Effect of superimposed electrical and thermal gradients in superconducting polycrystalline Bi-Pb-Sr-Ca-Cu-O

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We have studied thermoelectric effects in the presence of a superimposed transport current on a bar-shaped sample of polycrystalline Bi-Pb-Sr-Ca-Cu-O material with a superconducting transition at $\simeq 109$ K. The electrical current I_{ex} was applied parallel and then antiparallel to the temperature gradient $\nabla_x T$, which was directed along the longest sample axis X. We have detected asymmetry in the temperature dependence of the longitudinal voltage drop as a result of the current direction with respect to $\nabla_x T$. The result is interpreted in terms of the recently reported observation of the analog of the Fountain effect in an Y-Ba-Cu-O film [Europhys. Lett. **13**, 175 (1990)]. Our study supports the importance of weak-link-related effects in the thermoelectricity of high-T_c cuprates.

I. INTRODUCTION

Thermoelectricity in high-T_c materials has attracted significant research interest in the few years since its discovery in 1987. In particular, the Seebeck effect has been widely studied on various cuprates.[1-12] It is recently been pointed out by Ginzburg,[13] Huebener, Ustinov, and Kaplunenko[14] and Ustinov *et al.*[15] that the weak-link nature of these cuprates, coupled with the unusual range of parameter values, should produce interesting thermoelectric effects. Such effects were originally proposed by Ginzburg in 1944 (Ref. 16) and have subsequently been thoroughly discussed by Van Harlingen,[17] Ginzburg and Zharkov,[18] Kaplunenko, Ryazanov, and Shmidt[19] and many other researchers, but are generally small in conventional superconducting systems. For a review of the influence of Josephson junctions and weak-link structures on thermoelectricity in superconductors, the reader is referred to Refs. 17–19 and the literature cited therein.

Ustinov *et al.*[15,20] have recently reported an observation of the analog of the Fountain effect in superconducting Y-Ba-Cu-O. The authors applied a temperature gradient to a microbridge-shaped thick film of Y-Ba-Cu-O and observed asymmetry in the *I-V* characteristics as a function of the mutual orientation of the external electrical current density and the temperature gradient applied across the sample. This effect was originally reported by Clarke and Freake in 1972.[21] A nonzero voltage is detected when the total current density, $j_t = j_{ex} \pm j_s$, exceeds the temperature-dependent critical density, j_c. Here, j_{ex} is the external current density and j_s is the supercurrent that flows due to the temperature gradient applied across the weak-link array,[13,17,18] $j_s = S\nabla_x T/\rho_n$. S is the thermoelectric power (Seebeck effect) and ρ_n is the weak-link resistivity.

Here we report on a study of parallel and antiparallel current configurations with respect to the thermal gradient established across a bar-shaped sample of polycrystalline Bi-Pb-Sr-Ca-Cu-O material. It was of interest to check whether the effects observed by Ustinov *et al.*[15,20] would be apparent in this material, in which the nature of the weak links is rather different from Y-Ba-Cu-O. The sample was specially thermally tailored to produce degraded weak-link properties (thereby increasing the j_s component by raising the magnitude of the weak-link-related S) and low critical current densities. We were also interested in checking whether the channel complexity of the polycrystalline material ("intrinsic microbridges") would be sufficient for detecting the above-mentioned asymmetry in voltage signals. Therefore, we used a bar-shaped sample instead of the microbridge geometry studied by Ustinov *et al.*[15,20] Since $j_c \to 0$ as $T \to T_c$, we kept the magnitude of j_{ex} constant and studied the temperature dependence of the voltage drop across the sample, while flipping the current polarity with respect to the established temperature gradient. Our results support the recent findings of Ustinov *et al.*[15,20]

II. EXPERIMENTAL RESULTS AND DISCUSSION

We have studied a polycrystalline $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_2O_y$ material that has a critical temperature of superconducting transition around 109 K. The sample geometry is sketched in the inset of Fig. 1 where we also show the contact arrangement and the position of the sample with respect to the temperature gradient, $\nabla_x T$. The sample dimensions were $L_x \times L_y \times L_z = 13 \times 3 \times 0.3$ mm^3. The sample was clamped between two thermally massive, electrically isolated copper blocks with 100 Ω heaters wound onto each block. Two calibrated Lake Shore Pt temperature sensors were placed in good thermal contact with the sample and measured the temperatures indicated by T_c and T_h in Fig. 1, where the inset shows the sample configuration. A calibrated Lake Shore CGR-1500 was positioned between the Pt sensors and provided an independent check of the average sample temperature. The thermal stability was better than 40 mK for the duration of the measure-

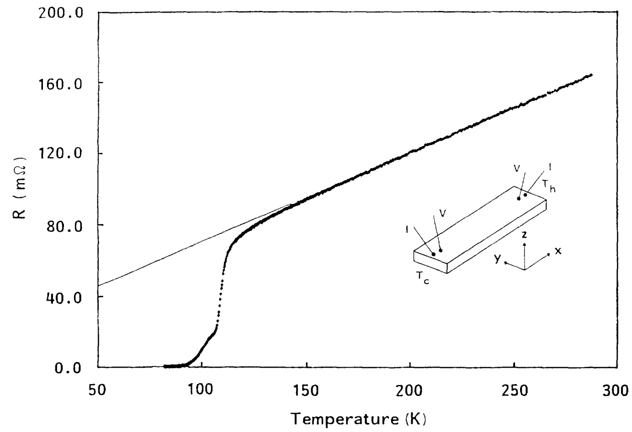

FIG. 1. Resistance $R=R(T)$ of the sample at an excitation current density of 0.1 A/cm^2. The inset shows the sample geometry and contact configuration used in this study. The straight line represents the normal resistance $R_n(T)$ extrapolated from $T>220$ K. T_c and T_h represent the temperatures of the cold and hot ends, respectively, of the sample as measured by two calibrated PT100 platinum thermometers.

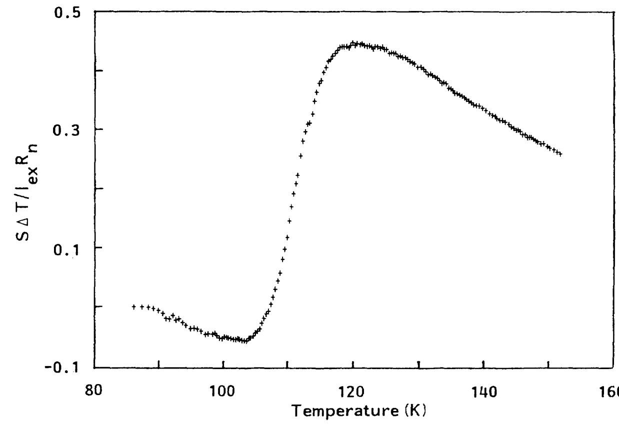

FIG. 2. The ratio of the thermoelectrically generated supercurrent ($I_s=S\Delta T/R_n$) to the external applied current (I_{ex}).

ment of each data point. The uncertainty of the thermal voltages was about 0.07 μV around 100 K and increased to 0.2 μV at room temperature. We used silver paste to make electrical contacts to the sample. These have proved their reliability during repeated thermal cycling in a similar experimental setup for the measurement of thermopower.[5] All contributions to the measured thermopower voltage which resulted from thermal mismatches at the wire connections were determined and subtracted from the data.

This batch of samples was deliberately reannealed at 840 °C in an oxygen-deficient atmosphere to reduce the superconducting quality of the sample for temperatures below 105 K. A long resistive tail for $T<105$ K is evident in Fig. 1, where we present the temperature dependence of sample resistance, $R=R(T)$, measured with electrical current $I_{ex}=1$ mA. This tail indicates poorly connected channels for carrier transport in our material due to the large number of degraded weak links. This thermal treatment was chosen following the approach of Ustinov *et al.*,[15,20] who studied the Fountain effect in their Y-Ba-Cu-O film, and specifically noted the importance of a sufficiently large number of weak links with low critical current for their observations. Using the extrapolated data for R_n (given by the straight line in Fig. 1) and the zero-current Seebeck coefficient data, $S=S(T)$ measured on the same sample, we can estimate the order of magnitude of the I_s component of the current relative to the external component, $I_{ex}=1$ mA. This is shown in Fig. 2 where we plot

$$I_s/I_{ex}=S\Delta T/I_{ex}R_n \tag{1}$$

as a function of the sample temperature. The nonzero value of S for $T<109$ K is due to the sizable weak-link effects in this material. Small negative values for the Seebeck coefficient just below the transition have been reported previously[22,23] and are believed to be associated either with sample imperfections, poor oxygen content, or both. The value of the external transport current $I_{ex}=1$ mA was chosen to ensure that the thermoelectric voltages $S\Delta T$ and the resistive potential drops V_R were of the same order of magnitude in the region of interest between 80 and 155 K. This current corresponds to a current density of about 0.1 A/cm^2, which is very close to the current density in the region where Ustinov *et al.*[15,20] measured asymmetry in their I-V curves. It is clear that the sign of the V_R component follows the external current polarity whereas $S\Delta T$ is independent of I_{ex}. Equivalently, we define V_R as a positive quantity and then the normalized total voltage drop $V/\Delta T$ across the sample for the parallel and antiparallel current orientation relative to the unchanged direction of $\nabla_x T$ will result in $V/\Delta T=V_R/\Delta T+S$ and $V/\Delta T=V_R-S$, respectively. The temperature dependence of $V/\Delta T$ for these two cases is shown in Fig. 3 for $\Delta T\simeq 4$ K. $V_R/\Delta T+S$ and $V_R/\Delta T-S$ are denoted by the solid and dashed curves, respectively. It is apparent that the S and $V_R/\Delta T$ contri-

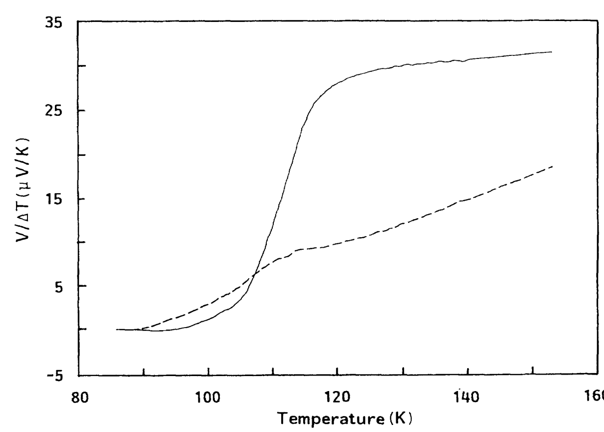

FIG. 3. The temperature dependence of $V_R/\Delta T+S$ and $V_R/\Delta T-S$. The former is represented by a solid line and the latter by the dashed line.

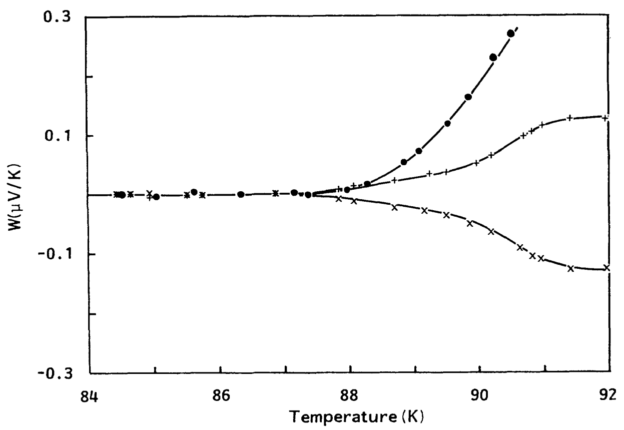

FIG. 4. The temperature dependence of $W_A \equiv V_R/\Delta T - S$, $W_B \equiv -V_R/\Delta T - S$, and $W_C \equiv V_R/\Delta T + S$ are labeled by ●, +, and ×, respectively. The solid lines are guides for the eye. Note that S starts to deviate from zero at $T \simeq 87.3$ K, whereas the resistive component $V_R/\Delta T$ only becomes nonzero at a higher temperature, $T \simeq 88.3$ K.

butions are nearly of the same magnitude. Since S is negative below $\simeq 108$ K, it results in a slight negativity of $V_R/\Delta T + S$ for $T \leq 95$ K. The region between 84 and 92 K is shown on a finer scale in Fig. 4 where we compare $W_A \equiv V_R/\Delta T - S$, $W_B \equiv -V_R/\Delta T - S$, and $W_C \equiv V_R/\Delta T + S$, which are presented by the upper, the middle, and the lower data sets, respectively. One observes that, to within the experimental uncertainty, W_B departs from W_A at $T \simeq 88.3$ K, whereas the departure of W_C from W_A occurs at about one degree lower, i.e., at $T \simeq 87.3$ K. This means that the thermoelectric component, S, is "switched on" approximately one degree earlier than the resistive contribution, $V_R/\Delta T$. Here we would like to stress that, in order to observe the Ustinov *et al.*[15,20] asymmetry of the *I-V* curves, there should be a "sizable" contribution of the thermoelectric term $S\Delta T$ to the total potential drop across the sample. It is seen in Fig. 4 that, under the chosen experimental conditions, the "thermoelectric tail" extends below the "resistive tail" for the chosen magnitude of external current (increasing I_{ex} produces an opposite pattern without affecting the asymmetry). The asymmetry between the W_A and W_C data sets, which is evident in Fig. 4, is the direct result of the thermoelectric contribution, which is believed to be related to the weak-link structure of high-T_c cuprates.[13–15,20] Our observation therefore supports the findings of Ustinov *et al.*[15,20] that the asymmetry in their *I-V* curves is due to the additional supercurrent density associated with the $S\nabla T$ term present in the weak-link array. Since the supercurrent due to the temperature gradient is intimately related to the phase difference across the boundaries in the sample, this experimental technique should be very useful for study of phase slippage and intergranular pinning effects in polycrystalline high-T_c materials. These effects are of prime importance to optimization of the transport properties of bulk superconductors.

III. SUMMARY

We have investigated polycrystalline Bi-Pb-Sr-Ca-Cu-O that has been thermally treated to enhance weak-link-related effects in its thermoelectric properties below the superconducting transition. Our study suggests a complementary approach to the recently reported findings of the Ustinov *et al.*[15,20] study in which the asymmetry in *I-V* characteristics was observed and was ascribed to superposition of the external current density and the additional supercurrents associated with the temperature difference across the weak-link array.

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