First-Order Vortex-Lattice Melting Transition in $YBa_2Cu_3O_{7-\delta}$ near the Critical Temperature Detected by Magnetic Torque

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High-resolution magnetic-torque studies on an untwinned $YBa_2Cu_3O_{7-\delta}$ single crystal near its critical temperature T_c reveal that the first-order vortex-lattice melting transition (VLMT) persists at least up to 0.5 K below T_c. The associated sharp discontinuity in magnetization is detectable even at temperatures where the torque signal deviates from mean-field behavior due to fluctuations. The magnetic irreversibility at the VLMT can be suppressed by applying a weak transverse ac magnetic field. This offers the possibility of separating the irreversibility line from the melting line near T_c. [S0031-9007(98)07569-3]

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Recent experimental work on the vortex-lattice melting in high-temperature superconductors clearly showed that the melting transition from the ordered vortex solid to a vortex-fluid phase is of first order [1–16]. Earlier muon-spin rotation [1] and neutron-scattering [2] experiments on $Bi_2Sr_2CaCu_2O_8$ (BSCCO) revealed an abrupt change in the local magnetic flux distribution and the diffracted intensity, respectively, at the vortex-lattice melting transition (VLMT), indicative of a first-order transition. Similarly, a discontinuity ΔM in the magnetization M at the VLMT was observed for single crystals of $YBa_2Cu_3O_{7-\delta}$ (YBCO) [5,6,9], BSCCO [3,4,16,17], and $(La,Sr)_2CuO_4$ [15], by using SQUID magnetometers [3,5,6,9,15,16], local Hall arrays [4], and torque magnetometry [17]. Specific-heat data that directly measure a related jump in the entropy ΔS are, so far, available only for YBCO [7–14].

Despite the considerable resolution of all of the techniques used to measure ΔM and ΔS, there are very controversial results about the lower-field part of the melting line (ML) $H_m(T)$ in YBCO. In some cases, a VLMT was observed only for large magnetic fields $\mu_0 H > 4$ T [10,11,14], in other cases it was no longer detectable below 0.75 T [6,13]. Earlier magnetic-torque measurements on untwinned YBCO showed no signature of a VLMT at all [18]. Thus it has not yet been possible to explore the magnetic phase diagram of YBCO very near T_c, where thermodynamic fluctuations and a possible vortex-loop unbinding mechanism [19] are expected to modify ΔS and even the phase diagram itself. In spite of a recent theoretical work suggesting that $H_m(T)$ of YBCO ends at an intrinsic lower end point at about $\mu_0 H \approx 0.5$ T [20], it is conceivable that the absence of a first-order transition in YBCO at low magnetic fields is simply due to pinning [10,11,14] or to a broadening of the superconducting transition due to sample inhomogeneities [6,9,13]. Moreover, it is still not clear from the existing data whether the onset of magnetic irreversibility (i.e., the "irreversibility line") in YBCO coincides with the ML or not [6].

In this Letter we demonstrate the existence of the VLMT in high-quality YBCO very close to T_c using torque magnetometry. This sets a new upper limit for an intrinsic end point of the VLMT. Besides we show that the duality between melting and irreversibility near T_c is not always valid. The experimental setup used in this work is described in detail in Ref. [21]. The sample was a tiny (380 μg) untwinned YBCO single crystal ($T_c = 93.3$ K) of very high quality with a transition width $\delta T_c \approx 40$ mK, which is considerably smaller than in earlier works [5–14].

In an external magnetic field $\mathbf{H}$, a sample with an anisotropic magnetization $\mathbf{M}$ and volume V experiences a magnetic torque $\boldsymbol{\tau} = V\mathbf{M} \times \mu_0 \mathbf{H}$. This torque can be measured by means of a microfabricated flexible cantilever. Recent improvements in the design of torque sensors and of corresponding detection electronics have led to an optimized sensitivity $\Delta\tau \leq 5 \times 10^{-13}$ N m [22]. In an external magnetic field of 1 T, one routinely reaches a resolution of the order of $\sim 10^{-13}$ A m^2, which is considerably higher than that of a commercial SQUID magnetometer.

Figure 1a shows a typical set of torque vs field data taken at $T = 90.7$ K. The external magnetic field $\mathbf{H}$, applied in the ac plane at an angle $\vartheta = 45°$ with respect to the a direction, was swept up and down at a rate $\mu_0 dH/dt$ varying between 0.1 and 30 mT/s. For each sweep rate a clear steplike feature in $\tau(H)$ was observed, corresponding to an abrupt decrease in the magnitude of the magnetization M by an amount ΔM (>0) that we attribute to the VLMT. At magnetic fields below and slightly above the step, the torque signal becomes hysteretic (irreversibility) and shows clearly more intrinsic noise below the VLMT. It is very plausible that, below

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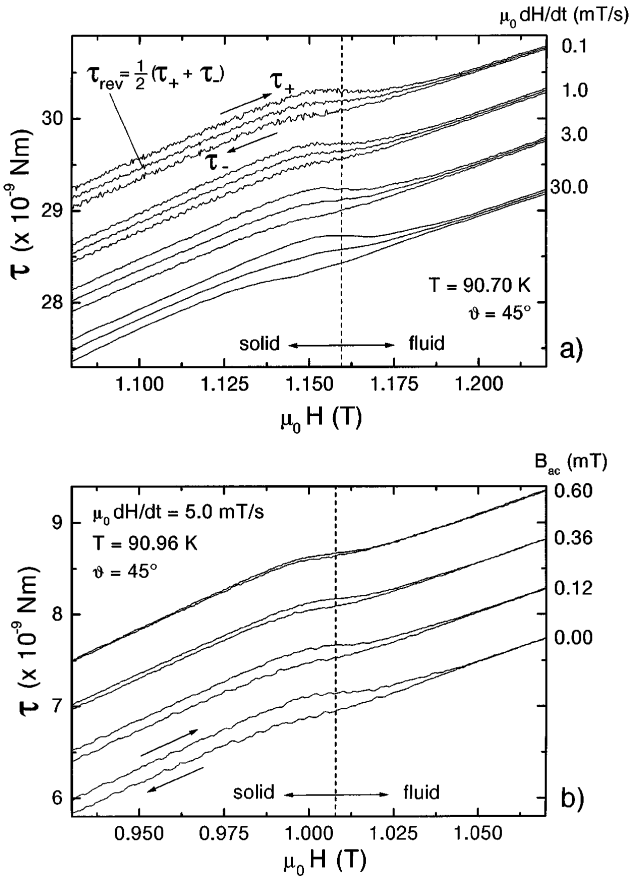

FIG. 1. The phase transition associated with vortex-lattice melting (dashed vertical line) as observed by field-dependent torque measurements on an untwinned single crystal of YBCO. (a) Magnetic torque τ for increasing (τ_+) and decreasing (τ_-) magnetic field $\mu_0 H$ for different sweep rates $\mu_0 dH/dt$, taken at 90.70 K and $\vartheta = 45°$. The data reveal a constant magnetization jump ΔM at the VLMT, determined from the reversible torque curve $\tau_{\rm rev}$. (b) Similar measurements (90.96 K, $\vartheta = 45°$) with an additional weak transverse ac magnetic field $B_{\rm ac}$ show a progressive suppression of irreversibility, the VLMT itself remaining unaffected. In both figures, each set of data is vertically shifted for clarity.

the VLMT, magnetic flux discontinuously enters or leaves the pinned vortex solid in successive bundles as the field is swept up or down, generating small stochastic vibrations of the sample and the cantilever. This effect does not occur in the vortex-fluid phase, because magnetic flux can freely penetrate or exit the sample in the absence of pinning effects. The reduction of the noise at higher sweep rates is simply a consequence of electrical filtering of the signal with a time constant of the order of $\sim$0.1 s, becoming comparable to the time it takes for the magnetic field to sweep through the region of interest.

The hysteresis width just below the VLMT corresponds to a critical-current density $J_c \approx 5$ A/cm^2, assuming that Bean's critical-state model is valid [23]. We observed that at fixed temperature the torque irreversibility is field independent over a wide range of external magnetic field (e.g., down to $\mu_0 H \approx 0.5$ T at $T = 90.7$ K and $\vartheta = 45°$). Thus the critical-current density J_c has a hyperbolic magnetic-field dependence over a wide range. The same behavior was seen also at smaller angles ϑ between **H** and the *ab* plane of the crystal. These observations, together with the fact that $H \gg H_{c1}$, rule out the occurrence of magnetic hysteresis generated by geometrical and surface barriers, as reported earlier in BSCCO for $\mathbf{H} \parallel c$ [17,24]. Therefore we can state here that the onset of bulk pinning in YBCO coincides with the VLMT; i.e., the irreversibility line and the ML collapse to a single line in the absence of external perturbations.

The application of an additional external ac magnetic field perpendicular to the main magnetic field has been demonstrated to enhance the vortex-lattice relaxation towards the thermodynamic equilibrium, even in the *a priori* irreversible vortex-solid state [25]. Consequently such an ac field should influence dramatically the magnetic irreversibility, but leave the VLMT unaffected. This is clearly demonstrated in Fig. 1b, with torque data taken at $T = 90.96$ K, $\vartheta = 45°$, at a sweep rate $\mu_0 dH/dt = 5$ mT/s, and in a perpendicular ac magnetic field of varying amplitude $B_{\rm ac}$ and fixed frequency $f = 1$ kHz. With increasing $B_{\rm ac}$ the torque curves become more and more reversible, and finally essentially collapse onto one single curve, still showing the first-order-like step in the torque signal. However, a hysteretic "bubble" still remains visible in the vicinity of the VLMT, before vanishing for $B_{ac} \approx 1$ mT. A slight increase of the hysteresis width of the torque signal near the transition is also observed at zero ac field (Fig. 1a). The observation of this "peak effect" supports the scenario of enhanced pinning of the vortices right below the VLMT, as observed earlier on twinned and untwinned YBCO single crystals [26,27]. It is likely that the external ripple field inhibits the occurrence of superheating/undercooling of the vortex solid/fluid at the first-order transition, which might be made responsible for the apparent increase of the hysteresis width of the torque signal around the phase transition. Our findings indicate that the irreversibility line and the ML are not necessarily identical near T_c. In fact, the application of a weak ac field [25] provides the possibility to tune the degree of irreversibility, and to clearly separate the irreversibility line from the ML. Magnetic irreversibility effects very near the VLMT of similar untwinned YBCO crystals could not be detected by moving-sample SQUID magnetometry [6,9]. However, our torque data taken on the same sample as used in Ref. [9] also show a magnetic hysteresis right below the VLMT. We believe that the process of moving a sample in the slightly inhomogeneous magnetic field of conventional magnetometers may lead to a suppression of any residual weak magnetic irreversibility.

The VLMT can also be identified in the angular dependence of the torque signal, which is usually well described by the mean-field expression [28]: $\tau(\vartheta) \propto \sin(2\vartheta)/\varepsilon(\vartheta)\ln[\eta H_{c2}^c/H\varepsilon(\vartheta)]$, where η is a constant of the order unity, H_{c2}^c is the upper critical field along c, and $\varepsilon(\vartheta) = (\sin^2\vartheta + \gamma^{-2}\cos^2\vartheta)^{1/2}$ where γ^2 is the effective mass anisotropy [29]. By fitting the experimental data to this expression, the anisotropy parameter γ was found to be constant over a wide temperature range well below T_c with $\gamma_{ac} = 8.2(1)$ and $\gamma_{bc} = 9.2(1)$ for $\mathbf{H}$ rotating in the ac and bc planes, respectively (see Fig. 2a). From these values, one obtains $\gamma_{ab} = \gamma_{bc}/\gamma_{ac} = 1.12(3)$. In the presence of irreversibility we considered the reversible torque given by $\tau_{\rm rev} = \frac{1}{2}(\tau_+ + \tau_-)$ (see Fig. 1a). Above 92.0 K, however, the fitted curves (Fig. 2b) show systematic deviations from the experimental data with unrealistic values of the fitting parameters. These deviations of $\tau_{\rm rev}(\vartheta)$ from mean-field behavior are a direct consequence of thermal fluctuations near T_c. However, it is important to note that the first-order character of the VLMT is very robust against the occurrence of such fluctuations (see the steplike transition at 92.0 K in the inset of Fig. 2a). The ΔM can be well resolved up to 92.8 K, where our torque data and the specific heat [30] already strongly deviate from mean-field behavior.

All torque data showing VLMT were analyzed assuming a Gaussian distribution of the magnetization step ΔM with half-widths δH_c and $\delta\vartheta$ for field- and angular-dependent measurements, respectively, and a polynomial background around the transition. In the presence of hysteresis we used the reversible torque $\tau_{\rm rev}$ in order to evaluate ΔM (see Fig. 1a). The magnetization M follows from the expression for τ mentioned above. Since the vector $\mathbf{M}$ is less than 1° away from the c axis in the investigated range of angles ϑ [28], we make the approximation $\Delta M \simeq \Delta M^c$. General scaling properties of anisotropic superconductors allow us to convert the applied magnetic field $\mathbf{H}$ to a field component along the c axis $H^c = \varepsilon(\vartheta)H$ [29]. Additional measurements were done by varying the angle ϑ at constant T (Fig. 2), not only to check the validity of this scaling procedure but also to obtain further ΔM data.

Figure 3a shows the step in the magnetization ΔM as a function of T as obtained from field- and angular-dependent torque. We found that ΔM and the width of the transition δH^c are virtually independent of the sweep rate. Steplike features can still be resolved at 92.8 K. Above this temperature, any melting-related anomaly, if present, remains below our detection limit. We compare, in the inset of Fig. 3a, our present data with $\Delta M(T)$ as reported earlier for a larger YBCO crystal [9]. Whereas $\Delta M(T)$ of the larger crystal drops abruptly at temperatures well below T_c, we were able to resolve ΔM up to $T/T_c \approx 0.995$

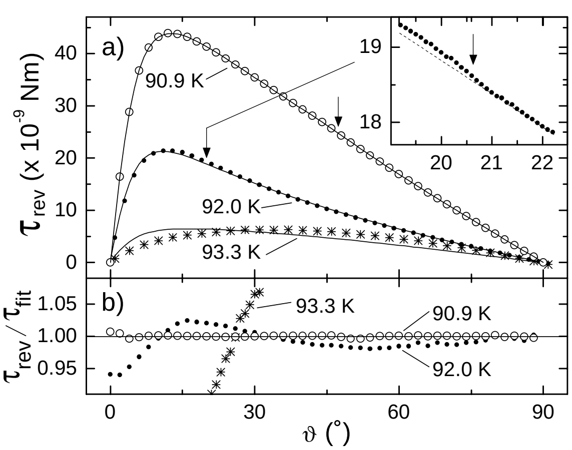

FIG. 2. (a) Reversible angular-dependent torque $\tau_{\rm rev}(\vartheta)$ obtained at three different temperatures, measured in a constant magnetic field of 1 T (for clarity the data at 93.3 K are amplified by a factor of 5). At 90.9 K the data are well described by the mean-field expression explained in the text, taking a Gaussian distribution of VLMTs into account (solid lines). The melting transition is indicated by the arrows for 90.9 and 92.0 K (inset). (b) Above 90.9 K pronounced deviations from mean-field behavior ($\tau_{\rm rev}/\tau_{\rm fit} = 1$) due to fluctuations are observed. For $T = 93.3$ K $\approx T_c$, the deviations exceed the frame of the figure.

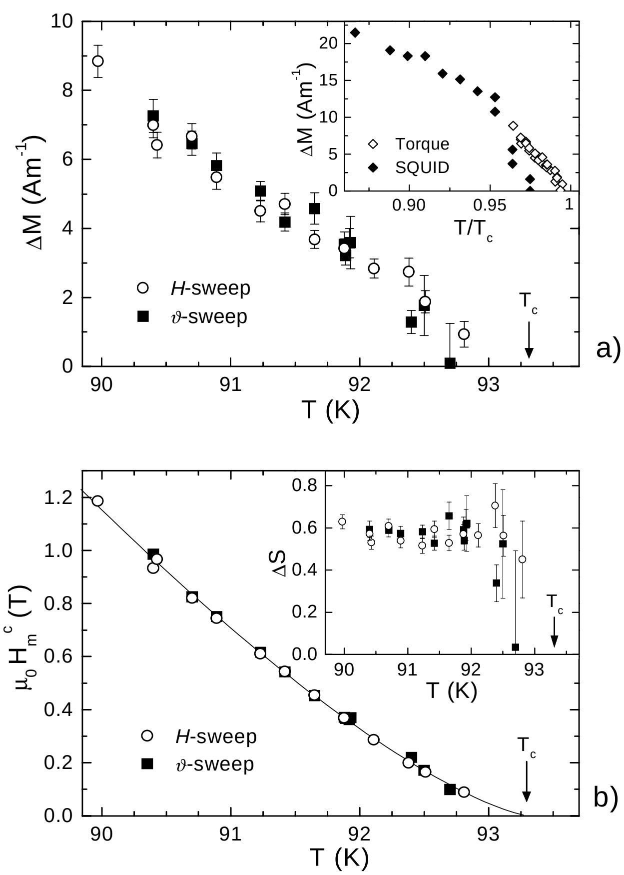

FIG. 3. (a) Magnetization jump ΔM, extracted from field- and angular-dependent torque measurements, as a function of T. A comparison between ΔM data from this work and from SQUID measurements taken from Ref. [9] on a similar but larger sample is shown in the inset. (b) Vortex-lattice ML $H_m^c(T)$ derived from field- and angular-dependent torque curves. The corresponding discontinuity in entropy ΔS (in units of k_B per vortex per Cu-O double layer) as a function of temperature is displayed in the inset.

with no sign of anomalous behavior. The Gaussian full-width of the VLMT for the investigated crystal is typically $2\mu_0 \delta H^c \approx 20$ mT. Using the temperature dependence of the ML, we calculate a transition width $\delta T \approx 40$ mK, which represents only about 25% of that reported in other YBCO single crystals [5,6,8–14]. It is thus very likely that the anomalous drop of ΔM and ΔS below T_c observed in other samples [9,11] and the absence of any meltinglike feature in earlier torque data on YBCO [18] are related to sample quality.

The ML extracted from the position of ΔM (see Fig. 3a) is displayed in Fig. 3b. Near T_c the data are well described by a power law $H_m^c(T) = H_0(1 - T/T_c)^n$ with $\mu_0 H_0 = 107(16)$ T, $T_c = 93.31(6)$ K, and $n = 1.35(5)$, as represented by the solid line in Fig. 3b. This value of n is consistent with the critical exponent $n = 2\nu \simeq \frac{4}{3}$ predicted for the 3D XY model [31]. Note that a power-law fit to previous high-field data on a similar crystal [13] yielded a somewhat smaller value, $n = 1.21(2)$. The entropy jump ΔS associated with melting was determined from the measured ΔM and the ML $H_m^c(T)$ using the Clausius-Clapeyron equation $\Delta S = -\Delta M \mu_0 dH_m^c/dT$. The results of ΔS as a function of temperature, in units of k_B per vortex per Cu-O double layer, are indicated in the inset of Fig. 3b. Excluding the two data points closest to T_c, we calculate an average value $\Delta S_{\mathrm{av}} = 0.57(7) k_B$ per vortex per double layer, in perfect agreement with specific-heat data [9,13] and the theoretical prediction of Dodgson *et al.* [32]. The absence of an anomalous increase in $\Delta S(T)$ close to T_c may indicate that the possible creation of vortex loops near T_c as proposed by Nguyen *et al.* [19] either does not take place in YBCO or occurs above $T/T_c = 0.995$, i.e., in a much lower magnetic-field range than anticipated [19].

In summary, high-resolution field- and angular-dependent magnetic-torque measurements were performed on an untwinned YBCO single crystal in order to investigate vortex-lattice melting near T_c. The first-order VLMT is observable up to 0.5 K below T_c. The corresponding discontinuities ΔM in the magnetization were detectable even at temperatures where the torque signal is perturbed by thermal fluctuations. Around and below the VLMT the torque signal is characterized by a noise related to the pinning/depinning of vortex bundles that vanishes in the fluid phase. We were able to significantly suppress the irreversibility below the VLMT by applying a transverse magnetic field, demonstrating that the ML and the irreversibility line can be separated from each other near T_c. The entropy change ΔS per vortex per Cu-O double layer associated with the VLMT remains constant up to at least 1 K below T_c.

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