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Anisotropy of magnetization discontinuity at vortex-lattice melting in untwinned $YBa_2Cu_3O_{7-\delta}$

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We measured the magnetic torque τ experienced by an untwinned $YBa_2Cu_3O_{7-\delta}$ single crystal in external magnetic fields up to $\mu_0 H = 7$ T below the critical temperature $T_c = 93.3$ K, as a function of Θ, the angle between H and the c axis of the crystal. At the vortex-lattice melting transition we observe discontinuities in both τ and $(\partial\tau/\partial H)_T$, that are related to changes in the transverse components of the magnetization vector **M** and $(\partial \mathbf{M}/\partial H)_T$, respectively. We use thermodynamic relationships to determine the direction of the vector $\Delta\mathbf{M}$ in space, and show that $\Delta\mathbf{M}$ is always directed parallel to **M**. The discontinuities in magnetization $\Delta\mathbf{M}$ and in entropy ΔS vanish slightly below the temperature where the melting field $H_m(T)$ extrapolates to zero, which could indicate the existence of a lower critical point of the vortex-lattice melting line. From our $(\partial\tau/\partial H)_T$ data we are able to extract the differences in the reduced specific heat $\Delta C/T$ between the vortex-fluid and the vortex-solid phases, and we compare these results with corresponding thermal data. We finally examine the validity of standard angular scaling rules for anisotropic superconductors for the melting fields $H_m(T,\Theta)$ at temperatures as high as $T/T_c = 0.99$.

I. INTRODUCTION

The solid-to-fluid transition of the vortex system in cuprate superconductors in an external magnetic field H separates a magnetically irreversible, zero-resistance state from a reversible state with dissipative transport properties.[1] This transition has been extensively studied by resistivity measurements,[2–4] torsional oscillator techniques,[5,6] muon-spin rotation[7] and neutron-scattering experiments,[8] and the measurement of thermodynamic properties such as magnetization[9–17] and specific heat.[18–30] Magnetization data have been obtained mainly on single crystals of $Bi_2Sr_2CaCu_2O_8$,[9,10,13–15] $YBa_2Cu_3O_{7-\delta}$,[11,12,17,20] and $(La, Sr)_2CuO_4$,[16] while associated thermal effects have been detected so far only in the $YBa_2Cu_3O_{7-\delta}$ family.[18–30] These thermodynamic data indicate that in sufficiently clean samples and in moderate external magnetic fields, the phase transition is of first order, while sufficient disorder and/or the application of large magnetic fields may drive the transition continuous.[10,19–22,25,29,30]

The magnetic phase diagram of anisotropic superconductors not only depends on the external magnetic field H and the temperature T, but also on the angle Θ between the magnetic field and the c axis of the crystal.[3,5,6,14,15,24,26,31] Hence all thermodynamic quantities related to the vortex-lattice melting transition, such as the melting field H_m and the discontinuities in entropy S and magnetization M, may depend on Θ.[32–34] For $Bi_2Sr_2CaCu_2O_8$, the angular dependence of magnetization anomalies ΔM has been measured,[14,15] and imply discontinuities in entropy ΔS at vortex-lattice melting via the Clapeyron equation. A direct thermal study about the angular dependence of both ΔS and related discontinuities in specific heat ΔC has been done only on $YBa_2Cu_3O_{7-\delta}$,[24,26] and confirmed theoretical treatments of the effects of anisotropy on the thermodynamic properties of anisotropic superconductors.[32,33] Although the angular dependence of ΔS in $YBa_2Cu_3O_{7-\delta}$ is now fairly precisely known,[26] no corresponding measurements of the angular dependence of the discontinuity in magnetization $\Delta\mathbf{M}$ have been reported. In very anisotropic materials such as $Bi_2Sr_2CaCu_2O_8$, **M** is virtually parallel to c for a wide range of angles Θ, except extremely close to $\Theta = 90°$, which considerably simplifies the geometry of the problem.[14,35] In $YBa_2Cu_3O_{7-\delta}$, however, the electronic anisotropy is rather low, and the magnetization vector **M** is markedly tilted away from the direction of the c axis of the crystal already for angles of the order of $\Theta = 75°$ and larger.[35] There are plausible arguments based on the London theory predicting that for an arbitrary angle Θ, $\Delta\mathbf{M}$ is always directed parallel to **M**. In this case, the magnetic induction **B** would slightly rotate at the melting transition. However, in an alternative view of the situation it is the thermodynamic variable **B** that shows the discontinuity $\Delta\mathbf{B}$

$=\mu_0\Delta\mathbf{M}$ along $\mathbf{B}$, which simply reflects a change in the number of vortex lines directed parallel to the magnetic-flux density vector $\mathbf{B}$. In this scenario, $\Delta\mathbf{M}$ is not always parallel to the magnetization $\mathbf{M}$, and would lead to a rotation of $\mathbf{M}$ at the melting transition. For the simple geometry $\Theta=0°$ that has already been studied in the literature,[11,12] $\mathbf{M}$, $\Delta\mathbf{M}$, $\mathbf{H}$, and $\mathbf{B}$ are all parallel to the c axis of the crystal, and the above distinction is not necessary.

Recent progress in torque magnetometery and in sample preparation techniques allowed us to detect the transverse component of the discontinuity $\Delta\mathbf{M}$ (i.e., the component perpendicular to the applied magnetic field $\mathbf{H}$) at the vortex-lattice melting in $YBa_2Cu_3O_{7-\delta}$ with previously unknown precision.[17,36,37] However, the determination of the direction of $\Delta\mathbf{M}$ in an uniaxial situation requires the knowledge of two components, the transverse and the longitudinal component (i.e., the $\Delta\mathbf{M}$ component parallel to $\mathbf{H}$). In this work we perform a series of systematic measurements of the transverse component of $\Delta\mathbf{M}$. Knowing the angular dependence of the associated discontinuity ΔS, and applying thermodynamics, we are able to extract also the longitudinal component of $\Delta\mathbf{M}$. This makes it possible to finally determine the direction of $\Delta\mathbf{M}$ in space.

The temperatures where the discontinuities in entropy $\Delta S(T)$ vanish define critical points of the vortex-lattice melting line, where the first-order character of the phase transition is lost. While the upper critical point can be very well identified from thermal data, the determination of a possible lower critical point from our magnetic data is less straightforward because it crucially depends on the definition of the critical temperature T_c. Our data indicate that ΔS vanishes below the temperature where $H_m(T)$ extrapolates to zero, which might mean that the first-order character of the phase transition at $H_m(T)$ disappears at $T<T_c$.

Along with the steps in entropy and magnetization at the vortex-lattice melting transition, simultaneous steplike variations in the reduced specific heat $\Delta C/T$ and changes in slope in magnetization, $\Delta(\partial M/\partial H)_T$ and $\Delta(\partial M/\partial T)_H$, respectively, have been observed.[11,12,17–19,22–30] These changes are related one to another by a thermodynamic relationship that is rather simple for $H\|c$.[24] In our experiments measuring the magnetic torque, $\boldsymbol{\tau}=\mu_0\mathbf{M}\times\mathbf{H}$ per volume of sample, we also observe corresponding changes in slope, $\Delta(\partial\tau/\partial H)_T$. We develop a suitable thermodynamic relationship to transform these Θ-dependent slope changes into angular-independent $\Delta C/T$ data, and compare the results with the data from a direct thermal measurement.

It has been verified by various experimental techniques[6,26,38] that, to first approximation, the standard scaling rules for thermodynamic quantities in anisotropic superconductors do apply to the moderately anisotropic $YBa_2Cu_3O_{7-\delta}$. The parameter describing the degree of anisotropy can be estimated by scaling the melting fields $H_m(T,\Theta)$ at fixed temperature. Since we are able to detect the vortex-lattice melting transition as a function of angle at temperatures as high as $T/T_c=0.99$, we can test the validity of these scaling rules for $H_m(T,\Theta)$ up to this temperature.

II. EXPERIMENT

Our homebuilt experimental setup to collect the magnetic-torque data has been described in detail in Refs. 36–37. We used capacitive cantilevers with a resolution in torque of better than 10^{-12} Nm. The experiments above $T=91.5$ K were done in a normal conducting magnet. Below 91.5 K, we used a commercial PPMS (where PPMS is a Physical Property Measurement System, Quantum Design) with a split-coil magnet that provided magnetic fields up to $\mu_0H=7$ T. The temperature scales in the two systems were carefully matched by detecting known reference points. The sample is a 380 μg naturally untwinned $YBa_2Cu_3O_{7-\delta}$ single crystal with a $T_c=93.3$ K. The very high quality of the sample is reflected in a small total width of the vortex-lattice melting transition, $\delta T\approx 60$ mK (see Sec. VI of this paper), which has to be compared to a δT of the order of 200 mK and more as observed in earlier samples.[11,12,19–28] Our torque data show very little magnetic hysteresis below the vortex-lattice melting transition,[17] therefore vortex pinning is much less pronounced than in other high-quality samples where the transition is also observable. All experiments described here were done by rotating the magnetic field in the plane that is defined by the a and the c directions of the crystal. Hence, all numerical values obtained in our data analysis are valid for this geometry.

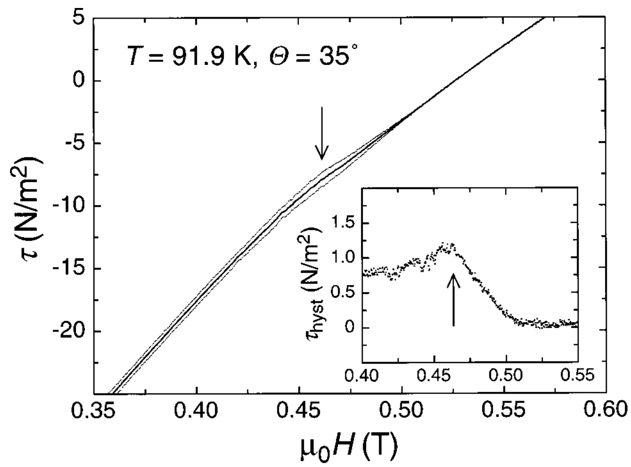

FIG. 1. Representative magnetic torque per sample volume τ vs magnetic-field data for an untwinned $YBa_2Cu_3O_{7-\delta}$ single crystal ($T=91.9$ K and $\Theta=35°$). The three curves are data taken for increasing (upper curve) and decreasing (lower curve) magnetic field, and the average of the two data sets (middle curve). In the inset we plotted the magnetic-hysteresis width τ_{hyst} that corresponds to a critical-current density of the order of $J_c\approx 2$ A/cm^2, and that shows a weak peak effect around the melting transition (indicated by an arrow).

In Fig. 1 we show typical torque-versus-magnetic-field raw data ($T=91.9$ K, $\Theta=35°$), for increasing and decreasing magnetic field around the vortex-lattice melting transition. In the case of hysteresis we assumed that the ''reversible'' torque signal is represented by the average of the two curves, i.e., that the hysteresis width (inset of Fig. 1) is symmetric for increasing and decreasing magnetic field. Each of these curves was analyzed assuming a Gaussian distribution of first-order transitions, centered around H_m, with a full-width δH and total amplitude $\Delta\tau$. At the same time $(\partial\tau/\partial H)_T$ was allowed to change its slope continuously according to the same Gaussian distribution. A smooth background was represented either by polynomials $P(H)$ in H of the order 3 or 4, or by polynomials in ln(H) of the form τ

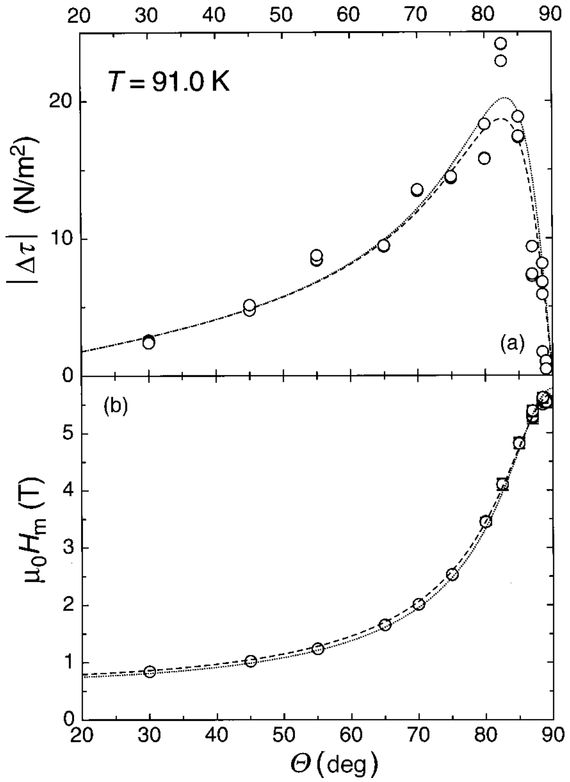

FIG. 2. (a) The discontinuity in magnetic torque, $|\Delta\tau|$ per sample volume, at the vortex-lattice melting transition for $T = 91.0$ K, plotted as a function of the angle Θ between the applied magnetic field and the c direction of the crystal. The dotted curve corresponds to a fit to Eqs. (1) and (A7) using $\gamma = 8.2$, while the dashed line represents a corresponding fit with $\gamma = 7.6$ (see text). (b) The melting fields H_m at $T = 91.0$ K, as a function of Θ.

$= A_1 + A_2 HP(\ln[H])$ (with additional fitting parameters A_1 and A_2) to account for the approximate logarithmic H dependence of the magnetization M. In the following figures we plot the results of all these fits that have been done in a data range of typically $H_m \pm 20\%$, to illustrate the effect of changing the model for the background on our results for $\Delta\tau$ and $\Delta(\partial\tau/\partial H)_T$.

III. DIRECTION OF THE MAGNETIZATION-DISCONTINUITY VECTOR ΔM

At a fixed temperature T and for each chosen magnetic field value H we varied the angle Θ between $\mathbf{H}$ and the c axis of the crystal. The resulting discontinuities in magnetic torque per sample volume, $\Delta\tau(\Theta)$, are shown in Fig. 2(a) for $T = 91.0$ K, and the corresponding melting fields $H_m(\Theta)$ are plotted in Fig. 2(b). From such $\Delta\tau(\Theta)$ data we are able to calculate the angle ε between the magnetization-discontinuity vector $\Delta\mathbf{M}$ and the c direction of the crystal [see Appendix with Fig. 9, Eqs. (A7) and (A9)]. All the $\varepsilon(\Theta)$ data, taken at six different temperatures, are shown in Fig. 3(a). For small angles (i.e., smaller than $\approx 75°$), $\Delta\mathbf{M}$ appears to be almost parallel to c. At larger angles, $\Delta\mathbf{M}$ is tilted away from c towards the direction of $\mathbf{H}$ (i.e., $\varepsilon < 0$). The fact that a discontinuity in τ can be detected in magnetic-torque measurements already indicates that $\Delta\mathbf{M}$ cannot be parallel to $\mathbf{B}$. If $\Delta\mathbf{M}$ were along $\mathbf{B}$, the magnetic induction vector would not rotate at the melting transition, and $\Delta\tau = 0$ according to Eq. (A10). The angle of rotation of $\mathbf{B}$, i.e., of the average direction of the magnetic-flux lines, is plotted in Fig. 4.

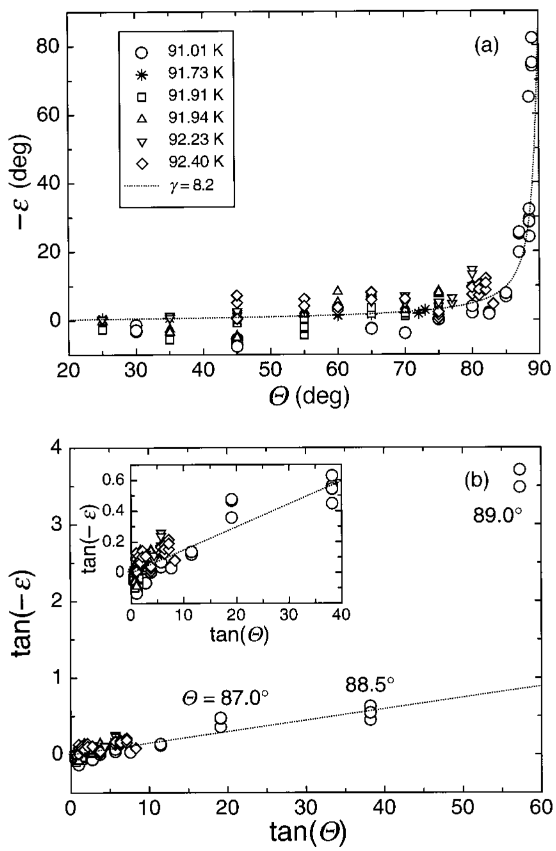

FIG. 3. (a) The angle ε between the magnetization-discontinuity vector $\Delta\mathbf{M}$ and the c direction of the crystal, as a function of Θ and for different temperatures T. The dotted line corresponds to our expectation according to Eqs. (A7) and (1) for $\gamma = 8.2$, assuming that $\Delta\mathbf{M}$ is parallel to $\mathbf{M}$ (see text). (b) The data from (a), plotted as $\tan(-\varepsilon)$ vs $\tan(\Theta)$. For $\Delta\mathbf{M}$ parallel to $\mathbf{M}$, the data should collapse onto a single line with a slope γ^{-2} [see Eq. (1) in the text]. The inset shows the same data, but excluding the data point at $\Theta = 89.0°$ for clarity.

The direction of $\Delta\mathbf{M}$ can be compared with predictions about the direction of the magnetization vector $\mathbf{M}$ in uniaxial superconductors.[35] The angle φ between $\mathbf{M}$ and the negative c direction is given by

$$\tan(\varphi) = \frac{1}{\gamma^2}\tan(\Theta), \tag{1}$$

where γ^2 is the effective-mass anisotropy for current transport parallel and perpendicular to the c direction.[35] This result should only weakly depend on the geometry of the vortex lattice, because in deriving Eq. (1) the geometry factors accounting for the vortex-lattice structure cancel out to first approximation. Thus, it is reasonable to assume that φ has the same value in the vortex solid and in the vortex-fluid state, and $\Delta\mathbf{M}$ should always be directed parallel to $\mathbf{M}$, with $\varepsilon = -\varphi$. In Fig. 2 we plotted our expectations according to Eq. (1) using a value $\gamma = 8.2$ that we derived earlier by systematically scaling all the melting fields $H_m(\Theta)$ from

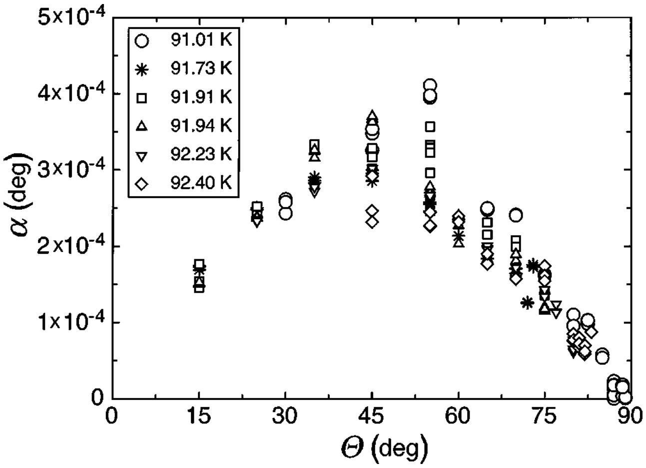

FIG. 4. The angle of rotation α of the magnetic-induction vector **B** (i.e., the average direction of the vortex lines) upon vortex-lattice melting, as a function of Θ and for different temperatures T.

angular-dependent thermal data on a similar crystal using Eqs. (A1)–(A2).[26] The melting fields $H_m(\Theta)$ that we measured directly with our torque magnetometer at this particular temperature are best fitted with $\gamma \approx 7.6$, however, which is slightly lower than the result from the global fit (see below). A possible variation of γ with a temperature that might account for this discrepancy is discussed later in Sec. VI of this paper. The corresponding $\varepsilon(\Theta)$ line for $\gamma = 8.2$ is shown in Fig. 3. In Fig. 3(b) we plotted the same data as $\tan(-\varepsilon)$ vs $\tan(\Theta)$. Except for the data point at $\Theta = 89°$, there is a fair agreement with Eq. (1) indicating $\Delta\mathbf{M}$ is indeed parallel to **M** for a wide range of angles Θ, even when **M** is already off by more than 20° from the c direction. The deviation of the last data point at $\Theta = 89°$ from Eq. (1) cannot be explained by a systematic offset of the direction of the applied magnetic field with respect to the c axis of the crystal, which we estimate to be less than 0.03°, leading to negligible error bars in Fig. 3. However, we cannot definitely conclude that this deviation is really significant and would indicate a change in the thermodynamics of the vortex-lattice melting very near the $H \| ab$ geometry.[39,40] It might be due to the effect of a weak, undetected mechanical vibration of the crystal disturbing the measurement, or to an unknown misalignment inside the crystal leading to additional errors in $\varepsilon(\Theta)$.

IV. MELTING ENTROPY AND CRITICAL POINTS OF THE FIRST-ORDER MELTING LINE

From our data and using Eqs. (A7) and (A9), we may extract values for the discontinuity in magnetization ΔM_0 for $\Theta = 0°$ [see Fig. 5(a)], and convert them to changes in entropy, ΔS, by making use of the Claussius-Clapeyron Equation (A4). In Fig. 5(b) we compare these results with the corresponding values that we deduced from our previously published $\Delta M_0(T)$ data taken on the same crystal [Fig. 5(a)],[17] and with ΔS data from a direct thermal study on a similar but larger crystal with a lower $T_c = 92.0$ K.[26] The data show a systematic, virtually linear decrease in the melting entropy (counted per mole of sample) as T approaches T_c. This decrease in ΔS has been quantitatively correctly reproduced by a recent theory that is based on the London model for describing the thermodynamics of the vortex system in $YBa_2Cu_3O_7$.[41] As we will see in the next paragraph, the theory only poorly describes the magnitude of the simultaneous steplike variations in the reduced specific heat,[33] $\Delta C/T$, that are associated with changes in the slopes $(\partial\tau/\partial H)_T$ at the vortex-lattice melting transition.

Both the magnetic $\Delta M_0(T)$ data and the $\Delta S(T)$ values from thermal measurements on a different sample extrapolate to zero around $T/T_c \approx 0.995 < 1$ [see Figs. 5(a)–(b)]. For both thermal and magnetic data the respective critical temperatures T_c were obtained by fitting the corresponding melting fields $H_m(T)$ to a power law, $H_m(T) = H_0(1 - T/T_c)^n$, with the fitting parameters H_0, T_c, and n [see Fig. 5(c)]. We observe that around $T/T_c \approx 0.985$ (i.e., at $H_{m0} \approx 0.36$ T for $\Theta = 0°$) and with increasing temperature, $\Delta M_0(T)$ starts deviating from the linear trend that extrapolates to zero at $T/T_c \approx 1$, thereby causing a kinklike feature in $\Delta M_0(T)$ [see Fig. 5(a)]. The fact that $\Delta S(T)$ vanishes below the temperature where the melting lines extrapolate to zero might indicate that the first-order character of the phase transition is lost at a lower critical point, that would be located around $\mu_0 H \approx 80$ mT in our crystals. However, the validity of this conclusion crucially depends on the definition of the critical temperature T_c. To demonstrate this, we tentatively ignore the upward curvature of the $H_{m0}(T)$ data obtained from the magnetic measurements [where $n \approx 1.35$ (Ref. 17)], and extrapolate $H_{m0}(T)$ above $T/T_c = 0.98$ *linearly* to zero [see Fig. 5(c)]. We find that with this new definition of T_c, the melting field becomes zero only slightly above the temperature where ΔS vanishes, making the assumption of the existence of a lower critical point perhaps unnecessary. This linear-extrapolation procedure for our $H_{m0}(T)$ data near T_c has no physical justification, however. We therefore define T_c in the following as the resulting value from a power-law fit to the $H_m(T)$ data as described above.

The temperature difference between T_c and the temperature where ΔS vanishes in our samples is $\approx$450 mK, which has to be compared with the width of the vortex-lattice melting transition and the width of the transition to superconductivity in zero magnetic field. We defined these transition widths as follows: Our analysis of the broadening of the steplike discontinuities in magnetic torque gives a full width of the vortex-lattice melting transition $\delta T \approx 50-70$ mK (see Sec. VI of this paper). From resistivity $\rho(H,T)$ data taken on a crystal of the same batch, in zero magnetic field and in $\mu_0 H \approx 1$ T at the vortex-lattice melting transition,[42] we can also estimate the widths δT_ρ of the respective transitions. For $H = 0$ we can fit $d\rho(T)/dT$ to a Gaussian with a full width δT_ρ and obtain $\delta T_\rho \approx 80$ mK. As an alternative, we can compare the well-defined temperatures where $d\rho/dT$ is maximum and where $\rho(T) = 0$. For both $H = 0$ (i.e., the transition to superconductivity) and $\mu_0 H \approx 1$ T (the vortex-lattice melting transition) we obtain in this way $\delta T_\rho \approx 100$ mK. The temperature difference between T_c and the temperature where ΔS vanishes ($\approx$450 mK) does not seem to be related in an obvious way to the above estimates for the ''chemical'' full width of the transition (50–100 mK). We therefore speculate that a lower critical point of $H_m(T)$ indeed does exist in our samples, that might be related to the amount and the character of disorder as recently shown in Ref. 43 for the

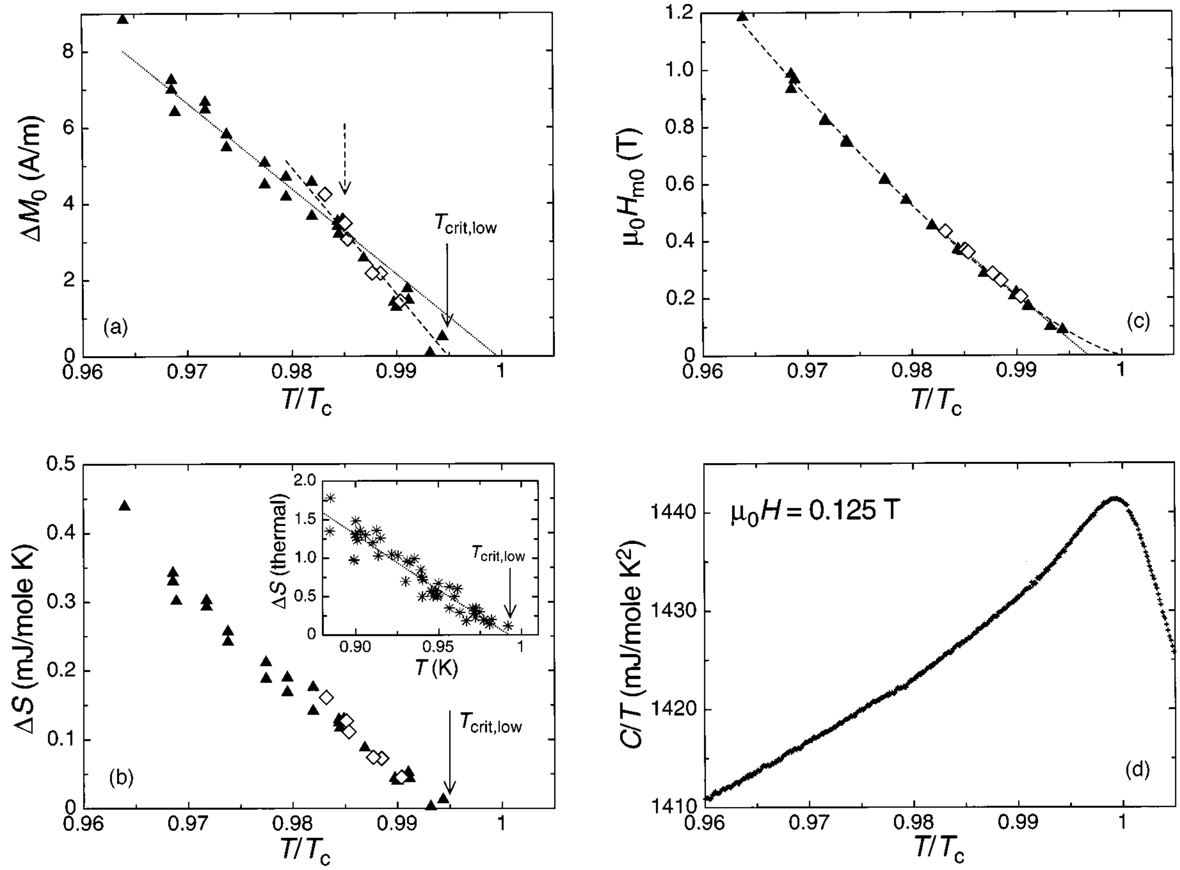

FIG. 5. (a) Magnetization discontinuities ΔM_0 for $H \| c$, obtained from the transverse components $\Delta M_{\rm trans}$ according to Eq. (A7), and plotted versus the reduced temperature. Triangles are the ΔM_0 data from Ref. 17, while diamonds correspond to additional data points from this work. The solid arrow indicates where the low-field $\Delta M_0(T)$ data extrapolate to zero. The dashed arrow shows where $\Delta M_0(T)$ changes its slope (see text). The dashed and the dotted lines are to guide the eye. (b) Discontinuities in entropy at the vortex-lattice melting transition, calculated from the data in Fig. 5(a) using the Clausius-Clapeyron equation (A4). The inset shows corresponding data from a direct thermal measurement on a different crystal (Ref. 26). The arrow indicates where $\Delta S(T)$ extrapolates to zero. (c) The melting fields from our magnetic experiments, reduced to $H \| c$ values H_{m0} using Eqs. (A1) and (A2) with $\gamma = 8.2$. The dashed line is a power-law fit to the data, while the dotted line represents a linear fit to the data above $T/T_c = 0.98$ (see text). (d) Specific-heat C/T vs T/T_c data for $\mu_0 H = 0.125$ T parallel to c. At this magnetic field and below (down to $\mu_0 H \approx 89$ mT parallel to c and $T/T_c = 0.994$) the first-order transition can still be observed magnetically, while C/T already shows a distinct upward curvature indicating the strong influence of fluctuations on thermodynamic quantities near T_c.

case of dilute columnar defects. It is worth mentioning here that in some moderately twinned or artificially detwinned $YBa_2Cu_3O_{7-\delta}$ crystals it was impossible to observe a first-order transition already in magnetic fields $\mu_0 H \approx 4$ T and lower, regardless of the sharpness of the transition to superconductivity and the high chemical purity of the samples.[19,21,22,28,29]

We have detected a discontinuity in the magnetic torque at temperatures as high as $T/T_c = 0.994$ (in $\mu_0 H \approx 89$ mT along c),[17] where the specific heat is already dominated by fluctuation effects, as demonstrated by the strongly upward curvature in C/T vs T in the same temperature and magnetic-field range [see Fig. 5(d)]. The first-order transition is obviously very robust against the occurrence of such fluctuations near T_c, which may be difficult to reconcile with the intuitive picture of the melting of an ordered crystal lattice of well-defined vortices so close to T_c. It can be explained in a natural way by interpreting the zero-field T_c as the critical point in a three-dimensional 3D-*XY* model, where all phase-transition lines within the critical region, including $H_m(T)$, eventually merge. Support for this interpretation comes from the excellent fit of our low-field $H_{m0}(T)$ data (i.e., $\mu_0 H < 1.2$ T) to a power law with $n \approx \frac{4}{3}$ (see above), which may be a consequence of the 3D-*XY* scaling of physical quantities in low magnetic fields and near T_c that has also been observed in other experiments.[11,44–50] These issues need further experimental clarification because they may question the validity of the common picture of a ''re-entrant'' melting line, that would bend over to lower temperatures with decreasing magnetic field and not terminate at T_c.[50,51]

Many experiments have indicated that in higher magnetic fields the first-order character of the transition is lost at an upper (multi-) critical point, the location of which is believed to depend on the anisotropy parameter γ and on the amount and the character of defects in the sample.[25,43,52–54] Unfortunately, the relevant magnetic fields are somewhat too high to do a systematic magnetic study on the upper critical point with our torque magnetometer. We have observed in thermal experiments on similar crystals that for $\Theta = 0°$ and above $\mu_0 H \approx 6.5$ T, $\Delta S(T)$ indeed starts to decrease with decreasing temperature, and finally vanishes around $\mu_0 H \approx 12.5$ T

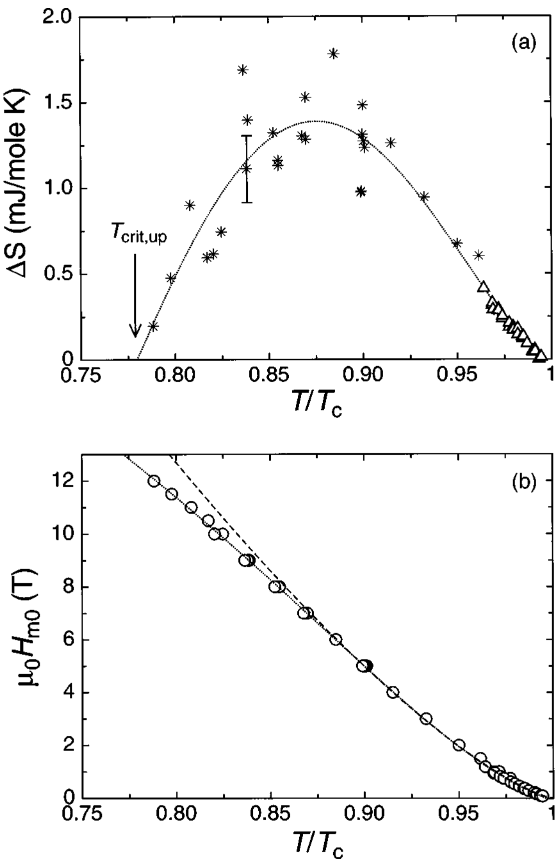

FIG. 6. (a) The discontinuities in entropy at vortex-lattice melting ΔS from both magnetic (triangles) and thermal experiments (stars). The arrow indicates where ΔS extrapolates to zero at $T/T_c = 0.78$, which corresponds to $\mu_0 H = 12.5$ T parallel to c. The dotted line is to guide the eye. The error bar represents the estimated error in ΔS for the data from thermal experiments. (b) The melting fields $H_{m0}(T)$ for $\Theta = 0°$ from both magnetic and thermal experiments. The dashed line is a power-law fit to the data below $\mu_0 H = 6$ T. The dotted line is to guide the eye and to illustrate the deviation of the $H_{m0}(T)$ data from a power-law behavior as the magnetic field approaches the upper critical point at $\mu_0 H = 12.5$ T.

[see Fig. 6(a)].[29,30] At the same time, with increasing magnetic field, the melting temperature is more and more shifted to lower temperatures when compared to an extrapolation of a low-field power-law fit to the $H_{m0}(T)$ data, thereby causing a slightly S-shaped melting curve with an inflection point around $\mu_0 H \approx 6.5$T [see Fig. 6(b)]. This behavior is, of course, not reproduced by a London theory that assumes a system without disorder.[33,41] A similar but somewhat much more pronounced feature in $H_m(T)$ has already been observed in the very anisotropic $Bi_2Sr_2CaCu_2O_8$.[10]

V. CHANGES IN $(\partial\tau/\partial H)_T$ AT THE VORTEX-LATTICE MELTING TRANSITION

Along with the steps in entropy and magnetization at the vortex-lattice melting transition, simultaneous steplike variations in the reduced specified heat $\Delta C/T$ have been observed in thermal experiments.[18,22–26] These changes represent the difference in specific heat between the vortex-fluid and the vortex-solid state, i.e., in C/T at temperatures right above and below the first-order transition, respectively. They are related to changes in slope in magnetization, $\Delta(\partial M/\partial H)_T$ and $\Delta(\partial M/\partial T)_H$, by a thermodynamic relationship.[24] In our experiments measuring the magnetic torque, $\boldsymbol{\tau} = \mu_0 \mathbf{M} \times \mathbf{H}$ per volume of sample, we also observe corresponding changes in slope, $\Delta(\partial\tau/\partial H)_T$ (see Fig. 1). While a thermodynamic consistency check between $\Delta(\partial M/\partial T)_H$ and $\Delta C/T$ data is rather straightforward for $H \parallel c$,[24] a transformation of corresponding $\Delta(\partial\tau/\partial H)_T$ data with $\Theta \neq 0°$ into thermal $\Delta C/T$ values is not as trivial (see Appendix). In Fig. 7(a) we show a representative set of $\Delta(\partial\tau/\partial H)_T$ data as a function of Θ for $T = 92.15$ K, indicating that this quantity strongly depends on the angle Θ. According to standard angular-dependent scaling rules for thermodynamic quantities in anisotropic superconductors, the associated changes $\Delta C/T$ at fixed T should not depend on Θ, however. In Fig. 7(b) we display the result of transforming our $\Delta(\partial\tau/\partial H)_T$ data into $\Delta C/T$ values by using Eq. (A24). We can conclude that at a given temperature T and within the scattering of the data, the resulting changes in reduced specific heat $\Delta C/T$ do not depend on the angle Θ, in a similar way as the entropy changes ΔS that also do not depend on Θ.[26,32,33] In Fig. 7(c) we compare the magnetically obtained $\Delta C/T$ data with the data from a direct thermal measurement on a similar crystal.[26,30] In a very contrast to specific-heat $C(T)$ data near the vortex-lattice melting transition, our field-dependent magnetic torque $\tau(H)$ data taken at fixed T do not exhibit any strong variation in the background signal that would give rise to a large uncertainty in choosing the correct model for fitting the data. Therefore we judge our ''magnetic'' $\Delta C/T$ data as more reliable than our thermal estimate for $\Delta C/T$ from a direct $C(T)$ measurement, the evaluation of which is strongly hampered by the presence of fluctuation effects near T_c.[26] It is clear that the magnitude of $\Delta C/T$ is of the order of 1 mJ/mole K^2 at temperatures as close as 0.99 K to T_c. This means that measured values of $\Delta C/T$ are at least twice as large as those predicted in Ref. 33, while the temperature dependence of $\Delta C/T$ may be correctly reproduced by the theory.

VI. ANGULAR SCALING OF THE MELTING FIELD AND OF THE TRANSITION WIDTH

Many experiments have confirmed that the standard scaling rules for thermodynamic quantities in anisotropic superconductors are valid for $YBa_2Cu_3O_{7-\delta}$ (Refs. 3, 6, 24, 26), which is only moderately anisotropic. This is not necessarily the case for a very anisotropic compound such as $Bi_2Sr_2CaCu_2O_8$. If the layered structure of a superconductor becomes important, anisotropy effects may perhaps no longer be described by a single, temperature and field-independent anisotropy parameter γ that is derived from an anisotropic effective-mass model.[31,34] Nevertheless, this parameter γ can be rather accurately determined for $YBa_2Cu_3O_{7-\delta}$ in the superconducting state, e.g., by scaling the melting fields $H_m(T,\Theta)$ or the melting temperatures $T_m(H,\Theta)$,[3,6,24,26,38] or by fitting the reversible magnetic torque $\tau(\Theta)$ to a suitable model.[55,56] This latter method is very simple and does not require the high instrumental precision that is necessary to detect the first-order melting transition, but it cannot be applied at temperatures too close to

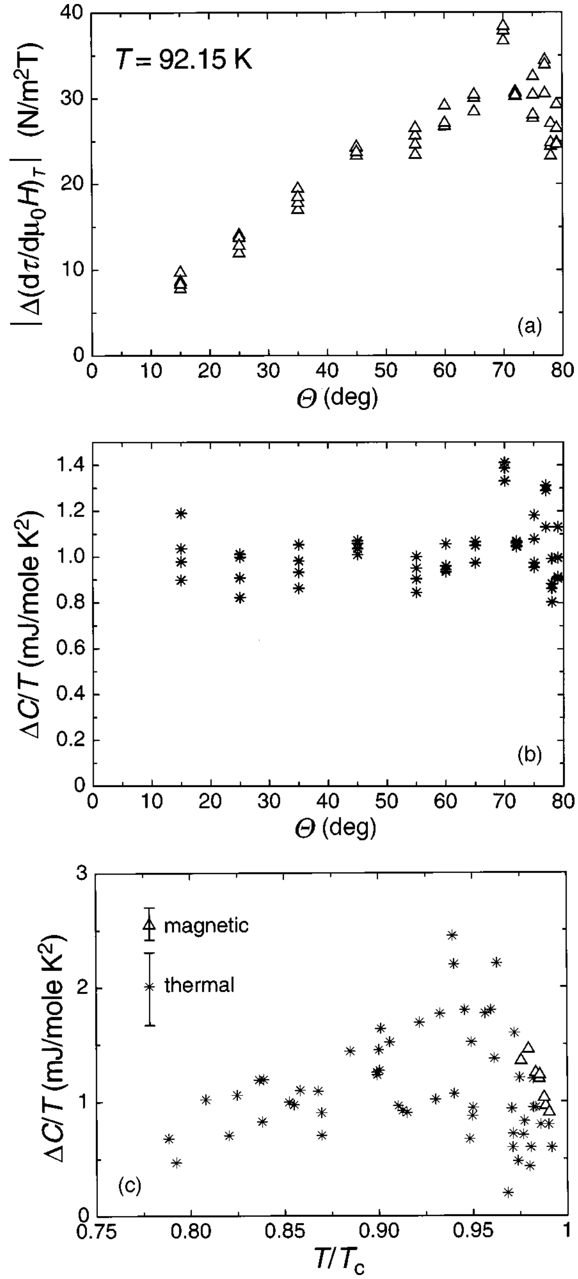

FIG. 7. (a) Changes in slope in the magnetic torque $\tau(H)$ at the vortex-lattice melting transition, $|\Delta(\partial\tau/\partial\mu_0 H)_T|$, as a function of Θ for $T = 92.15$ K. (b) The difference in the specific heat between the vortex-fluid and the vortex-solid state, $\Delta C/T$, calculated from the data from Fig. 7(a) using Eq. (A24). (c) Comparison between our magnetic estimates (triangles, see Figs. 7(a)–(b)) and direct thermal measurements of $\Delta C/T$ (stars) for different temperatures. The error bars represent the estimated errors for the respective sets of data.

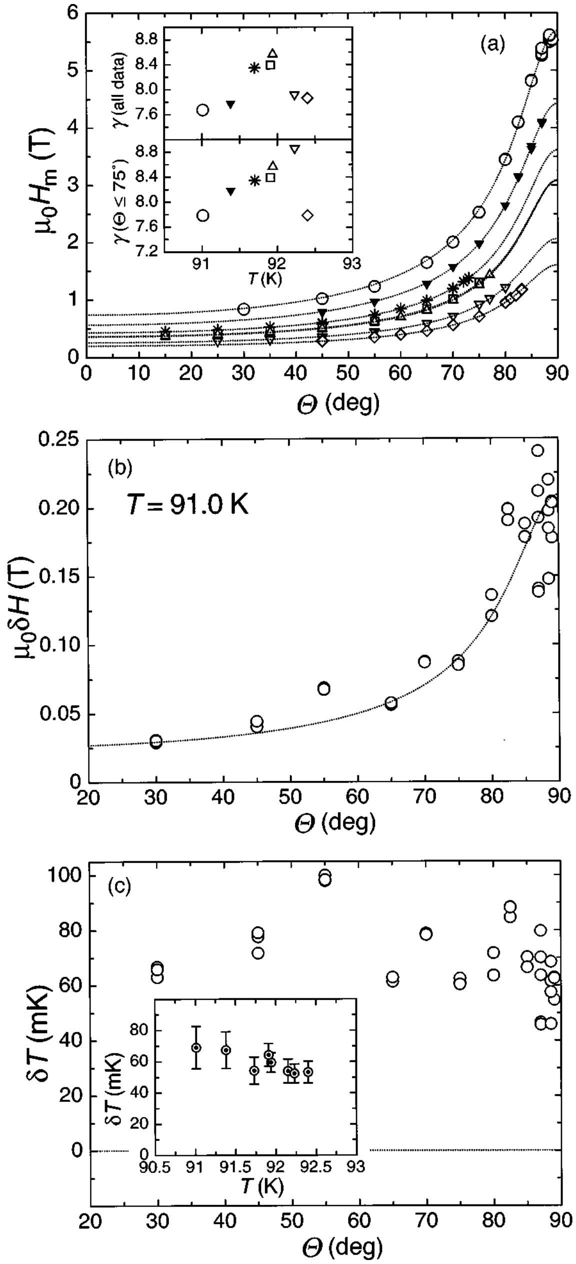

FIG. 8. (a) The melting fields $H_m(\Theta)$ for different temperatures near T_c. The dotted lines correspond to fits according to Eqs. (A1) and (A2) to obtain values for the anisotropy parameter γ (upper inset). The lower inset shows γ values that we obtained by fitting the data for $\Theta \leqslant 75°$ only. (b) Total transition width δH (i.e., the full width according to a Gaussian distribution of first-order transitions), as a function of the angle Θ for $T = 91.0$ K. The dotted line corresponds to a scaling of δH according to Eq. (A2) with $\gamma = 8.2$. (c) Transition widths in temperature δT for $T = 91.0$ K. Within the experimental scatter, δT is independent of Θ. Inset: The respective mean values of the transition widths δT as a function of temperature.

the critical temperature T_c where fluctuation effects lead to a pronounced distortion of the mean-field torque signal.[57] We were able to measure the angular dependence of the vortex-lattice melting transition at temperatures as high as $T/T_c = 0.99$, which allows us to test the validity of the standard scaling rules for $H_m(T,\Theta)$ up to this temperature.

In Fig. 8(a) we show the corresponding melting fields $H_m(T,\Theta)$ for different temperatures. Each curve can be fitted according to Eqs. (A1) and (A2), and we obtain an anisotropy parameter γ for each temperature. The result of this analysis is shown in the upper panel of the inset of Fig. 8(a). To first approximation, all the melting fields $H_m(T,\Theta)$ scale very well up to $T/T_c = 0.99$. In order to test whether or not the weak variation in γ [as shown in the inset of Fig. 8(a)] is significant or not, we examined to what extent changing the range of fitting a set of $H_m(T,\Theta)$ data influences the resulting value of γ. A simultaneous linear fit with varying fitting range to all the available data reveals that the parameter γ weakly depends on the selected range of angles Θ. For $0° < \Theta \leqslant 75°$ we obtain $\gamma = 8.09$, for $\Theta \leqslant 85°$ we calculate $\gamma = 7.91$, and for the full range of data we have an optimum $\gamma = 7.69$. We therefore tentatively fixed the range of angles for fitting the data to $\Theta \leqslant 75°$ for all temperatures. The resulting values for γ are plotted in the lower panel of the inset of Fig. 8(a), and they still show a certain change with temperature. However, all these variations of γ are within a standard deviation of only $\pm 5\%$ around the mean value $\gamma = 8.2$. Therefore, there are no profound consequences on the conclusions that we have drawn in Secs. III–V of this paper, and we can confirm that scaling the melting fields $H_m(T,\Theta)$ is a reliable method to determine γ near T_c.

Finally, we want to analyze the width of the vortex-lattice melting transition in $YBa_2Cu_3O_{7-\delta}$, that, according to common interpretation schemes, reflects the chemical homogeneity of the sample. Many previous experiments on $YBa_2Cu_3O_{7-\delta}$ single crystals of different sources indicated that the respective total transition widths (usually defined as the width of the first-order peak in specific-heat measurements) are rather narrowly distributed around a seemingly common value of the order of $\delta T = 200$ mK.[11,12,19–28] This observation gave rise to suggestions that a virtually constant δT (more precisely, a δT that depends very weakly on sample size) may be a consequence of a peculiar underlying mechanism that also produces the observed discontinuities in magnetization and in entropy, but that does not invoke a thermodynamic first-order transition.[58] In Fig. 8(b) we show the total transition width (according to a Gaussian distribution of first-order transitions with a half-width $\delta H/2$), as a function of the angle Θ for $T = 91.0$ K. If the broadening of the transition were of a chemical nature leading to a total width δT in temperature, the resulting magnetic full-widths δH should scale as $\delta H = \delta T |dH_m(T,\Theta)/dT|$, i.e., according to the scaling function (A2). Using $\mu_0 dH_m(T,0)/dT = -0.4$ T/K at this temperature, we calculate with (A2) and $\gamma = 8.2$ the δT values shown in Fig. 8(c), that are independent of Θ within the experimental scatter. In the inset of the same figure we have plotted the respective mean values of the transition widths δT as a function of temperature. The resulting averaged value, $\delta T \approx 60$ mK, is significantly smaller than those reported for earlier samples. We therefore conclude that δT in $YBa_2Cu_3O_{7-\delta}$ indeed most likely depends mainly on sample quality, and that the observed broadening of the melting transition is of chemical origin. We expect that any additional broadening of the transition due to a geometry-induced inhomogeneous flux distribution amounts to much less than $M|dH_m|dT|^{-1} \approx 3$ mK for $T = 91$ K in $YBa_2Cu_3O_{7-\delta}$, but it may become relevant in a strongly anisotropic compound such as $Bi_2Sr_2CaCu_2O_8$.

VII. CONCLUSIONS

We have performed a detailed study on the angular dependence of the discontinuity in magnetization at the vortex-lattice melting transition in $YBa_2Cu_3O_{7-\delta}$. We have shown that within the accuracy of our measurement, the vector $\Delta\mathbf{M}$ is always parallel to the magnetization $\mathbf{M}$ itself, even for large angles Θ between the applied magnetic field and the c direction of the crystal, where $\mathbf{M}$ is already significantly tilted away from the c direction. The smallest magnetic field at which the first-order transition is still detectable within the resolution of our experiments is $\mu_0 H \approx 89$ mT along the c direction at $T/T_c = 0.994$, where other physical quantities are already strongly affected by fluctuation effects. The discontinuities $\Delta\mathbf{M}$ and ΔS vanish slightly below the temperature where $H_m(T)$ extrapolates to zero, which could indicate the existence of a lower critical point of the vortex-lattice melting line in our samples, where the first-order character of the transition is lost. Using an appropriate thermodynamic relationship we can convert the changes in slope in magnetic torque at the melting transition, $\Delta(\partial\tau/\partial H)_T$, to differences in specific heat $\Delta C/T$ between the vortex-fluid and the vortex-solid phase. The thus obtained data agree well with thermal experiments, but also suggest that $\Delta C/T$ is somewhat larger than predicted by theory. We have shown that the melting fields $H_m(T,\Theta)$ scale, to first approximation, very well according to scaling rules for anisotropic superconductors up to temperatures $T/T_c = 0.99$.

ACKNOWLEDGMENTS

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APPENDIX

1. Measuring the Direction of the Magnetization-Discontinuity Vector ΔM

In this paragraph we want to show that the direction of the magnetization-discontinuity vector $\Delta\mathbf{M}$ at the vortex-lattice melting transition can be determined by measuring its transverse component alone, if we assume that standard angular-dependent scaling rules for thermodynamic quantities in anisotropic superconductors do apply. We also consider how a possible change in the direction of the magnetization vector $\mathbf{M}$ at the transition would influence other measurable scalar magnetic quantities, such as the longitudinal component of $\Delta\mathbf{M}$, or its projection along the c axis of the crystal.

A possible scenario is sketched in Fig. 9. We choose the axes in a way that the unit vector $\mathbf{e}_z$ is directed along c, with an angle Θ between the magnetic field $\mathbf{H}$ and $\mathbf{e}_z$. The vectors $\mathbf{H}$ and $\mathbf{M}$ are chosen to be in the xz plane. We ignore here the effect of a possible in-plane anisotropy that would make a distinction between x and y (or a and b, respectively) necessary. We assume that the magnetization vector discontinuously jumps from $\mathbf{M}_1$ (solid phase, with an angle φ_1 between $\mathbf{M}_1$ and $-\mathbf{e}_z$) to $\mathbf{M}_2$ (fluid phase, with a respective angle φ_2) at the melting transition, and rotates by $\Delta\varphi = \varphi_2 - \varphi_1$ (here and in the following, Δ denotes a difference between the high-temperature fluid and the low-temperature solid phases). If $\Delta\varphi \neq 0$ the rotation would lead to a $\Delta\mathbf{M} = \mathbf{M}_2$

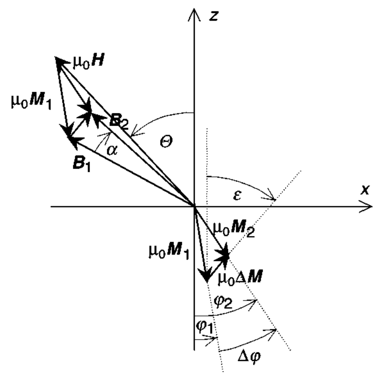

FIG. 9. Sketch of the situation for a magnetic first-order transition in a tilted magnetic field. The magnetization jumps discontinuously from $\mathbf{M}_1$ to $\mathbf{M}_2$, which might, in principle, lead to a rotation of the magnetization vector $\mathbf{M}$ at the transition. If the discontinuity $\Delta\mathbf{M}$ is parallel to $\mathbf{M}$, we expect to have $-\varepsilon=\varphi_1=\varphi_2$.

$-\mathbf{M}_1$ that is neither parallel to $\mathbf{M}_1$ nor to $\mathbf{M}_2$. The magnetic-flux density $\mathbf{B}$ would show a rotation by an angle α. The angle ε between $\Delta\mathbf{M}$ and $\mathbf{e}_z$ is unknown, and will be extracted from our experimental data. If $\Delta\mathbf{M}$ were parallel to $\mathbf{M}_1$, we would have $\varepsilon=-\varphi_1$, and $\Delta\varphi=0$. In $YBa_2Cu_3O_{7-\delta}$, the absolute value of the melting field $H_m(T,\Theta)$ at fixed T scales as

$$H_m(T,\Theta)=H_{m0}(T)f(\Theta), \tag{A1}$$

with

$$f(\Theta)=\frac{\gamma}{[\sin^2(\Theta)+\gamma^2\cos^2(\Theta)]^{1/2}}, \tag{A2}$$

where $\gamma\geqslant 1$ is the anisotropy ratio as defined in the main text of the paper, and $H_{m0}(T)$ is the melting field for $\Theta=0°$.[31] The magnetic torque per sample volume

$$\boldsymbol{\tau}=\mu_0\mathbf{M}\times\mathbf{H} \tag{A3}$$

is a vector along the unit vector $+\mathbf{e}_y$. Angular-dependent scaling rules for thermodynamic quantities in moderately anisotropic superconductors such as $YBa_2Cu_3O_{7-\delta}$ require that the step in entropy at vortex-lattice melting, ΔS, depends only on T, but not on the angle Θ.[31,32] This has been confirmed by thermal experiments.[26] Therefore, the Clausius-Clapeyron equation

$$\begin{aligned}\Delta S&=-\mu_0\Delta\mathbf{M}\cdot\frac{d\mathbf{H}_m}{dT}\\&=+\mu_0|\Delta M|\left|\frac{dH_m}{dT}\right|\cos(\Theta+\varepsilon)=+\mu_0|\Delta M_0|\left|\frac{dH_{m0}}{dT}\right|\end{aligned} \tag{A4}$$

gives a useful relationship between the absolute values of the vectors $\Delta\mathbf{M}$ and $d\mathbf{H}_m/dT$ and the respective values ΔM_0 and dH_{m0}/dT for $\Theta=0°$. The quantities $\Delta M_0(T)$ and $H_{m0}(T)$ are well known from experiments.[11,12,17–29] Using (A2) and (A4) we obtain

$$\Delta M=\frac{\Delta M_0}{f(\Theta)\cos(\Theta+\varepsilon)}. \tag{A5}$$

Note that in our situation $|d\mathbf{H}_m/dT|>0$, while $dH_m/dT<0$. The longitudinal component (parallel to $\mathbf{H}$) becomes

$$\Delta M_{\mathrm{long}}=e_H\cdot\Delta\mathbf{M}=\frac{\Delta M_0}{f(\Theta)}, \tag{A6}$$

with the unit vector $\mathbf{e}_H$ along $\mathbf{H}$. It does not depend on ε, and a possible rotation of $\mathbf{M}$ cannot be detected in an experiment probing only ΔM_{long}. The transverse component perpendicular to $\mathbf{H}$ can be experimentally determined by detecting the discontinuity in magnetic torque per sample volume, $|\Delta\tau|=|\mu_0\Delta M_{\mathrm{trans}}H|$, and is

$$\Delta M_{\mathrm{trans}}=\frac{\Delta M_0}{f(\Theta)}\tan(\Theta+\varepsilon), \tag{A7}$$

which depends on ε. A rotation of $\mathbf{M}$ can therefore be detected by measuring $\Delta M_{\mathrm{trans}}$, e.g., in a sensitive transverse superconducting quantum interference device arrangement or in a magnetic-torque experiment.

For the component parallel to c we calculate

$$\Delta M_c=\frac{\Delta M_0\cos(\varepsilon)}{f(\Theta)\cos(\Theta+\varepsilon)}=\frac{\Delta M_0}{f(\Theta)[\cos(\Theta)-\tan(\varepsilon)\sin(\Theta)]}. \tag{A8}$$

This component, that is measured in local Hall-array experiments, also depends on ε. In our magnetic-torque experiment, we can detect $|\Delta\tau|$ at fixed temperature T and for a series of angles Θ. For each Θ, $\Delta\tau$ will occur at the melting field $H=H_m(\Theta)$. The angle ε can then be calculated from $\Delta M_{\mathrm{trans}}$ using Eqs. (A2) and (A7) if the corresponding values ΔM_0 and γ are known from another experiment. In practice we can rely on the fact that for small values of Θ, $\Delta\mathbf{M}$ is virtually parallel to c,[34] and

$$|\Delta M_0|\approx\frac{|\Delta\tau|}{\mu_0 H_m\sin(\Theta)}.\quad \Theta\to 0. \tag{A9}$$

The anisotropy parameter γ can be easily obtained in the same experiment by scaling the measured melting fields $H_m(\Theta)$ at fixed T according to Eqs. (A1) and (A2).
The vector $\mathbf{B}$ will also show a slight rotation by the angle α. Since in $YBa_2Cu_3O_{7-\delta}$, $\mu_0 M\ll B$ we can use $\mu_0 H\approx B$, and with Eq. (A7) we obtain

$$\frac{2\pi\alpha}{360°}\approx\frac{|\Delta M_{\mathrm{trans}}|}{H_m}=\frac{|\Delta M_0|}{H_m f(\Theta)}\tan(\Theta+\varepsilon)=\frac{|\Delta\tau|}{\mu_0 H_m^2}. \tag{A10}$$

2. Difference in the Specific Heat between the Vortex-Fluid and the Vortex-Solid Phases

In this section we will calculate the difference in the reduced specific heat $\Delta C/T$ between the vortex-fluid and the vortex-solid phases, expressed by the measured changes in slope $\Delta(\partial\tau/\partial H)_T$ of the magnetic torque per sample volume at the vortex-lattice melting transition, and other magnetic quantities that are known from experiments. The situation is complicated by the fact that the magnetic torque per sample volume, $\boldsymbol{\tau}=\mu_0\mathbf{M}\times\mathbf{H}$, is a vector, while experiments only give the absolute value $|\tau|$. Therefore one has to carefully consider the geometry of the problem, and to check the signs

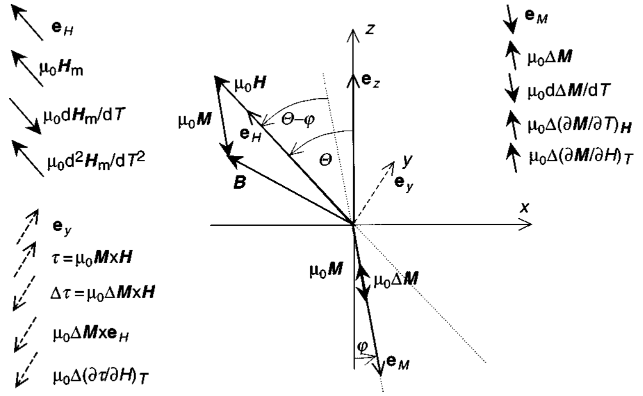

FIG. 10. Sketch of the same situation as in Fig. 9, but for $\Delta\mathbf{M}$ parallel to $\mathbf{M}$. The arrows indicate the direction of the respective vectors in space that are used for the calculation of the difference in the specific heat between the vortex-fluid and the vortex-solid state, $\Delta C/T$, from $\Delta(\partial\tau/\partial H)_T$ data (see Appendix).

of all scalar quantities that are related to vectors such as $\mathbf{M}$, $\Delta\mathbf{M}$, $\boldsymbol{\tau}$, $\Delta\boldsymbol{\tau}$, $\mathbf{H}_m$, and their derivatives (see Fig. 10). We already include here the experimental result that $\Delta\mathbf{M}$ is parallel to $\mathbf{M}$, i.e., $\varphi_1=\varphi_2=\varphi=-\varepsilon$. The angle φ can be calculated from Eq. (1) in the main text.

The derivative $(\partial\boldsymbol{\tau}/\partial H)_T$ is

$$\left(\frac{\partial\boldsymbol{\tau}}{\partial H}\right)_T=\mu_0\left(\frac{\partial\mathbf{M}}{\partial H}\right)_T\times\mathbf{H}+\mu_0\mathbf{M}\times\mathbf{e}_H \tag{A11}$$

(with the unit vector $\mathbf{e}_H$ along $\mathbf{H}$), with a change

$$\Delta\left(\frac{\partial\boldsymbol{\tau}}{\partial H}\right)_T=\mu_0\Delta\left(\frac{\partial\mathbf{M}}{\partial H}\right)_T\times\mathbf{H}\times\mu_0\Delta\mathbf{M}\times\mathbf{e}_H \tag{A12}$$

at the phase transition. To substitute the derivative $(\partial\mathbf{M}/\partial H)_T$ we calculate

$$\frac{d\Delta\mathbf{M}}{dT}=\left(\frac{\partial\Delta\mathbf{M}}{\partial T}\right)_H+\left(\frac{\partial\Delta\mathbf{M}}{\partial H}\right)_T\frac{dH_m}{dT} \tag{A13}$$

for a fixed Θ, where $\Delta\mathbf{M}(T)$ is the step in magnetization as a function of the melting temperature T. We note that the changes in the slope of $\mathbf{M}$ at the transition are identical to the corresponding slopes of the difference $\Delta\mathbf{M}$, i.e.,

$$\Delta\left(\frac{\partial\mathbf{M}}{\partial T}\right)_H=\left(\frac{\partial\Delta\mathbf{M}}{\partial T}\right)_H \quad\text{and}\quad \Delta\left(\frac{\partial\mathbf{M}}{\partial H}\right)_T=\left(\frac{\partial\Delta\mathbf{M}}{\partial H}\right)_T. \tag{A14}$$

From Eq. (A12) we obtain

$$\Delta\left(\frac{\partial\mathbf{M}}{\partial H}\right)_T=-\frac{|\Delta(\partial\boldsymbol{\tau}/\partial H)_T-\mu_0\Delta\mathbf{M}\times\mathbf{e}_H|}{\mu_0 H\sin(\Theta-\varphi)}\mathbf{e}_M \tag{A15}$$

with the unit vector $\mathbf{e}_M$ along $\mathbf{M}$. With Eqs. (A12)–(A15) we calculate

$$\frac{d\Delta\mathbf{M}}{dT}=\Delta\left(\frac{\partial\mathbf{M}}{\partial T}\right)_H-\frac{|\Delta(\partial\boldsymbol{\tau}/\partial H)_T-\mu_0\Delta\mathbf{M}\times\mathbf{e}_H|}{\mu_0 H\sin(\Theta-\varphi)}\cdot\frac{dH_m}{dT}\cdot\mathbf{e}_M. \tag{A16}$$

The change in reduced specific heat, $\Delta C/T$, is given by

$$\Delta\frac{C}{T}+\mu_0\Delta\left(\frac{\partial\mathbf{M}}{\partial T}\right)_H\cdot\frac{d\mathbf{H}_m}{dT}+\mu_0\frac{d\Delta\mathbf{M}}{dT}\cdot\frac{d\mathbf{H}_m}{dT}+\mu_0\Delta\mathbf{M}\cdot\frac{d^2\mathbf{H}_m}{dT^2}=0. \tag{A17}$$

This formula is an extension of an Ehrenfest relationship (used to describe second-order phase transitions where $\Delta\mathbf{M}=0$) for phase transitions that are of first order.[24] It can be obtained by using the fact that the difference in the free energy between the fluid and the solid states, ΔG, is zero along $H_m(T)$, and therefore $d^2\Delta G/dT^2=0$ [note that $d\Delta G/dT=0$ yields the Clausius-Clapeyron equation (A4)]. Combining (A16) and (A17) we obtain

$$\Delta\frac{C}{T}=-2\mu_0\frac{d\Delta\mathbf{M}}{dT}\cdot\frac{d\mathbf{H}_m}{dT}-\frac{|\Delta(\partial\boldsymbol{\tau}/\partial H)_T-\mu_0\Delta\mathbf{M}\times\mathbf{e}_H|}{H\sin(\Theta-\varphi)}\cdot\frac{dH_m}{dT}\cdot\frac{d\mathbf{H}_m}{dT}\cdot\mathbf{e}_M-\mu_0\Delta\mathbf{M}\cdot\frac{d^2\mathbf{H}_m}{dT^2}. \tag{A18}$$

To eliminate all the vectors in (A18), we first write

$$\tau=\mu_0|M|H\sin(\Theta-\varphi)>0, \tag{A19}$$

(i.e., $\boldsymbol{\tau}$ is directed along $+\mathbf{e}_y$) and the difference

$$\Delta\tau=-\mu_0|\Delta M|H\sin(\Theta-\varphi)<0 \tag{A20}$$

(along $-\mathbf{e}_y$). With (A14) and $|\partial\Delta\mathbf{M}/\partial H|=\partial|\Delta\mathbf{M}|/\partial H$ we have

$$\Delta\left(\frac{\partial\tau}{\partial H}\right)_T=-\mu_0\left(\left|\Delta\left(\frac{\partial M}{\partial H}\right)_T\right|H+\left|\Delta M\right|\right)\sin(\Theta-\varphi)<0 \tag{A21}$$

along $-\mathbf{e}_y$. The negative signs of both $\Delta\tau$ and $\Delta(\partial\tau/\partial H)_T$ are observed in our experiments (see Fig. 1).

The cross product in (A18) is a vector with the absolute value

$$|\Delta\mathbf{M}\times\mathbf{e}_H|=|\Delta M|\sin(\Theta-\varphi), \tag{A22}$$

that is also directed along $-\mathbf{e}_y$. Therefore,

$$\begin{aligned}\left|\Delta\left(\frac{\partial\boldsymbol{\tau}}{\partial H}\right)_T-\mu_0\Delta\mathbf{M}\times\mathbf{e}_H\right| &=\left|-\left|\Delta\left(\frac{\partial\tau}{\partial H}\right)_T\right|+\mu_0\left|\Delta M\right|\sin(\Theta-\varphi)\right|\\ &=\left|\left|\Delta\left(\frac{\partial\tau}{\partial H}\right)_T\right|-\frac{\mu_0|\Delta M_0|}{f(\Theta)}\tan(\Theta-\varphi)\right|.\end{aligned} \tag{A23}$$

Inserting Eqs. (A2), (A5), and (A23) into Eq. (A18) we obtain the final result

$$\frac{\Delta C}{T}=-2\mu_0\left|\frac{d|\Delta M_0|}{dT}\right|\left|\frac{dH_{m0}}{dT}\right|-\mu_0|\Delta M_0|\left|\frac{d^2H_{m0}}{dT^2}\right| +\frac{f(\Theta)^2}{H}\left|\left|\Delta\left(\frac{\partial\tau}{\partial H}\right)_T\right|\cot(\Theta-\varphi)-\frac{\mu_0|\Delta M_0|}{f(\Theta)}\right| \times\left(\frac{dH_{m0}}{dT}\right)^2. \tag{A24}$$

This formula is valid only if all the relevant vectors are directed as indicated in Fig. 10. In other situations, one has to reconsider the signs of some of the terms in (A24). In a different physical context (e.g., for a first-order transition in other anisotropic magnetic materials) one has also to use an appropriate scaling function $f(\Theta)$, and needs to verify that $\Delta S(\Theta)$ scales according to similar rules as in anisotropic superconductors.

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