Magnetic field dependence of the thermoelectric power of superconducting Bi-Sr-Ca-Cu-O

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Measurements of the temperature and magnetic field dependence of the thermopower $S(B,T)$ are presented for a polycrystalline sample of the $T\sim85$ K phase of Bi-Sr-Ca-Cu-O. Between 90 and 140 K, $S(B,T)$ is positive and shows a small decrease with increasing temperature. In this temperature interval the application of an external magnetic field does not influence $S(B,T)$. Below 90 K, $S(0,T)$ drops sharply at the superconducting transition temperature. The magnetic field B causes a suppression of the transition temperature similar to the effect observed in resistivity data. The field data are interpreted in terms of the Abrikosov vortex lattice for the mixed state of the type-II superconductor. Relating the thermopower to the transport entropy of the vortex motion due to the temperature gradient across the sample, we find that the sign of the charge carriers is positive, and the principal unit $\hbar/2m$ of the particle flux is within 13% of its tabulated value.

I. INTRODUCTION

The transport properties of conventional type-II superconductors in the mixed state differ fundamentally from those in type-I superconductors. They exhibit a resistive behavior and characteristic thermomagnetic effects[1] that are relatively well explained in terms of vortex motion due to magnetic pressure and the influence of Lorentz force and pinning effects.[2-4]

With the discovery of high-temperature superconductors, much experimental and theoretical work has been done to explain the properties of these materials. Studies of the zero-field thermoelectric power in some high-T_c compounds have also been reported recently.[5] In this paper we present measurements of the temperature and magnetic field dependence of the thermoelectric power of the $T_c=85$ K phase of Bi-Sr-Ca-Cu-O. We explain the results within the previously established framework of flux motion in conventional low-T_c, type-II superconductors.

II. EXPERIMENTAL PROCEDURE

The sample used in this study was prepared from a mixture of Bi_2O_3, $SrCO_3$, $CaCuO_3$, and CuO in the molar ratio of Bi:Sr:Ca:Cu=2:2:1:3. After milling in methanol and drying, the reactants were melted together in a gold crucible at 1050 °C air, and then cooled to room temperature. The gold crucible was then replaced by a flat bottomed alumina combustion boat and the material was remelted and solidified over a 12 h period and then cooled to room temperature in 5 h. A rectangular sample with dimensions $15\times3\times0.25^3$, hereafter referred to as the X, Y, and Z axes, was cut from the ingot adjacent to the bottom of the boat so that a 1 mm thickness of aluminum adhered to the xy plane of the material, providing mechanical support. Analysis by backscattering electron microscopy (BSEM) and energy-dispersive analysis by x rays (EDAX) revealed that the sample was primarily of a single phase with little contamination either by additional phases or by aluminum reactants at the border with the substrate.

A mask was used to place six silver paste electrical contacts onto the surface of largest dimension in a conventional Hall-resistivity configuration, after which the sample was baked for 12 h at 300 °C. Copper wires were attached to these contacts with a further quantity of silver paste. High-purity lead wire, chosen because of its well-characterized thermopower,[6] was mechanically pressed onto the sample at either end of the XY surface to produce good electrical contacts. The ends of these wires were taken to a thermal anchoring point within the sample chamber and connected to the copper wires leading to the measuring device.

The sample was clamped between two electrically isolated brass blocks, which contained the heaters necessary for producing the thermal gradient along the X axis. The temperature was monitored at each end by a calibrated Pt resistance thermometer; each was accurate to better than 0.1 K in the temperature range $35\text{K}<T<300$ K. The thermometers were in thermal contact with the sample and the lead contact. Two vacuum jackets allowed thermal isolation from the liquid-helium cryostat and wide control of the average temperature of the sample.

In general, a typical data set was taken by warming the sample from 4.2 to 140 K with a temperature difference along the X axis ΔT of 1.5–2.5 K. Magnetic field measurements were taken with field applied along the Z axis of the sample. Since the thermometry below 35 K was deemed only roughly accurate, the data below 35 K were neglected. Contributions to the thermopower voltage from thermal mismatches at the wire connections and from the thermopower of the lead wires were accounted and subtracted from the data. In addition, the magnetoresistivity, Hall effect, and magnetization in the sample were measured. All of these quantities showed qualitative agreement with previous measurements in the literature,[7–12] and indicate that the sample had a superconducting transition temperature of approximately 85 K in zero field. There was no evidence of the $T_c=110$ K phase observed by other researchers.

III. RESULTS

Typical experimental data of the absolute thermoelectric power $S(B,T)$ of our sample are plotted in Fig. 1 as a function of temperature T for two values of magnetic field: $B=1.5$ and 0.0 T. The application of the field results in considerable broadening of the transition region compared to the zero-field curve and in the appearance of several shoulderlike features below $T=75$ K. The origin of these features, more pronounced in the lower field data although not present at zero field, is uncertain and will not be discussed further. Below $T=45$ K and above $T=95$ K the curves are coincident for all field values to within the reproducibility of our study. We note that our zero-field data show quite satisfactory agreement with the recently reported study of the Bi-Sr-Ca-Cu-O system.[13]

In order to more closely investigate the influence of the magnetic field on the thermopower,we present in Fig. 2 the quantity $\Delta S(B,T)$, defined by

$$\Delta S(B,T)=S(B,T)(-S(0,T) \ . \tag{1}$$

The temperature position of the main peak decreases slightly with increasing B. This results from the depression of the transition temperature by the magnetic field. This shift in the transition temperature is also observed in the resistivity.[8] Asymmetry of the line shape of $\Delta S(B,T)$ is also evident in Fig. 2.

In Fig. 3 we plot, as a function of the field strength B, the area enclosed between the $\Delta S(B,T)$ curves and the line $\Delta S(B,T)=0$ in Fig. 2. The area $A(B)$ is defined by

$$A(B)=\int_{T_1}^{T_2} S(B,T)dT \ , \tag{2}$$

where $T_1=40$ K and $T_2=95$ K. The data points are fitted to the linear expression

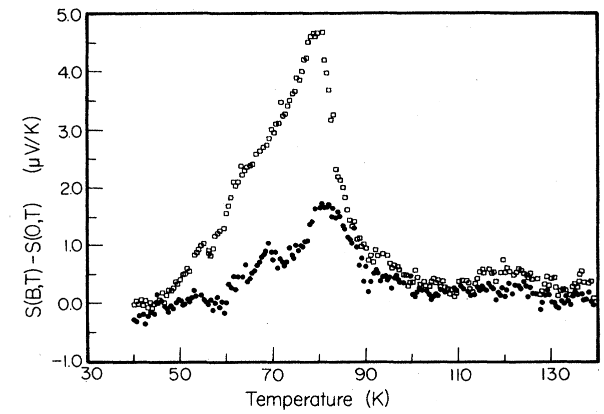

FIG. 2. Temperature dependence of the increment in the thermopower due to the application of the magnetic field [see Eq. (1) for definition]. □ and ● correspond to the $B=1.6$ T and $B=0.2$ T data, respectively.

$$A(B)=RB+D \ , \tag{3}$$

where $R=65\pm9$ (μV/T), $D=3\pm9$ (μV), and B is in T. Since D may be considered to be zero to within the experimental accuracy, there is a simple proportionality between $A(B)$ and the magnetic field B.

IV. DISCUSSION

It has been pointed out by Heubener and Seher[14] and discussed by a number of other authors[15] that the driving force acting on a vortex line due to a temperature gradient across the sample is related to the transport entropy carried by that line. This is usually attributed to the excitations present in the nonsuperconducting core of the vortex. The physical origin of this force results from the mutual repelling of the vortex lines, which are greater in number at the hot end than at the cold one. If we let σ be the transport entropy per unit volume of the sample, then the equilibrium condition for magnetic pressure is written as[2]

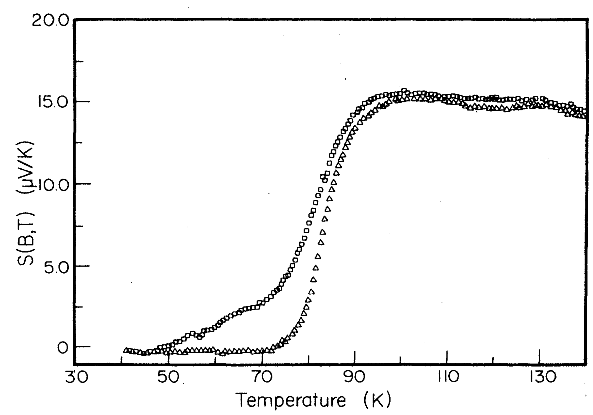

FIG. 1. Temperature dependence of the thermoelectric power $S(B,T)$ for $B=1.5$ T and $B=0.0$ T marked by □ and △, respectively.

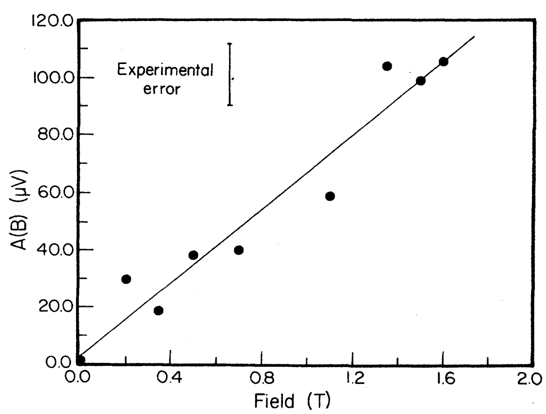

FIG. 3. Magnetic field dependence of the area enclosed by the curves $\Delta S(B,T)$ and $\Delta S(0,T)=0$.

$$\sigma \nabla T = -\frac{1}{4\pi} \mathbf{B} \nabla \mathbf{H} \ , \tag{4}$$

where $\mathbf{B}$ is the average magnetic induction and $\nabla\mathbf{H}$ is the gradient of the average local field strength in the sample with a temperature gradient ∇T. Assuming that the ∇T and $\nabla \mathbf{H}$ gradients are uniform across the sample we may write

$$\sigma = \frac{-1}{4\pi} \mathbf{B} \frac{\partial \mathbf{H}}{\partial T} \ . \tag{5}$$

It has been shown by Maki[16] and de Gennes[17] that in the dirty limit of a type-II superconductor Abrikosov's[18] description of the mixed state in terms of the vortex lattice can be extended to all values of the reduced temperature $t = T/t_c$, provided that the external field $B_0 \simeq H_0$ is much smaller than the zero-temperature value of the upper critical field $H_{c2}(0)$.[19] Since the coherence length of high-T_c samples is small,[20] and the upper critical field of these materials is much larger[21] than the fields used in our study, the Maki–de Gennes extension of the Ginzburg-Landau-Abrikosov-Gor'kov theory is assumed to hold thereafter.[22]

It is well known[23] that the magnetization $M = (B - H_0)/4\pi$ is expressed as

$$M = -(H_{c2} - H_0)/\{[2k_2^2(T) - 1]4\pi\} \ , \tag{6}$$

$$H_{c2} = k_1(T)\sqrt{2} H_c \ . \tag{7}$$

The values $k_1/k = k_2/k$ increase slightly[24] (from 1 to 1.2) when $t = T/T_c$ changes from 1 to 0, where k is the Ginzburg-Landau parameter, which is almost temperature independent.[18] An alternative form of the magnetization for the Abrikosov vortex lattice can be given in terms of the order parameter $\langle |\psi|^2 \rangle$ as[15,23]

$$M = (\mathbf{B} - H_0)/4\pi = \mathbf{H}/4\pi = -\frac{2\pi}{4\pi}(e\hbar/mc)\langle |\psi|^2 \rangle \ . \tag{8}$$

Here e and m are the values of the electronic charge and mass, respectively; $\langle \cdots \rangle$ denotes the space average of $|\psi|^2$ over the sample volume. Since $|\psi|^2 = n_S(r)$, where $n_S(r)$ is a local density of the electrons in the superconducting state, then $\langle |\psi|^2 \rangle$ represents the space average of $\langle n_S(r) \rangle = n_S$, which we write as $n_s = n\langle |\phi|^2 \rangle$; here $\langle |\phi|^2 \rangle$ is the normalized order parameter, which in the absence of the external field has the values of 1 and 0 at $T = 0$ K and $T = T_c$, respectively; n is the total density of conducting electrons in the material. The last two equalities on the right-hand side of Eq. (8), which describes the isothermal case, i.e., $\Delta T = 0$, enable us to express the partial derivative $\partial \mathbf{H}/\partial T$ in terms of $\partial \langle |\phi|^2 \rangle / \partial T$, when $\Delta T \neq 0$. Furthermore, for $H_0 \gg \mathbf{H}$, we substitute the approximate relation

$$H_0 \frac{\partial \mathbf{H}}{\partial T} = B_0 2\pi (e\hbar/mc) \partial \langle |\phi|^2 \rangle / \partial T$$

for $\mathbf{B}\partial\mathbf{H}/\partial T$ in Eq. (5) and obtain the expression for the transport entropy σ:

$$\sigma = B \left[e \frac{\hbar}{2m} c \right] n \partial \langle |\phi|^2 \rangle / \partial T \ . \tag{9}$$

Here the subscript "0" in the labeling of the external field $H_0 \simeq B_0$ is omitted. The absolute thermoelectric power of material S, is given as the ratio of the transport entropy density, σ at constant pressure, to the carriers charge density, qn, i.e., $S = \sigma/qn$.[25] (This result can be derived directly for small external fields in the proximity of H_{c2} by a phenomenological approach, given in Ref. 2, for a conventional superconductor with electron pairing and $q = e$.) Thus the absolute thermopower as a function of magnetic field and temperature is expressed:

$$S(B,T) = B(e\hbar/qm2c)\partial\langle |\phi|^2 \rangle / \partial T \ . \tag{10}$$

Finally, integrating Eq. (10) over the $T = (0, T_{c2})$ interval, and using the approximation that for the fields $B \ll H_{c2}(0)$, the value of the order parameter $\langle |\phi|^2 \rangle$ at $T = 0$ K is close to the zero-field value of 1 and that $\langle |\phi|^2 \rangle = 0$ at $T = T_{c2}$ we obtain the expression

$$\int_0^{T_{c2}} S(B,T)dT = -B(e\hbar/q2mc) \ . \tag{11}$$

For a superconductor $S(0,T)$ is essentially zero below T_c [see also Fig. 1 and Eq. (1)]. Therefore Eqs. (10) and (11) correspond to the observations of Figs. 2 and 3, respectively. We note that the sign of the charge q is opposite to that of the electron, since $\Delta S(B,T)$ is opposite in sign to that of lead used for calibration[6] and positive for all the temperatures measured; thus, $q = -e$ in Eqs. (10) and (11). Secondly, the slope $R = 65$ (μV/T) in Eq. (3) is predicted to be $\hbar/2mc$ by Eq. (11). The prediction is satisfied by $R/(\hbar/2m) = 1.13$ (when $c = 1$ and B is in T) to within experimental uncertainty. Thirdly, the thermopower in Fig. 2 reaches zero above and well below the transition, since above T_{c2} and below T_{c1} the transport entropy of the vortex motion goes to zero. It is noted, however, that with small values of $H_{c1}(0)$ comparative to H_0, $S(B,T)$ at low temperatures could be nonzero.

V. SUMMARY AND CONCLUSIONS

In this paper, we report measurements of the magnetic field dependence of the thermoelectric power in the high-transition-temperature type-II superconductor Bi-Sr-Ca-Cu-O (85 K phase). We present interpretation of data in terms of the Abrikosov vortex lattice of the mixed state of a type-II superconductor. The field and temperature dependence of the thermoelectric power are connected to the transport entropy associated with the vortex motion resulting from the thermal driving forces on the vortex when a temperature gradient is applied across the sample. The integral of the thermopower over the temperature range of the superconducting state predicts a linear magnetic field dependence that is observed in the study. This analysis enables us to determine the principal unit of the particle flux $\hbar/2m$ to within 13% of its tabulated value. The analysis required several simplifying assumptions, and therefore, a more rigorous treatment of the problems is welcome and encouraged.

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