Observation of vortex-lattice melting in $YBa_2Cu_3O_{7-\delta}$ by Seebeck-effect measurements

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Observation of vortex-lattice melting by Seebeck-effect measurements is reported. This technique does not require a transport current and may be related to the analogous resistivity measurements in the limit of zero current. The $YBa_2Cu_3O_{7-\delta}$ single crystals displayed a measurable effect in the b axis direction in the as-grown condition but virtually no signal in the a axis direction. The b axis data are combined with the results of Blatter and Ivlev to produce a phase diagram taking into account both thermal and quantum fluctuations. [S0163-1829(96)05738-4]

The transition between the normal state and the superconducting state in the presence of a magnetic field is richer in terms of physical phenomena in the high-T_c superconductors (HTSC) than in the conventional materials. For example, thermal fluctuations allows melting of the vortex lattice well below T_c.[1] Thus the resistive transition in untwinned single crystals of HTSC occurs in two steps: as the temperature is decreased, the resistivity decreases smoothly until it reaches a kink and then proceeds to decrease extremely rapidly to the zero resistance state, constituting a first order transition.[2–5] The latter is a manifestation of flux-lattice freezing or flux-lattice melting depending on the direction of the temperature sweep.

While the expression ''flux lattice'' is this regard implies long range order, such is not always the case. For example, in intense magnetic fields, the high density of vortices leads to their entanglement and a solidification into a glass state.[5] If the sample contains a few twin boundaries, the first order transition persists but the height of the kink varies with the orientation of the magnetic field[4] illustrating the corresponding variation of the effectiveness of twin-boundary pinning. On the other hand, the creation of a sufficient number of point defects in untwinned samples will lead to a second order transition.[4]

In the case of lattice melting, the resistivity above and below the kink has been shown to be respectively Ohmic and non-Ohmic.[4,5] Nonlinear I-V curves are also observed in the case of vortex glasses whose resistivity is often described by its value for currents tending to zero. All these measurements involve a variation of electrical current which must of course assume finite values. Is it possible then to observe lattice melting with essentially no transport current? One method would be the measurement of the Seebeck coefficient which in HTSC displays a transition similar to that of the resistive transition. In fact it was shown[6] that

$$S \approx \frac{S_n \rho}{\rho_n},$$

where S and ρ are the Seebeck coefficient and resistivity in the flux flow state and the subscript n refers to the normal state properties. Numerous articles have shown the similarity of the Seebeck and resistivity transitions in these materials.[6,7] Should lattice melting occur, however, the above relation will not be applicable quantitatively throughout the transition. Nevertheless, one would expect a behavior of the Seebeck coefficient analogous to that of the resistivity. Another method, a thermodynamic one, involving the measurement of the local magnetic field at the sample surface was applied by Zeldov *et al.*[8] A discontinuous change at the melting transition was observed in Bi-Sr-Ca-Cu-O single crystals.

A completely different approach was attempted by Gammel *et al.*[9] using a high-Q mechanical oscillator. The results were difficult to interpret, possibly due to the high frequency (2×10^3 Hz) involved and to the large pinning in the samples. Later, Beck *et al.*[10] resorted to a low frequency torsional-oscillator technique in an untwinned single crystal in which pinning is expected to be weaker.

In this paper we report on the observation of vortex-lattice melting by the Seebeck effect, i.e., with no applied current. In subsequent work, a comparison will be made between the effects along the a and b axes in order to separate the contributions from the chains and the planes. It will be shown, however, that the samples will require some treatment before this can be accomplished. Previous work concerning this separation has been done in the past[11–13] but in the absence of a magnetic field and therefore of vortices.

The growth of the $YBa_2Cu_3O_{7-\delta}$ (YBCO) single crystals involved in this work was done in a manner described elsewhere.[14] Sample 1 is a square platelet and is completely detwinned except for a very small region in one corner which is not expected to produce observable results. Sample 2 is rectangular, with the a axis on the longest side but contains several twin boundaries. Silver epoxy contacts were added to the ends of both samples and the Seebeck effect was measured as a function of temperature in the vicinity of T_c in magnetic fields up to 5 T parallel to the c axis.

Our measurement technique is an improved version of the one already described.[15] As illustrated in Fig. 1, the sample

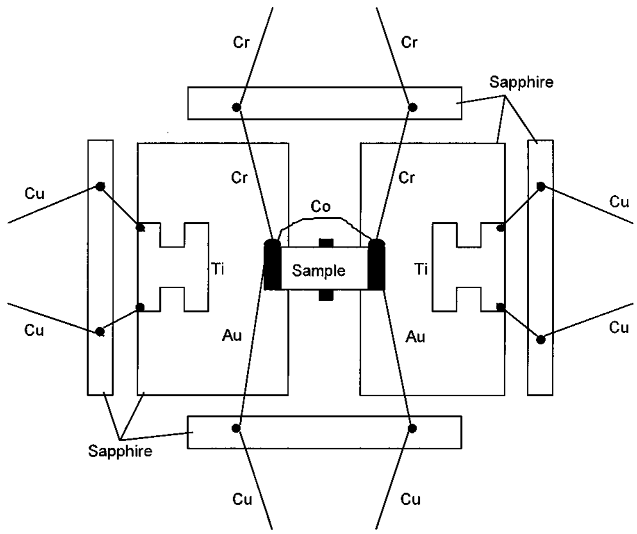

FIG. 1. Schematic of the sample mount for the Seebeck-effect measurements.

is now a bridge between two sapphire sheets which in turn are glued to a large copper block. Four smaller sapphire sheets act as temperature sinks. On the main sapphire sheet we evaporated Ti heaters through which square wave currents are circulated. The currents in the two heaters are out of phase by π so that ac signals are generated allowing detection by lock-in techniques. The Seebeck voltage ΔV is obtained through the gold wires attached to the ends of the sample whereas the temperature difference ΔT is obtained through the chromel constantan thermocouple junctions adjacent to the sample ends but isolated electrically by Stycast cement. We recall that the Seebeck coefficient is defined as the ratio $\Delta V/\Delta T$ in the limit $\Delta T \rightarrow 0$. The ac technique has a higher sensitivity and allows a smaller ΔT while maintaining a reasonable signal to noise ratio. This is particularly important in the case of YBCO whose Seebeck coefficient near T_c is only about 3 μV/K. Typically $\Delta T \sim 0.15$ K in the reported measurements. As one may gather from Fig. 1, we measure the Seebeck effect of YBCO with respect to gold. The measured ΔV is thus given by

$$\Delta V = \int_{T_1}^{T_1+\Delta T} (S_{\mathrm{Au}} - S)\, dT,$$

where S_{Au} represents the absolute Seebeck coefficient of gold and S that of the YBCO sample. If ΔT is sufficiently small, $(S_{\mathrm{Au}} - S)$ may be treated as constant over that interval and taken out of the integral so that one measures $(S_{\mathrm{Au}} - S)$. S_{Au} is sufficiently constant for all the magnetic fields considered here.[16] But such is not the case for S at T_c in the absence of a magnetic field. The intrinsically narrow transition as measured by S will appear broadened with a certain rounding at the beginning and end of the transition. However, a real broadening greater than ΔT is created by magnetic fields $\geqslant 0.5$ T so that our finite ΔT ceases to be a factor. Thus the narrow ΔT allowed by the ac technique along with the application of a magnetic field eliminates the need for involved calculations to compensate for the artificial rounding of the transition.[17] The absolute thermoelectric power of YBCO, S, is easily obtained since this effect is zero below T_c. We merely subtract the Seebeck voltage measured below T_c (due to S_{Au}) from all the Seebeck data of that particular run, using the fact that the Seebeck coefficient of gold does not vary over the range of the temperature sweep for the relevant magnetic fields.[16]

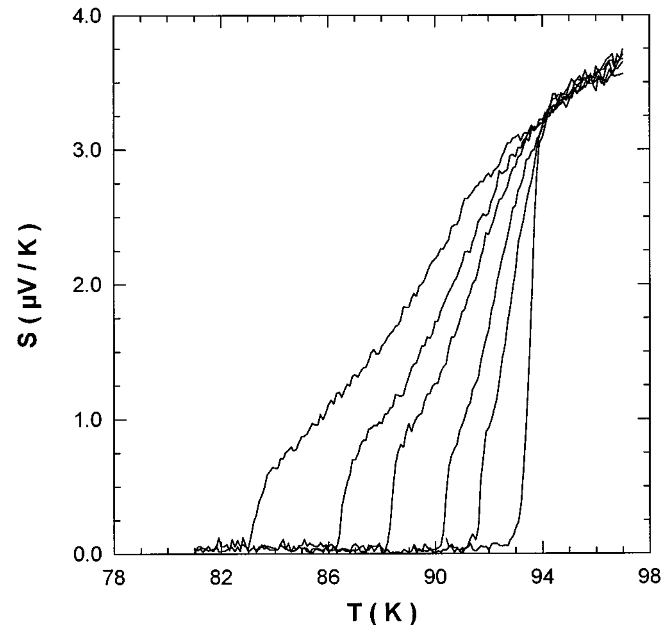

FIG. 2. Seebeck coefficient of sample 1 along the b axis as a function of temperature for magnetic fields of 0, 0.5, 1, 2, 3, and 5 T (from right to left).

The results presented here are limited to temperatures varying from 78 to 98 K but the apparatus can be used from liquid helium to room temperature. The temperature was ramped at a rate of 0.2 K/min, the detector being a carbon glass thermometer. The main improvement over the earlier version of our technique is in the mounting of the sample which allows more control over the orientation of the sample with respect to the magnetic field and of the temperature gradient with respect to the sample length. The two heaters are also more symmetric than in the previous version.

The Seebeck effect initially observed along the b axis of sample 1 in the presence of various magnetic fields is shown in Fig. 2. Instead of a smooth variation from zero to the normal state value as reported in the literature for twinned single crystals or for thin films, one observes a sharp rise and a kink followed by the smooth variation. The sharp rise corresponds to that of the resistivity and is attributed to vortex-lattice melting. This result was expected, following the remarks in the Introduction. However, the melting transition does not appear to be as sharp in the case of the Seebeck effect due to the finite temperature variation along the sample as explained above. Nevertheless, as in Ref. 4 the width of the melting transition at 2 T is narrower $[\Delta T_m(10\text{–}90\,\%) \sim 250$ mK$]$ than in zero field $[\Delta T_c(10\text{–}90\,\%) \sim 600$ mK$]$. At higher fields, however, ΔT_m increases with magnetic field as in the published resistivity data.

The melting process occurs in a finite temperature interval as already noted. To define the melting temperature T_m we use the maximum of the $\partial S/\partial T$ vs T curve. The same criterion applied at zero field gives $T_c = 93.55$ K. By combining the T_m values obtained at various magnetic fields, one may plot the melting field H_m as a function of T_m thus constituting a phase diagram. A power law of the form $(T_c - T_m)^n$ is obtained with an n similar to that reported by Kwok *et al.*[4] However, Blatter and Ivlev[18] stress that such a power law lacks any theoretical basis. To fill this gap, they present an analysis which describes the melting transition in terms of both thermal and quantum fluctuations. As usual, the melting criterion is that of Lindemann, which applies when the mean displacement amplitude $\langle u^2 \rangle^{1/2}$ of the vortices reaches a fraction $c_L < 1$ of the vortex lattice constant a_0. After appropriate simplifications Blatter and Ivlev obtain

$$\frac{\langle u^2 \rangle}{a_0^2} = \left[\frac{G}{\beta_{\mathrm{th}}}\right]^{1/2} \frac{\sqrt{b}}{1-t-b} \left\{ t + q\sqrt{b}\left(1 - \frac{b}{1-t}\right)\right\} = c_L^2,$$

similar to the earlier relation obtained by Houghton *et al.*[19] Here G is the Ginsburg number, β_{th} is a numerical parameter, $b = B/H_{c2}(0)$ is the scaled magnetic field, and $t = T/T_c$ is the scaled temperature. The suppression of the order parameter close to the upper critical field has been taken into account by the factor $[1 - b/(1-t)]$. Finally $q = 2.4 v/K_F \xi$, where $v = \omega_c \tau_r$ is the product of the cyclotron frequency and the relaxation time, K_F the Fermi wave vector, and ξ the in-plane coherence length. Neglecting the term of order b^2,

$$b_m = \frac{4\theta^2}{[1 + \sqrt{1 + 4S\theta/t}]^2},$$

where

$$\theta = c_L^2 \left(\frac{\beta_{\mathrm{th}}}{G}\right)^{1/2} \left(\frac{1-t}{t}\right) = \theta_0 \left(\frac{1-t}{t}\right)$$

and

$$S = q + c_L^2 \left(\frac{\beta_{\mathrm{th}}}{G}\right)^{1/2} = q + \theta_0.$$

Note that for $\theta \to 0$ (or $T \to T_c$), b_m varies as $(1-t)^2/t^2$, the thermal result. A fit of our data may be attempted with the above expressions by assuming literature values for $H_{c2}(0)$ in order to deduce values of θ_0 and q. Using $dH_{c2}/dT = -1.9$ T/K from Ref. 18 and our value of $T_c = 93.55$ K, $H_{c2}(0) = 177$ T which yields $\theta_0 = 3.6$ and $q = 5.4$. With $dH_{c2}/dT = -2.3$ T/K from Ref. 20, $H_{c2}(0) = 215$ T, $\theta_0 = 3.3$, and $q = 6.6$. Both sets of parameter values yield the same curve illustrated in Fig. 3. They are similar to those obtained by Blatter and Ivlev after fitting resistivity data to their theory.

As grown, sample 1 produced a very small *a* axis Seebeck effect providing no hope for a meaningful study as a function of magnetic field. It was then left under a partial vacuum for six months. It was found that the *a* axis Seebeck effect had become much larger. The *a* axis Seebeck effect of two other samples was also found to be very small in the as-grown condition. It therefore appears that a certain degree of deoxygenation is necessary to study the Seebeck effect along the *a* axis as a function of field, a phenomenon that is now under investigation.

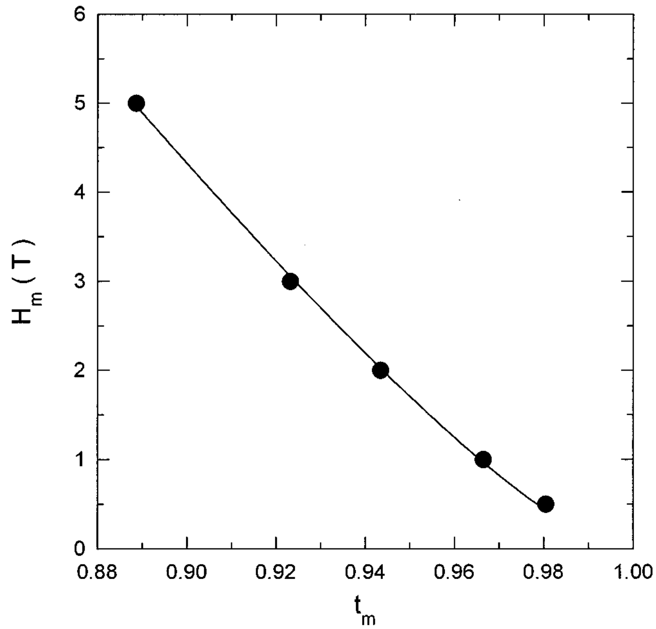

FIG. 3. Plot of H_m as a function of $t_m = T_m/T_c$. The circles are the experimental points and the curve represents the fit described in the text.

After the treatment just described, sample 1 was subjected to a new *b* axis Seebeck-effect study. This revealed an apparent deterioration of the sample, probably due to a loss of oxygen. Nevertheless this loss is too small to change T_c. According to Cohn *et al.*,[12] the electronic structure of $YBa_2Cu_3O_{7-\delta}$ is highly sensitive to the oxygen deficiency for $\delta \leqslant 0.2$ but T_c remains essentially unchanged. The melting transition was less pronounced and completely absent at 5 T. At higher fields, the vortices interact, some may become entangled,[5] and the first order nature of the freezing disappears. In fact condensation into a solid (glassy) state does not occur until S reaches zero.

We also studied the lightly twinned sample 2 and observed a slope in the melting regime that is less pronounced than that of sample 1 before deterioration. This is compatible with the effect of twinning observed in resistivity experiments.[4]

In conclusion, lattice melting has been observed by Seebeck-effect measurements, which involve virtually no transport current. The effect is readily seen in as-grown YBCO single crystals along the *b* axis. The analysis of the data allowed us to draw the phase diagram of the melting process taking into account thermal and quantum fluctuations. A very small signal is observed along the *a* axis, unless the sample is subjected to some treatment, presumably a change in the oxygen content. A systematic study of the effect of this parameter on the Seebeck-effect signal will be undertaken. Since the oxygen content affects the density of carriers and the Seebeck effect in the normal state depends on the density of states at the Fermi level, some correlation is expected. When the *a* axis effect becomes large enough to be analyzed, anisotropy studies with this type of measurement will be possible.

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