

## Precise magnetization measurements of single crystalline $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$

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Precise dc-magnetization measurements, using a superconducting quantum interference device magnetometer (SQUID), have been performed on single crystalline  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  in magnetic fields both parallel to the  $c$  axis ( $H\parallel c$ ) and tilted away from the  $c$  axis towards the  $ab$  plane to investigate the vortex state at various temperatures between 40 K and  $T_c = 83.0$  K. The magnetization curves at a fixed temperature as a function of magnetic field ( $H\parallel c$ ) show a clear jump at  $H_M$ , which corresponds to the flux-line-lattice-melting transition (FLLMT) as observed previously. In the vicinity of  $T_c = 83.0$  K, it is shown that the  $H_M(T)$  line changes its character near  $T^* = 79.5$  K, below which it can be described well by the FLLMT and above which the change of magnetization,  $\Delta M$ , and the corresponding change of the entropy,  $\Delta S$ , at  $H_M$ , falls sharply as  $T_c$  is approached. A simultaneous decoupling of the Josephson coupling and the melting of the flux-line lattice may be an appropriate picture below  $T^*$ , whereas above  $T^*$  an additional degree of freedom makes a contribution to the FLLMT due, perhaps, to vortex-antivortex creation and annihilation processes. In a tilted magnetic field, it is found that the angular dependence of  $H_M$ , as well as  $\Delta M$  and  $\Delta S$ , obeys a scaling law up to  $\theta \leq 80^\circ$ . The deduced anisotropy parameter  $\gamma_s \approx 9$  is found to be much lower than the value of  $\gamma_l \approx 150\text{--}200$  found in the liquid state. We interpret such a discrepancy as due to the difference of the dimensionality of the vortex pancake state above and below  $H_M$ . Furthermore, from the scaling behavior of  $\Delta S$  in tilted fields, it is inferred that the FLLMT is predominantly ruled by the number of pancake vortices,  $n$ , in the superconducting  $\text{CuO}_2$  layers and that Josephson vortices do not play an important role for the FLLMT. [S0163-1829(98)02618-6]

### I. INTRODUCTION

Since the flux-line-lattice state in a type-II superconductor between  $H_{c1}$  and  $H_{c2}$  was first predicted in 1957 by Abrikosov,<sup>1</sup> using the Ginzburg-Landau approach, the phase diagram of the type-II superconductor had been thought to have been established until theoretical predictions of the flux-line-lattice-melting transition (FLLMT) by Nelson and Seung,<sup>2</sup> Houghton *et al.*,<sup>3</sup> and Brandt<sup>4</sup> in 1989. These works were triggered by the anomalous resistivity broadening phenomena<sup>5</sup> in the vortex state in high-temperature superconductors, which were discovered in 1986. Since then, although numerous experimental evidence strongly suggested the FLLMT, it was not convincing until experimental thermodynamical confirmation had been achieved by Zeldov *et al.*<sup>6</sup> and others<sup>7,8</sup> for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{7+\delta}$  and Liang *et al.*<sup>9</sup> and Welp *et al.*<sup>10</sup> for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . Zeldov *et al.*<sup>6</sup> measured the thermodynamical equilibrium magnetization curve in single crystalline  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ , by a micro-Hall-probe technique, as a function of both magnetic field and temperature. They found a tiny jump in equilibrium magnetization,  $\Delta M$ , in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  in the reversible region. Following their fascinating observations, a number of similar experiments, using various experimental techniques, have been performed.<sup>11-13</sup> There had also been more indirect evidence such as small-angle neutron diffraction,<sup>14</sup> muon spin resonance,<sup>15,16</sup> precise resistivity measurements,<sup>17-19</sup> torque magnetometry,<sup>20,21</sup> etc. The anomalies so far reported as an indication of the FLLMT have been detected only in the

purest samples both for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  and  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , and the corresponding physical quantities show an abrupt change at the critical temperature  $T_M$ , suggesting that the transition may be first order. More recently, Schilling *et al.*<sup>22,23</sup> have measured the specific heat of a very pure sample of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  and discovered a small, but distinct spikelike feature in the specific heat, which indicates a thermodynamical first-order phase transition associated with the FLLMT.

Most experiments have been carried out for the case of the magnetic field applied parallel to the  $c$  axis ( $H\parallel c$ ). It is reasonable to assume that all flux lines below  $T_M$  penetrate perpendicularly to the superconducting  $\text{CuO}_2$  planes in this case. When the magnetic field is rotated towards the  $ab$  plane, it has been believed that the flux lines become like a staircase structure and form a Josephson vortices between the superconducting layers. When the magnetic field direction is nearly parallel to the  $ab$  plane, it is highly intriguing to ask what kind of vortex structure is more stable and, if it exists, what kind of melting sets in below  $T_M$ . Does it melt in the same manner as that for  $H\parallel c$ , or is there a transition at a certain angle  $\theta$ ? These questions have been raised, in particular, in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  because of its very high anisotropy parameter in the superconducting state. In a previous study, we have indeed observed a peculiar feature in resistivity measurements in tilted fields. The anomaly corresponding to the FLLMT persists up to a value very close to  $\theta \approx 90^\circ$ , even within  $1^\circ$ .<sup>24</sup> In such a highly tilted field, it has been com-

monly thought that the pancake vortices are all shifted uniformly along the external field direction at the adjacent layers, between which Josephson vortices are formed with a characteristic length scale of  $\gamma_s \approx \lambda_{ab} \sim 2100 \text{ \AA}$ , resulting in a very uniform magnetic field distribution between superconducting  $\text{CuO}_2$  layers.

In order to understand the nature of the FLLMT phenomenon more deeply, particularly to answer the question raised above, we have measured precise magnetization curves of high-quality single crystalline  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  grown by us. We have discovered that the experimentally obtained FLLMT line can be described very well by the flux-line-lattice-melting transition with Josephson coupling as predicted by Blatter *et al.*<sup>25</sup> We believe that a simultaneous melting and decoupling phase transition of first order is the most appropriate picture in this compound. Very close to  $T_c = 83.0 \text{ K}$ , the melting transition becomes suddenly weak at  $T^* = 79.5 \text{ K}$ . This anomalous behavior can be correlated with strong thermal fluctuation effects, which may induce spontaneous creation and annihilation of vortex-antivortex pairs. Since this process creates an additional degree of freedom, it can naturally explain the divergent behavior of the entropy associated with each pancake vortex.

The scaling behavior of  $T_M$  as a function of the tilt angle was observed up to at least  $80^\circ$ , where our measurements are limited by the experimental resolution of our superconducting quantum interference device (SQUID). This is in good agreement with our recent resistivity measurements in tilted fields<sup>24</sup> and is in contrast with the recent preliminary results of Yamaguchi *et al.*<sup>26,27</sup> They found scaling behavior up to  $\theta < 45^\circ$ . Although scaling works excellently, we found that the anisotropy parameter  $\gamma \approx 9$ , deduced from the present study, is much lower than  $\gamma \approx 150\text{--}200$  determined by other methods in this compound. We attribute this large difference in the anisotropy parameter to the difference in the anisotropy of the different vortex states:  $\gamma_s \ll \gamma_l$ , where  $\gamma_s$  and  $\gamma_l$  are the  $\gamma$  values defined in the solid and liquid phases, respectively. Moreover, the excellent scaling behavior of  $H_M$  as well as  $\Delta M$ ,  $\Delta S$ , and  $\Delta s$  observed in the present study naturally leads to the conclusion that the first-order phase transition at  $H_M(\theta)$  in a tilted field is driven mainly by the number of pancakes in the  $\text{CuO}_2$  plane and that Josephson vortices do not play an important role in the phase transition in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ . We discuss the angular dependence of the jump in magnetization,  $\Delta M$ ; the associated entropy change  $\Delta S$ ; and the entropy change per pancake,  $\Delta s$ . We compare them with the results obtained recently by the micro-Hall-probe technique used by Morozov *et al.*<sup>28</sup> and Schmidt *et al.*<sup>29</sup>

## II. EXPERIMENTAL DETAILS

A piece of single crystal of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  was cut from a bulk single crystal, which was grown by the traveling solvent floating zone (TSFZ) method. The dimensions of the sample are  $2.8 \text{ mm} \times 1.55 \text{ mm} \times 29 \text{ }\mu\text{m}$  with a mass of  $0.5846 \text{ mg}$ . The superconducting transition temperature  $T_c$  was observed at  $83.0 \text{ K}$  in  $2 \text{ Oe}$ .

High-precision dc-magnetization measurements have been performed on this piece of single crystalline  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  in magnetic fields parallel to the  $c$  axis

using a SQUID magnetometer (Quantum Design, MPMS-5S). The reciprocal sample option (RSO) as well as the conventional method with a 4-cm scan length was used for the measurements. For data acquisition, a programmed sequence was executed with varying field-up and field-down steps of a few gauss each at constant temperatures between  $40 \text{ K}$  and  $T_c = 83.0 \text{ K}$ . The data at each point were accumulated 5 times with RSO setting parameters such as the frequency at  $4 \text{ Hz}$ , an amplitude of  $1 \text{ cm}$ , and  $50$  cycles.

The background contribution due to the sample holder was subtracted by measuring the magnetization without the sample under the same conditions as those with the sample present.

Since the sample shape is platelike thin and flat, a considerable demagnetization effect is expected, especially in the low-field region near  $H_{c1}$ . We estimated the demagnetization factor from the sample dimensions, which are approximated as a flat, oblate ellipsoid with principal axes of  $a \approx b \gg c$ . Using the value of  $c/a \approx 1.6 \times 10^{-2}$  for the sample used in the present experiments, we obtain a value of  $N_z \approx 0.97$ . This yields a large enhancement of the initial slope of the magnetization curve, i.e., the initial susceptibility  $\chi_{\text{ini}}^{\text{cal}}$ , by a factor of  $1/(1 - N_z) \approx 40$ , which is in fairly good agreement with the observed value of  $\chi_{\text{ini}}^{\text{expt}} \approx -3.0$  instead of  $-1/4\pi = -0.0795$  for the case without a demagnetization effect. The magnetic field corresponding to the initial flux entry is also shown in the inset of Fig. 3 for this particular sample.

Resistivity measurement in magnetic fields had been performed in detail prior to the magnetization measurements. It was confirmed by both resistivity and magnetization measurements that there is no additional structure near  $T_c = 83.0 \text{ K}$ , which had a width of  $\Delta T_c \approx 1.5 \text{ K}$ . A more detailed characterization of the single crystals will be described elsewhere.

Although the doping level of the sample used in the present study was not determined, it is most likely that the sample lies in the slightly overdoped region judging from the  $T_c$  value in our as-grown sample.

## III. RESULTS, ANALYSES, AND DISCUSSIONS

### A. FLLMT in $H \parallel c$

In our previous studies, the anomalous behavior of the resistivity in a tilted magnetic field was reported in single crystalline  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ .<sup>24</sup> Based on these results, we have been motivated to measure the equilibrium magnetization of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  in detail.

In Fig. 1 a set of magnetization curves, measured as a function of magnetic field, are presented in order to show an overview of the anomaly in magnetization associated with the FLLMT, which is indicated by small arrows in the figure. As the temperature is decreased below  $T_c = 83.0 \text{ K}$ , the diamagnetic magnetization grows gradually with a small hysteresis region in the lower-field region and is accompanied by a small jump in magnetization at higher fields. Within the accuracy of our measurements, in which the experimental resolution is limited by a value  $\Delta M \approx 1.5 \times 10^{-3} \text{ G}$ , which is equivalent to  $11 \text{ mA/cm}^2$  in the critical current density  $J_c$ , the hysteresis disappears just at the FLLMT. This guarantees that those measurements are done under thermodynamical

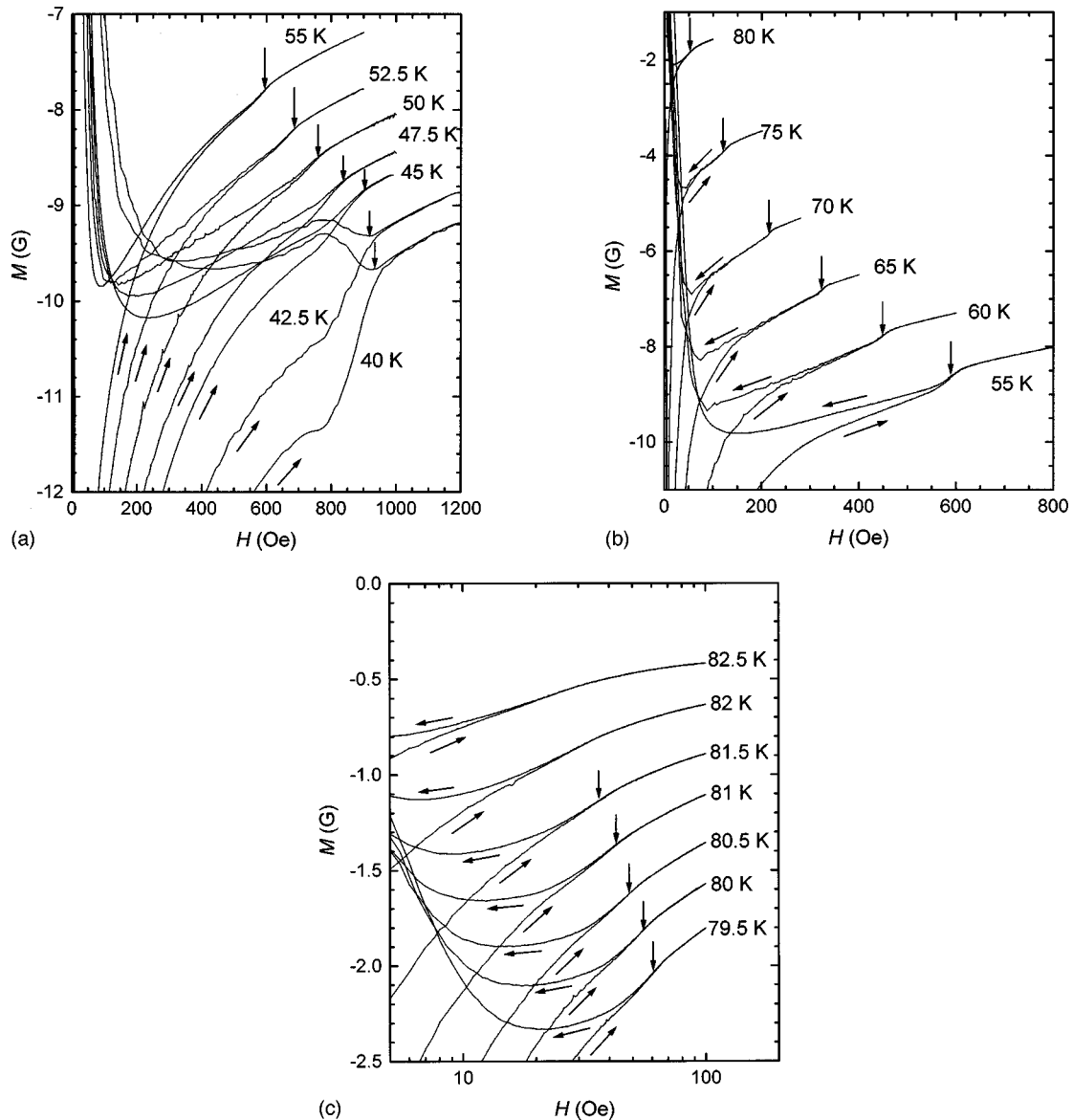


FIG. 1. Magnetization curves of single crystalline  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  as a function of magnetic field at various fixed temperatures as indicated. The magnetic field directions are indicated by arrows. (a) Between 40 and 55 K, (b) between 55 and 80 K, and (c) between 75 and 82.5 K. The vertical arrow represents the FLLMT point defined by the peak in the derivative (see text).

equilibrium. However, this situation is violated gradually as the temperature is lowered. As seen in Fig. 1, the magnetization curves begin to show hysteresis just at  $H_M$  at low temperatures and the jump in magnetization becomes nearly indistinct below about 45 K. With a further temperature decrease, the jump in magnetization at  $H_M$  is no longer observable and changes its character to an anomalous peak effect at about 45 K as previously reported.<sup>11</sup> According to Majer *et al.*,<sup>30</sup> the irreversibility in magnetization is due to the geometrical effect in the higher-temperature region, and bulk pinning comes into play effectively only below the region where the peak effect begins to appear.

In Fig. 2, derivative curves of the magnetization around  $H_M$ , selected from Fig. 1, are shown in order to show the sharpness of the transition. A sharp peak associated with the jump in magnetization is clearly visible, but its width gradually increases as the temperature is decreased. This broadening effect of the FLLMT is attributed to the effect of a

gradual participation of the bulk pinning effect with decreasing temperature. In the vicinity of  $T_c$ , which is shown in Fig. 2(b), the sharp-peak feature is no longer visible even in the derivative of  $M$ , but there remains a hump, which is associated with the peak at lower temperatures as indicated by arrows. This hump also becomes faint at 82 K, just 1 K below  $T_c$ .

In Fig. 3 the peak position in the derivative of the magnetization curve, which is taken as the FLLMT point, is plotted in an  $H$ - $T$  diagram. The overall behavior agrees with the results previously obtained. At lower temperatures the FLLMT line tends to saturate because of merging into the peak effect region, while at higher temperatures it behaves as a concave curve as a function of temperature. The temperature dependence of  $H_M$  has been analyzed by taking models according to recent theoretical predictions. As pointed out by Blatter *et al.*,<sup>25</sup> the vortex lattice may undergo phase transitions differently in different regions of temperature and mag-

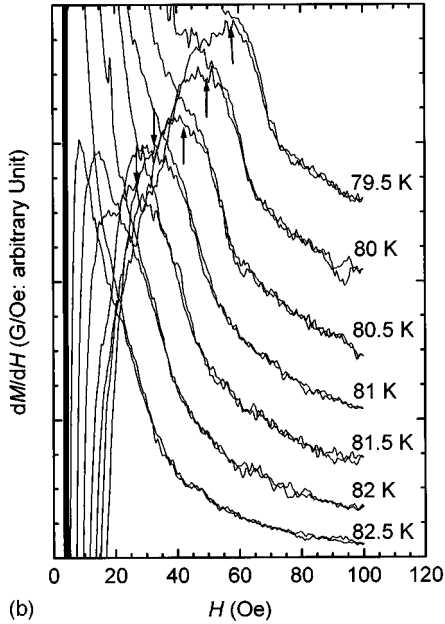
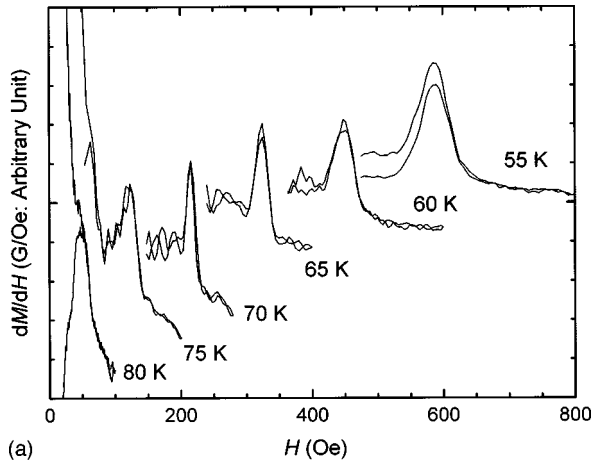


FIG. 2. Examples of the derivative curves of the magnetization with respect to the magnetic field in single crystalline  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  at the various temperatures shown in Fig. 1. (a) Between 55 and 80 K and (b) between 79.5 and 82.5 K. The arrow represents the FLLMT point.

netic field. They suggested the important role of Josephson coupling in addition to electromagnetic coupling, which may alter the nature of the FLLMT. Therefore, we take two models to analyze the data: one is the flux-line-lattice-melting model and the other the decoupling model, which are described by the formulas

$$H_M = H_{M0}(1 - T/T_c)^\alpha \quad (1)$$

and

$$H_D = H_{D0}(T_c/T - 1), \quad (2)$$

respectively. As seen in Fig. 3, it is surprising that both the flux-line-lattice-melting and the decoupling models give a reasonably good fit, if we neglect the data below 50 K. When  $T_c$  is taken as a free parameter in the melting model, the fitted value of  $T_c = 86.3$  K is far greater than the actual  $T_c$ . This is seen more clearly in the inset of Fig. 3. If  $T_c$  is fixed at 83.0 K, the fit becomes much worse in the low-field re-

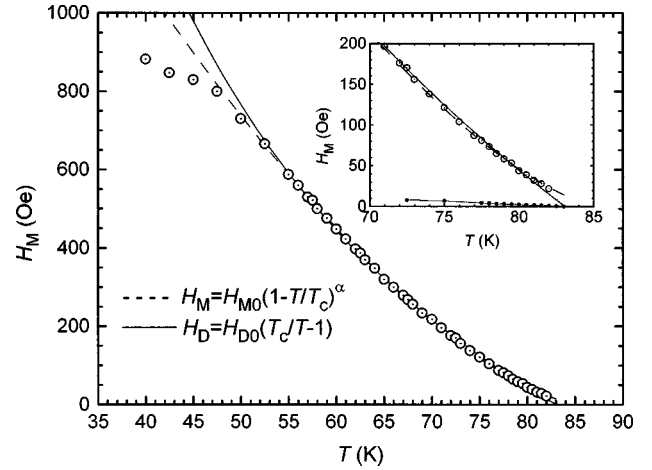


FIG. 3. Phase diagram of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ . The solid and dotted lines are obtained by fitting the data to the flux-line-lattice-melting model and the decoupling model, respectively. The inset shows the phase diagram near  $T_c$ . The solid circles represent the flux-entry field.

gion. This problem does not exist in the decoupling model:  $T_c$  obtained by the fit is  $83.08 \pm 0.07$  K, which is very close to the actual  $T_c = 83.0$  K. Although the fit has large deviations in the low-field region below about 40 Oe just below  $T_c$  in the melting model, the fitted value for  $\alpha$  is  $1.50 \pm 0.05$ , which is very close to the theoretically expected value of  $3/2$  predicted by Blatter *et al.*<sup>25</sup> They took into account both electromagnetic and Josephson interactions for their calculation of the melting transition. Moreover, using the parameter obtained in this fitting, the Lindemann parameter  $c_L$  is correctly obtained to be 0.11. This excellent agreement suggests that the FLLMT may be dominated by a decoupling character associated with the Josephson interaction rather than a simple flux-line-lattice-melting phenomenon. This is supported by the fact that the decoupling model fits better and predicts the real  $T_c$  value correctly. We note that the change in the slope in the FLLMT line at  $T^{\text{em}} \sim 68$  K obtained by  $\mu\text{SR}$  measurements by Lee *et al.*<sup>16</sup> was not found in our study. The present result, however, is also consistent with our recent low-field resistivity measurements, which will be published elsewhere.<sup>24</sup> However, we also note that there is a crossing point in the two fitted curves at  $T^*$ , which is reminiscent of  $T^{\text{em}}$  found by Lee *et al.*<sup>16</sup> Hence we do not exclude the possibility that it might exist below 40 Oe, in the temperature interval between 80 K and  $T_c$ , where an anomalous resistivity behavior is also observed in our recent measurement.<sup>24</sup> Although in the present magnetization study such an anomalous behavior could not be resolved very well just below  $T_c$ , perhaps due to the insufficient sensitivity in our SQUID magnetometer, we speculate that such anomalous behavior may indeed exist and may be related to the existence of a reentrant nature of the phase diagram in this low-field region.<sup>25</sup> However, the discrepancy of  $T^*$  and  $T^{\text{em}}$  between Lee *et al.*<sup>16</sup> and us may possibly be caused by a different sample quality, since the lattice formation is known to be very sensitive to a small amount of disorder in the system as shown in the recent scanning Hall probe microscopy (SHPM) measurements by Oral *et al.*,<sup>31</sup> who observed a semimacroscopic stripe formation of a vortex cloud sepa-

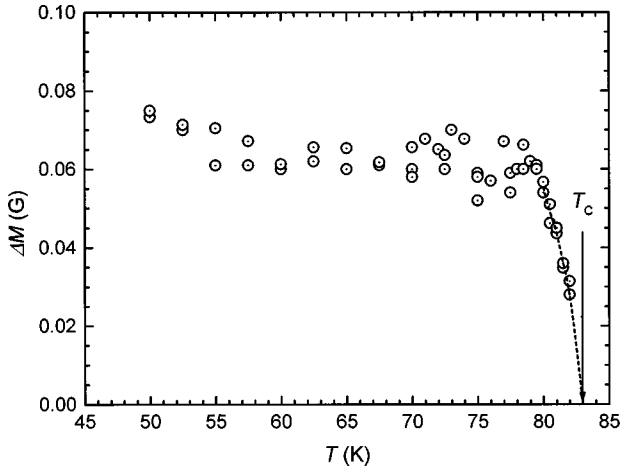


FIG. 4. Jump in magnetization,  $\Delta M$ , of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  at  $T_M$  plotted as a function of temperature.

rated on the order of  $10^{-4}$  cm along the crystallographic  $a$  or  $b$  axis. This observation strongly implies possibilities that the inhomogeneities such as dislocation networks, low-angle grain boundaries, etc., may drastically modify the nature of the melting transition of the flux-line lattice, in particular, in the low-field region.

### B. Thermodynamical analyses

In Fig. 4 the jump in magnetization,  $\Delta M$ , is plotted as a function of temperature. Although determination of the step height  $\Delta M$  becomes rather difficult in both the low- and high-temperature ends because of the growing additional peak effect and the smearing of the transition, respectively, we obtain, more or less, a constant value of  $\Delta M \approx 0.06$  G in the intermediate-temperature region between 55 and 80 K. It is noted that the error in  $\Delta M$  is considerable, approximately  $\pm 10\%$  of  $M$ , which is higher than that expected from the raw data shown in Fig. 1. This is due to the data-extracting procedure for  $\Delta M$ , which is immersed in the background slope of the  $M$ - $H$  curve.

In data by Zeldov *et al.*,<sup>6</sup> a linear temperature dependence was obtained: the value of  $\Delta M$  linearly increased with increasing temperature and  $\Delta M \approx 0.030$  G at about 80 K, which is twice as high as the value at 40 K and is half the value of the present data at the same temperature. It is interesting to note that the values for  $\Delta M$  as measured by SQUID magnetometers<sup>8,12</sup> are approximately the same, ranging from 0.04 to 0.08 G, whereas their values measured by a Hall probe are considerably smaller, being less than 0.03 G.<sup>6,11,13</sup> We speculate that this discrepancy may originate from the difference of the experimental technique.

Above about 80 K, the temperature dependence of  $\Delta M$  changes drastically as seen in Fig. 4. An unusual sudden decrease of  $\Delta M$  towards zero at  $T_c = 83.0$  K is found. Such an anomalous behavior was also observed in Hall probe measurements.<sup>6</sup> It appears that this anomalous behavior occurs in the same temperature region above the crossing temperature  $T^* \approx 79.5$  K as mentioned above. We think that this correlation cannot be accidental, and a change of the character of the FLLMT, as pointed out in Sec. III A, is inferred above  $T^*$ .

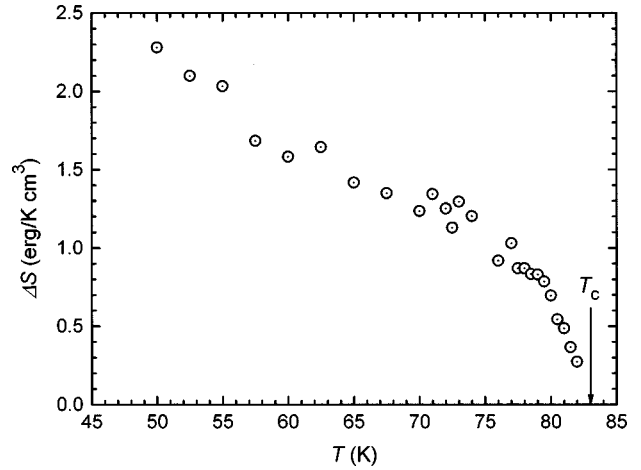


FIG. 5. Total entropy change  $\Delta S$  of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  at  $T_M$  as a function of temperature.

From the above result, the entropy change  $\Delta S$  involved in the FLLMT can be calculated by the thermodynamical Gibbs free energy  $G = U - TS$ , where  $U$  is the internal energy and  $S$  is the entropy in the system:

$$dG = \mu dN - S dT + M dH. \quad (3)$$

The Gibbs free energy should be equal at the phase transition point in the two phases. The Clausius-Clapeyron relation for a first-order phase transition is

$$\Delta S = -\Delta M \frac{dH_M}{dT}. \quad (4)$$

Associated with this change of entropy is the latent heat  $L$ :

$$L = T_M \Delta S. \quad (5)$$

Both quantities  $\Delta S$  and  $L$ , deduced from the present data, show almost linear decreases with increasing temperature up to about 79 K; then, they sharply decrease above 80 K as shown in Figs. 5 and 6. Such a sharp drop of  $\Delta S$  and  $L$  corresponds to the sudden decrease of  $\Delta M$  as described above. Although  $\Delta S$  decreases as the temperature approaches  $T_c$ , the change of the entropy per vortex per layer,

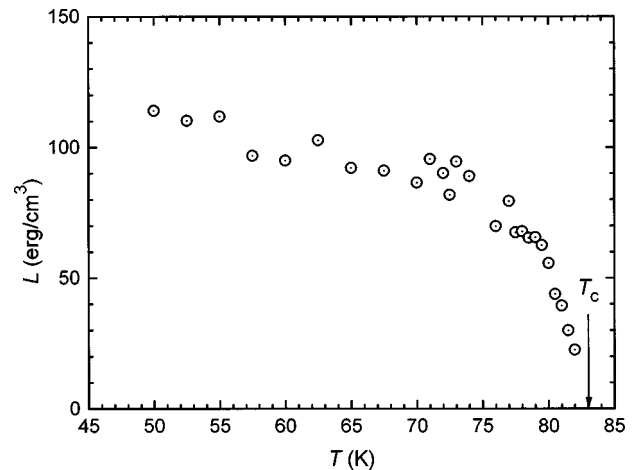


FIG. 6. Latent heat  $L$  associated with the first-order phase transition at  $T_M$  in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  as a function of temperature.

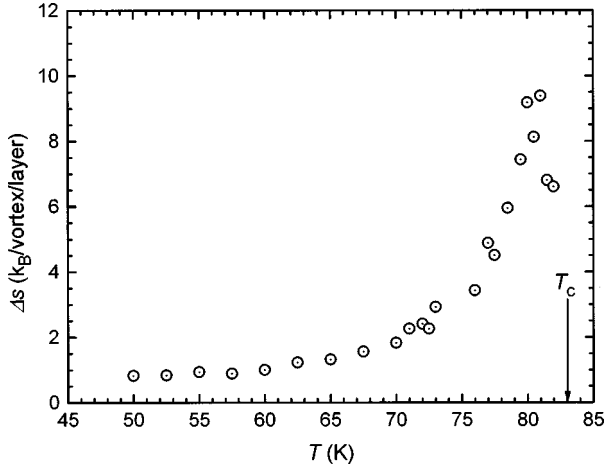


FIG. 7. Entropy change per vortex per layer,  $\Delta s$ , at  $T_M$  in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  as a function of temperature.

$\Delta s$ , strongly increases from a value of approximately  $(0.6\text{--}0.7)k_B$  at low temperatures to more than  $10k_B$  near 80 K as shown in Fig. 7. The value of  $\Delta s \approx 0.6k_B$  is in good agreement with the values predicted by recent calculations,<sup>32,33</sup> but contradict the results of Zeldov *et al.*,<sup>6</sup> which is even smaller than  $0.1k_B$ . Our results are also consistent with other recent experimental results in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ .<sup>28</sup> Although the discrepancy may arise from differences in the experimental techniques, we also consider possibilities of other degrees of freedom contributing to the entropy in the melting phenomenon. Recently, numerical Monte Carlo simulations by Hu *et al.*<sup>34</sup> and Koshelev<sup>33</sup> have pointed out that the configurational degree of freedom and superconducting fluctuations, which induce the creation and annihilation of vortex and antivortex pairs, play a significant role in the entropy, especially at low fields and high temperatures. The former authors showed a drastic increase of the number of vortices above  $T_M$  as well as the disappearance of the helicity modulus at  $T_M$ . Taking their results into consideration, we suggest a physical picture in the case of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  that has a simultaneous decoupling and decomposition into a gas of pancakelike objects in the ‘‘liquid phase’’ which occurs at  $H_M$ . Our argument given in Sec. III A is not inconsistent with this picture.

Interestingly, a similar value of  $\Delta s \approx 0.6k_B$  was also found experimentally<sup>9,10</sup> in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . In contrast with the rather temperature-independent behavior of  $\Delta s$  in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ,  $\Delta s$  in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  shows a remarkable temperature dependence as seen in Fig. 7. It reaches over  $8k_B/\text{vortex}/\text{layer}$  at the maximum at 80 K and then shows a sharp decrease above that temperature. Such an unusual temperature behavior of the entropy change associated with the FLLMT strongly implies that there are considerable extra contributions from vortex and antivortex creation and annihilation processes to the entropy participating in the phase transition starting even at very low temperatures, and such contributions progressively increase as the temperature approaches  $T_c$ , as described above. This effect is certainly not included correctly in the theoretical calculations based on the vortex-lattice-melting models using the Lindemann criterion, which result in too small values for  $\Delta s$ . This situation is apparently the essential difference that exists in the melting

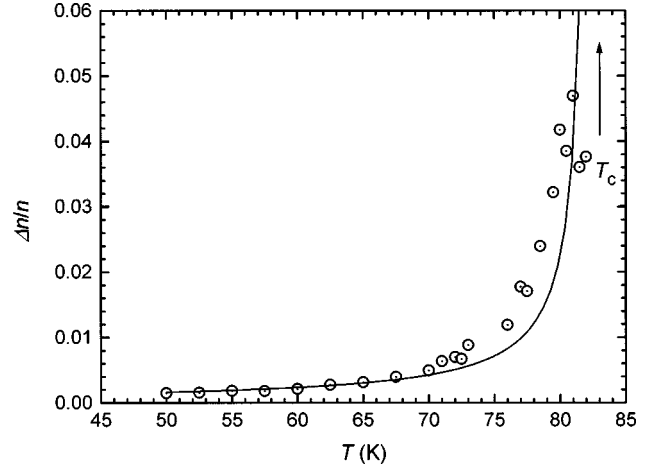


FIG. 8. Change of the density of the vortex lines,  $\Delta n$ , at  $T_M$  in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  as a function of temperature. The solid curve represents the fitted curve to  $\lambda(T)^{-2}$ .

process in the case of vortex matter, especially in highly anisotropic cases like in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ , from the conventional melting transition in a system of atoms or molecules. Vortex matter in a highly anisotropic case like  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  is, therefore, a unique system and presents an opportunity for study of the nature of the vortex phase transition in the region where the thermal fluctuation contribution is dominant.

From the results shown in Fig. 4, the relative change of the number of vortices,  $\Delta n/n$ , in a unit area at the FLLMT is calculated. It ranges from  $1.61 \times 10^{-3}$  at 50 K to  $4.91 \times 10^{-2}$  just below  $T_c$ . This large expansion of the vortex lattice in going from the liquid phase to the solid phase, especially at low fields, is of the order of 2.5%, which is about two to three orders of magnitude larger than the values estimated for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ .<sup>10</sup> This large expansion is reminiscent of the water-to-ice transition where the volume change is about 8%. According to the recent boson theory of the vortex state by Nordborg and Blatter,<sup>33</sup> the temperature variation of the vortex density,  $n$ , is expected to follow  $\Delta n(T) \propto \lambda(T)^{-2}$ , which yields a reasonably good fit to the experimental data as shown by the solid line in Fig. 8.

### C. Angular dependence of the FLLMT

When the magnetic field is tilted towards the superconducting plane, it is in general believed that the line of pancake vortices is also tilted as if the average position of the vortices lies along the field direction. In such a configuration of pancake vortices, it is of great interest to study how the FLLMT phenomenon is influenced and modified by the tilted magnetic field. The stability of the tilted flux-line lattice can be estimated by calculating the tilt energy  $E_{\text{tilt}}$  of the single flux line, which is given as<sup>35,36</sup>

$$E_{\text{tilt}} = \frac{\theta^2 C_{44}}{2} d, \quad (6)$$

where  $C_{44} \approx \phi_0^2 / (4\pi\lambda_c)^2$  and  $d$  is the distance between two pancakes. Taking values of  $\lambda_c \approx 30 \mu\text{m}$ ,  $d \approx a_0 \approx 200 \text{ \AA}$  at 500 G,  $\theta \approx \pi/2$ , and  $\phi_0 = 2.07 \times 10^{-7} \text{ G cm}^2$ , the value  $E_{\text{tilt}} \approx 6 \text{ K}$  can be obtained. Since  $E_{\text{tilt}} \ll T_M$ , this crude esti-

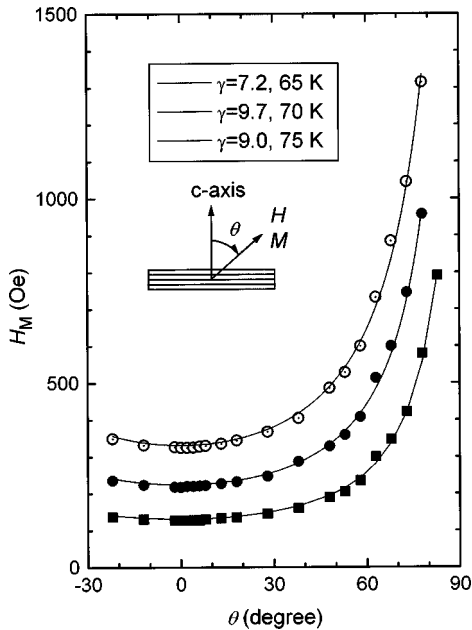


FIG. 9. Angular dependence of  $H_M(\theta)$  in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  at three temperatures 65 K (open circles), 70 K (solid circles), and 75 K (solid squares). The solid curves represent the fit to Eq. (7), with  $\gamma$  values as indicated in the figure.

mate strongly implies that the pancake vortex line in a tilted field is not stable at high temperatures. Therefore, it is expected that the FLLMT may become unstable at a certain angle  $\theta$ , when the magnetic field is tilted towards the  $ab$  plane.

Recently, specific heat measurements in a tilted magnetic field have been done in single crystalline  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  by Schilling *et al.*,<sup>23</sup> who showed the scaling behavior of the FLLMT line. They found the anisotropy parameter  $\gamma \sim 8$ , which is surprisingly identical to the values obtained by other measurements. They also found a sharp transition associated with the FLLMT even when fields were parallel to the  $ab$  plane. Such behavior is not surprising and can be expected, since the tilt energy for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  is  $E_{\text{tilt}} \approx 520 \text{ K} \gg T_M$ .

In contrast to  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , such thermodynamical measurements have not yet been successful in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ , mainly because the anomaly in the specific heat is estimated to be three orders of magnitude smaller than that of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . Therefore, it is so far only the equilibrium magnetization measurements that provide a clue to this problem.

In Fig. 9 the experimental results of the angular dependence of the FLLMT field,  $H_M(\theta)$ , in a tilted field at three different temperatures 65, 70, and 75 K are plotted as a function of the angle  $\theta$  measured with respect to the  $c$  axis as indicated in the inset. As seen in Fig. 9, it is surprising that the FLLMT persists up to  $80^\circ$ . This fact sharply contradicts the above-mentioned expectation. Furthermore, the  $H_M(\theta)$  field tends to progressively increase at larger angles.

In Fig. 10 the same data as in Fig. 9 converted to the perpendicular field component are plotted. If the system behaves as a perfect two-dimensional (2D) system, the value of  $H_M(\theta) \cos \theta / H_M(0)$  should not depend on angle. As seen in Fig. 10, however, it begins to deviate to a smaller value

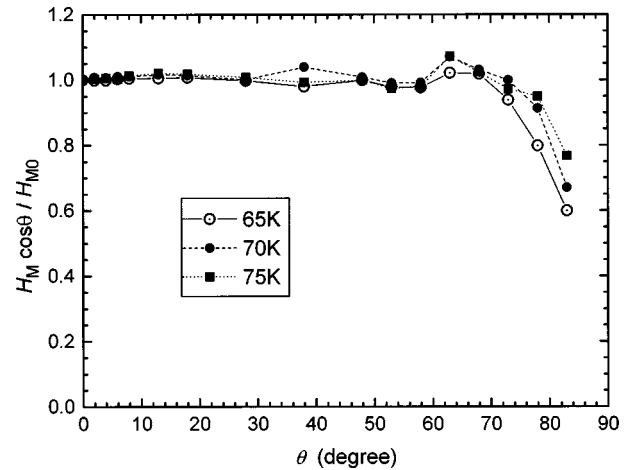


FIG. 10. Normalized perpendicular component of  $H_M$  as a function of angle in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  at three temperatures 65 K (open circles), 70 K (solid circles), and 75 K (solid squares).

above about  $70^\circ$ , indicating a breakdown of the 2D behavior and 3D interactions begin to play a role. The 2D scaling behavior, observed up to  $70^\circ$ , is not consistent with the results of Yamaguchi *et al.*<sup>26</sup> We believe that this discrepancy may originate from the different disorder levels of the samples as discussed below.

Assuming the scaling behavior in anisotropic superconductors,  $H_M(\theta)$  was fitted by the following formula:

$$H_M(\theta)/H_M(0) = [\cos^2(\theta) + \gamma^{-2} \sin^2(\theta)]^{-1/2}, \quad (7)$$

where  $\gamma$  is the anisotropy parameter. As presented in Fig. 9, we found an excellent fitting curve with two parameters  $H_M(0)$  and  $\gamma$ . From these fitting parameters, we estimated the  $\gamma$  value to range from 7.2 to 9.7, which is considerably lower than the value of  $\gamma \approx 150\text{--}200$  obtained from other measurements. The solid curves in Fig. 9 show the fitted curves using Eq. (7). It should be noted that this is not contradiction, since the values are deduced from different phases in the phase diagram: the  $\gamma$  values obtained from the present data are deduced from the vortex solid phase, while much higher values reported previously are for the liquid phase. We point out a very important fact that the anisotropy factor in the vortex solid phase is different from the intrinsic anisotropy parameter of the superconductor. The reason for this is that the anisotropy of the flux-line-lattice state can strongly be modified by the nature of the vortex lattice, which is influenced not only by the intrinsic parameters associated with the superconductivity, but also by the extrinsic effects such as vortex pinning mechanisms. This interpretation is also supported by the recent detailed resistivity measurements of the FLLMT,<sup>24</sup> as well as the Josephson plasma resonance,<sup>37,38</sup> in which a continuous reduction of the anisotropy parameter  $\gamma$  is observed with increases in the number of columnar pinnings in the solid vortex phase without a progressive reduction of  $\gamma$  in the vortex liquid phase. For example, the sample with columnar defects equivalent to 0.5 T gives rise to both a great reduction and a remarkable temperature dependence of the  $\gamma$  value:  $\gamma \approx 3$  at 45 K in the ‘‘vortex glass phase’’ and  $\gamma \approx 28$  at 84 K in the ‘‘viscous liquid phase.’’ It is noted that the  $\gamma$  value before the irradiation is about 200 in the liquid phase. The reduction of the

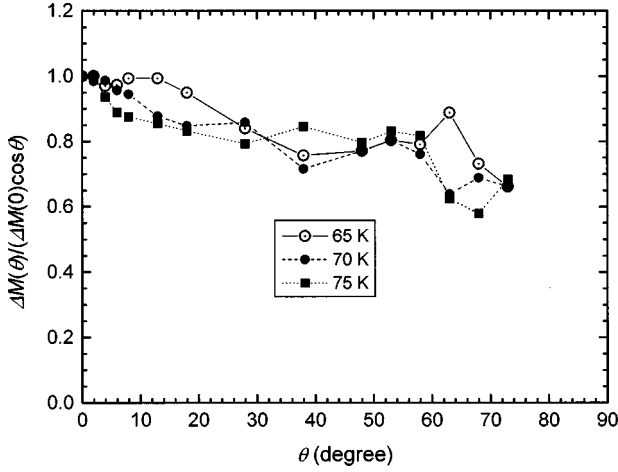


FIG. 11. Jump in magnetization,  $\Delta M$ , projected on the  $c$  axis as a function of angle in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  at three temperatures 65 K (open circles), 70 K (solid circles), and 75 K (solid squares).

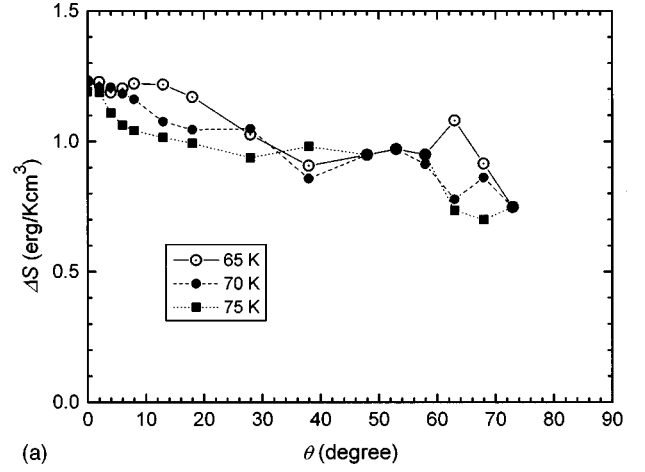
anisotropy parameter obtained in the present experiment is the first significant result of this paper.

In Fig. 11 the jump height  $\Delta M$ , normalized by its perpendicular component  $\Delta M(0)\cos(\theta)$ , is plotted at 65, 70, and 75 K. It is clearly observed that  $\Delta M/\Delta M(0)\cos(\theta)$  is almost independent of the angle up to the maximum angle measured, with a small decreasing tendency near the maximum angle, which approaches 25% at the maximum angle.

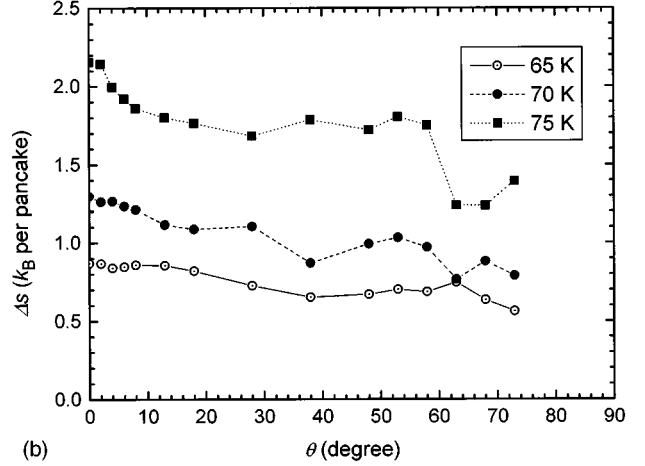
From  $\Delta M$ , shown in Fig. 11, the entropy change  $\Delta S$  is estimated as a function of  $\theta$ . This is shown in Fig. 12(a). Using Eq. (4), the entropy change per vortex pancake,  $\Delta s$ , is also estimated and presented in Fig. 12(b). It is remarkable that the both entropies are nearly independent of the magnetic field orientation over a wide range of angle to about  $70^\circ$ . As we mentioned earlier, in such a tilted field, it is expected that the vortices lying in adjacent layers will considerably displace each other, especially at higher angles. In such a case the flux-line-lattice state may not form. However, even in this case the flux line may become a staircase structure, where the vortex pancakes are connected by the Josephson vortex between layers. It is very interesting and intriguing to ask the question about how such a vortex staircase arrangement melts, keeping the same thermodynamical parameters at the FLLMT. The answer to this question can reasonably be inferred from the present experimental results. Because of the scaling of  $H_M(\theta)$ , as well as the independent nature of all thermodynamical quantities as a function of angle, we come to the conclusion that the essential parameter in the FLLMT phenomenon exists mainly in the number of pancake vortices,  $n$ . Surprisingly, in such tilted fields, it seems that the Josephson vortices play only a very minor role for the FLMMT phenomenon in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ . This is the second central result of this paper.

#### IV. CONCLUSIONS

We have measured the dc magnetization, using a conventional SQUID magnetometer, in single crystalline  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  in magnetic fields for both  $H\parallel c$  and tilted away from the  $c$  axis in order to study the nature of the FLLMT phenomena. The magnetization curves show a clear



(a)



(b)

FIG. 12. Total entropy change  $\Delta S$  (a) and the entropy change per vortex per layer,  $\Delta s$  (b), as a function of angle  $\theta$  in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  at three temperatures 65 K (open circles), 70 K (solid circles), and 75 K (solid squares).

jump at  $H_M(T_M)$ , corresponding to the FLLMT. The anomalies can be observed above 40 K, very close to  $T_c = 83.0$  K, and can be considered to be first-order phase transitions. In the vicinity of  $T_c$ , it is found that the  $H_M$  line changes character at  $T^*$ , below which it can be described well by the FLLMT as given by Blatter *et al.*<sup>25</sup> and above which the change of magnetization,  $\Delta M$ , at  $T_M$  falls sharply as  $T_c$  is approached. From the result of the fitting analysis to the existing theories, we interpret this as the occurrence of a simultaneous decoupling of the Josephson coupling and the flux-line-lattice melting above  $T^*$ . An additional degree of freedom takes a significant part in the FLLMT above  $T^*$  due to the thermal excitation of vortex-antivortex creation and annihilation processes. Although the clear crossover temperature  $T^{\text{em}} \sim 68$  K observed by Lee *et al.*<sup>15,16</sup> was not found in our sample, we do not exclude such a possibility. We speculate that  $T^{\text{em}}$  shifts to higher temperatures and lower fields and may correspond to the  $T^*$  found in our sample.

The entropy associated with the FLLMT was calculated by assuming the Clausius-Clapeyron relation for a first-order phase transition. Although the change of the total entropy,  $\Delta S$ , associated with the transition decreases with increasing temperature, the entropy change  $\Delta s$  per vortex per layer has a remarkable diverging tendency at  $T^*$ . In our case  $\Delta s$



$\approx 0.6k_B$  at low temperatures, while it rises to about  $10k_B$  at 80 K. Such anomalous behavior can be understood by considering an additional entropy participating in the phase transition as proposed by Hu *et al.*<sup>34</sup> and Koshelev.<sup>32</sup> It is noted that the value of  $\Delta s$ , as well as its temperature dependence, disagrees with the previous data of Zeldov *et al.*<sup>6</sup> They observed a linearly increasing behavior with temperature in contrast to that shown in Fig. 7. Their values of  $\Delta s \approx 0.03k_B$  at 40 K and  $2k_B$  just below  $T_c$  can be compared with  $0.8k_B$  at 40 K and  $8k_B$  at  $T^*$ . The substantially higher value of  $\Delta s$  that is reported here is consistent with the recent work of Morozov *et al.*<sup>28</sup> Their results are very similar to ours including a sharp drop in  $\Delta s$  in the vicinity of  $T_c$ .

The angular dependence of the FLLMT in single crystal-line  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  was measured at three temperatures of 65, 70, and 75 K in detail. Applying the similar thermodynamical analyses as that for  $H\parallel c$ , and using the scaling function of Eq. (7), we found a remarkable scaling behavior of  $H_M(\theta)$  in the entire range of angles up to  $80^\circ$ . The anisotropy parameter  $\gamma$  from the fitting analysis, however, resulted in a very small value of  $\gamma \approx 7-9$  at the three temperatures, compared with  $\gamma \approx 150-200$  as found usually by other methods. We attribute this discrepancy to the difference of the flux lines between two different states: in the flux-line solid phase,  $\gamma_s$  is much reduced compared with that of the liquid phase  $\gamma_l$ . This is also observed in our resistivity measurement<sup>24</sup> as well as in the Josephson plasma resonance.<sup>37,38</sup> It is concluded that the reduction of the  $\gamma$  value originates from the nature of the vortex solid phase,

which may significantly alter the intrinsic properties by extrinsic contributions such as pinning centers.

It is noted that our result, mentioned above, is not consistent with the recent report by Schmidt *et al.*,<sup>29</sup> who claim that no significant improvement was found in their analysis of  $H_M(\theta)$  by using Eq. (7). Although they did not deduce the exact  $\gamma$  value, the significant deviation from 2D scaling of the 70 K data above about  $70^\circ$  was mentioned. This behavior is very similar to our experimental finding. Therefore, reinterpreting their results, we believe that the similar  $\gamma$  value would be expected in their results too.

The scaling behavior of the FLLMT and the angular dependence of  $\Delta S$ , as well as  $\Delta s$ , strongly suggest that the FLLMT in a tilted field is governed mainly by the number of pancakes, and the Josephson vortices associated with the kink formation do not contribute to the FLLMT phenomenon. This may be a natural consequence in highly anisotropic superconductors like  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  where the tilt energy of the vortex line is so weak that the interlayer coupling does not play a role in maintaining a rigid flux line, which results in 2D-like vortex lattice order within  $\text{CuO}_2$  layers. Such speculation should be confirmed by further experiments in the future.

#### ACKNOWLEDGMENTS

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