm) is maintained during the growth. Using thoria crucibles allows us to obtain high-quality crystals with T_c's approaching 90–91 K after an oxygenation in flowing O_2 at 450 °C of only a few days. A more detailed description is given elsewhere.[17,18]

For the magneto-Seebeck and Nernst-effect measurements, as mentioned above, a sensitive ac technique was developed. An alternating temperature gradient of 1 Hz whose amplitude can be varied from 50 to 500 mK is applied to the sample. By means of this technique, the resolution is greatly increased over dc methods. This gradient is established by two heaters carrying square-wave pulses in opposite phase. The heaters are made of chromel wire and are wound around the two copper wires that support the sample (see Fig. 1 and Ref. 19 for more details). A differential chromel-gold thermocouple is used to measure the temperature gradient. Platinum paint is utilized to glue the sample to the copper wires and to make electrical contacts. The crystal is etched, before installation, in 1 % Br (by volume) in methanol for 30 min,[20] improving in this way the electrical contacts ($\sim 5\ \Omega$). The measurements are made with two lock-in amplifiers. A ramp in temperature is programmed and data are taken each 0.1 K between 70 and 100 K for applied magnetic fields parallel to the c axis, up to 5 T.

The experimental results presented in this paper were all obtained from the same sample with a temperature oscillation across the sample equal to 250 mK, peak to peak. The sample was oxygenated for three weeks to make sure of the oxygen homogeneity.

Figure 2 shows the Seebeck coefficient as a function of temperature for fixed magnetic fields up to 5 T. The original data correspond to the relative Seebeck effect, the reference being the lead wires and the platinum paint. This reference is assumed constant throughout the relevant temperature interval. Furthermore, far below T_c, the Seebeck coefficient associated with the sample is equal to zero. Accordingly, the data were shifted to reflect this fact. For $H=0$ T, the transition is very

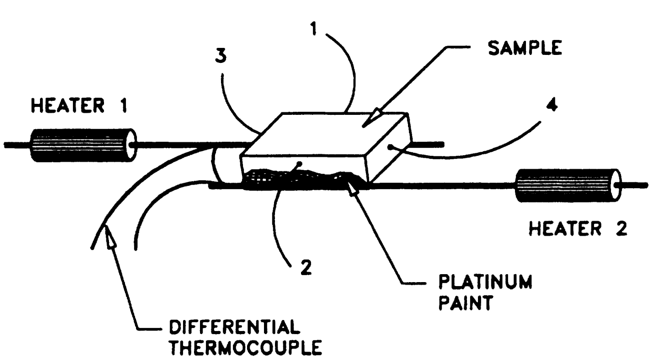

FIG. 1. Electrical connections to the sample. 1, 2 are used for the Seebeck effect and 3, 4 for the Nernst effect. H is applied parallel to the c axis.

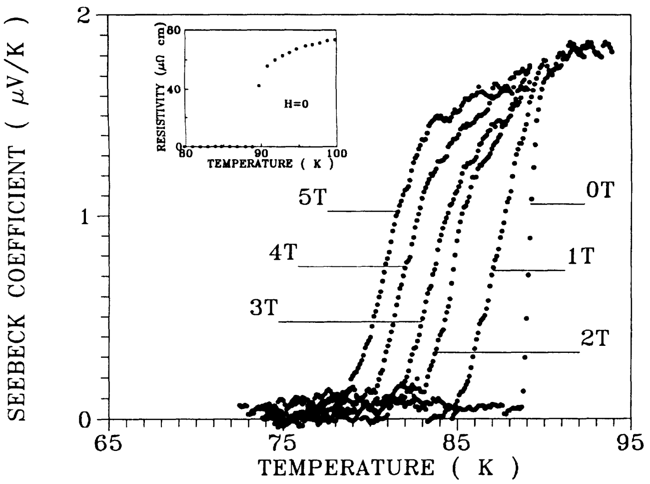

FIG. 2. Seebeck coefficient of a $YBa_2Cu_3O_{7-x}$ single crystal vs temperature for the indicated magnetic fields. Inset: the electrical resistivity as a function of temperature under zero applied magnetic field.

sharp. It begins at 90 K and terminates at 89 K, indicating the good quality of the crystal. The transition broadens with increasing magnetic field. The inset shows the resistivity versus temperature at $H=0$ T, measured in a van der Pauw geometry. T_c deduced from the resistivity is the same as the one obtained from the Seebeck curve at $H=0$ T.

Figure 3 shows the measured Nernst effect versus temperature for fields up to 5 T. For temperatures just below T_c, the Nernst voltage increases with decreasing temperature up to a maximum and then decreases reaching zero far below T_c. As one increases the magnetic field, the peak increases in magnitude, becomes broader, and shifts to lower temperatures.

We have already mentioned that voltages in the mixed state are commonly attributed to the motion of vortex lines[1,2] or, in a microscopic description,[21-24] to the moving order parameter Δ. Vortex pinning thus leads to a decreasing Nernst voltage with decreasing temperature on the left side of the peak in Fig. 3 and to vanishing magneto-Seebeck and Nernst voltages far below T_c. In other measurements[5-7] it has been shown that close to T_c and at high fields, the magnetization becomes reversible. This means that vortices are no longer pinned under these conditions. Pinning forces are overcome by means of thermal activation (pinning forces are of the order of thermal energies[25,26]) and the repulsive force between vortices that increases with increasing magnetic field (this force varies as $\sim\log(\lambda/d)$ when $d \ll \lambda$, for d equal to distance between vortices and λ equal to penetration length).[27] In this field and temperature range, a thermal force, induced by even a small temperature gradient, gives rise to the flux flow regime.

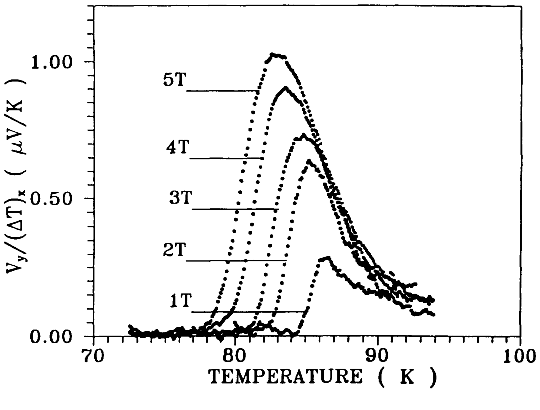

FIG. 3. Nernst voltage of a $YBa_2Cu_3O_{7-x}$ single crystal vs temperature for the indicated magnetic fields.

Using Caroli and Maki's theory developed in the framework of the time-dependent Ginsburg-Landau theory, one obtained the following expressions, valid near T_c, for the magneto-Seebeck coefficient and Nernst effect:[21-24]

$$S_s = S_n \frac{\rho_f}{\rho_n} , \tag{1}$$

$$\frac{E_y}{\nabla_x T} = -\frac{\rho_f S_\phi(T)}{\phi_0} , \tag{2}$$

where ρ_n and ρ_f are the normal and flux flow resistivities, respectively, and $\phi_0 = h/2e$ is the flux quantum. S_ϕ is the diffusion entropy and is given in SI units by

$$S_\phi(T) = \frac{\phi_0}{\mu_0 T} \frac{[H_{c2}(t) - H] L_\phi(t)}{1.16[2\kappa^2(t) - 1] + 1} , \tag{3}$$

where κ is the Ginsburg-Landau parameter, $\mu_0 = 4\pi \times 10^{-7} H/m$, t is the reduced temperature T/T_c and $L_\phi(t)$ is a constant close to one.

As the crystal used is square, one can write $|E_y/\nabla_x T| = V_y/(\Delta T)_x$. Combining Eqs. (1) and (2) and using the experimental results of Figs. 2 and 3, one can obtain the transport line energy $U_\phi = TS_\phi$ (ρ_n and S_n are taken to be equal to 7×10^{-7} Ωm and 1.8×10^{-6} V/K, respectively, according to Fig. 2).

Figure 4 presents the calculated transport line energy U_ϕ as a function of temperature T. As expected from theory, U_ϕ varies linearly with temperature near T_c. Our values of U_ϕ have the same order of magnitude as those determined from well oxygenated $YBa_2Cu_3O_{7-x}$ (YBCO) epitaxial films.[16] However, they are tenfold smaller than those determined from YBCO single crystals by means of Ettingshausen measurements.[28] These authors predicted a Nernst effect approximately ten times higher than that obtained experimentally in our case. It is worth noting that the slope $dU_\phi/dT \approx -0.67\times10^{-13}$ J/K m for $H=3$ T as deduced from Fig. 4, is between that determined, at the same field, for slightly oxygen deficient YBCO epitaxial films ($\sim -0.14\times10^{-13}$ J/K m) (Ref. 15) and that deduced for well oxygenated epitaxial films ($\sim -2.2\times10^{-13}$ J/K m).[16] It has already been reported that the Seebeck coefficient in the normal state goes from positive values to negative ones as the oxygenation of the sample progresses.[29,30] Our results are thus consistent with other Seebeck measurements in the normal state since we have

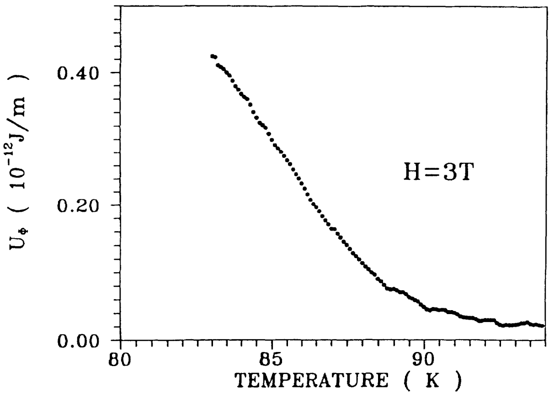

FIG. 4. Transport line energy $U_\phi = TS_\phi$ vs temperature for $H = 3$ T.

1.8×10^{-6} V/K for our crystal compared to 9×10^{-6} V/K for the oxygen deficient film and -3.3×10^{-6} V/K for the well oxygenated film. It seems then that the transport line energy depends on the oxygen concentration. This idea is further supported by the fact that here U_ϕ is deduced from magneto-Seebeck measurements instead of magnetoresistivity, avoiding in this way the problem of contributions other than that coming from Abrikosov vortices flow such as those due to Josephson vortices flow.[15,16] On the other hand, S_ϕ and hence U_ϕ are transported by excitations localized about the vortices.[28,31] The density of these bound states is proportional to the density of normal electron states at the Fermi surface.[32] The nature of this dependence is not clear in $YBa_2Cu_3O_{7-x}$ since in this case the dominant carrier species changes[30] from electronlike to holelike with increasing x. Nevertheless, it seems that the increase of U_ϕ with oxygen content may be associated with the increase of the localized excitations.

Using the expression of the thermal force exerted on a single vortex, given by Palstra *et al*:[28] $f_{th} = (\partial U_\phi / \partial T)\nabla T$, we obtain approximately 6.7×10^{-12} N/m for a temperature gradient of 10^2 K/m. This corresponds to the Lorentz force produced by a current density of the order of 0.34×10^4 A/m^2 comparatively to the case of conventional superconductors[3,31] ($\sim5\times10^4$ A/m^2). As the oxygenation of the YBCO samples progresses, a tendency towards the latter value is observed (a value of 10^4 A/m^2 is deduced from data of well oxygenated films[16]). This force is vanishingly small compared to pinning forces ($\sim10^{-5}$ N/m at 4.2 K),[33] as mentioned above. By extrapolating $U_\phi(T,H)$ to zero one can obtain either $T_c(H)$ or $H_{c2}(T)$ and then deduce the slope[28] dH_{c2}/dT. In our case we deal with variations of $T_c(H)$ of the order of 0.1 K or less leading to only a rough estimation of the slope which is approximately equal to $dH_{c2}/dT \approx -10\pm3$ T/K. This value is comparable to that deduced by Palstra *et al.*[28] Using experimental values of dH_{c2}/dT, dU_ϕ/dT, and Eq. (3) yields the Ginzburg-Landau parameter $\kappa \approx 10^2$.

In summary, following our development of a sensitive ac technique, we have succeeded in measuring magneto-Seebeck and Nernst voltages of small crystals such as those of $YBa_2Cu_3O_{7-x}$. The results presented are comparable to those obtained in epitaxial films. The magnitude of the transport line energy U_ϕ is shown to be probably dependent upon oxygen concentration.

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