Discontinuity of Reversible Magnetization in Untwinned YBCO Single Crystals at the First Order Vortex Melting Transition

Ruixing Liang, D. A. Bonn, and W. N. Hardy
Department of Physics, The University of British Columbia, 6224 Agricultural Road, Vancouver, British Columbia, Canada V6T 1Z1
(Received 26 June 1995)

We report magnetization measurements on clean and untwinned $YBa_2Cu_3O_{6.95}$ single crystals with extremely low pinning. For the first time, an irreversibility line well below the vortex melting line has been achieved. A discontinuity in the magnetization at the vortex melting transition is observed, indicating a first order transition. The entropy change at the phase transition is $0.8k_B$ per vortex per CuO_2 double layer at 10 kOe and $0.6k_B$ per vortex per CuO_2 double layer at 40 kOe, in reasonable agreement with theoretical calculations.

PACS numbers: 74.60.Ge, 74.25.Ha, 74.72.Bk

The vortex dynamics of the mixed state of high T_c superconductors has been a topic of particularly intense interest in recent research. Due to strong thermal fluctuations, the behavior of these materials in the mixed state is very different from conventional superconductors: The vortex lattice melts into a vortex liquid at much lower temperature and magnetic field than the mean-field H_{c2} line; the mean-field H_{c2} line is no longer a phase boundary but a broad crossover [1].

For $YBa_2Cu_3O_{7-x}$ (YBCO), the vortex lattice–vortex liquid phase transition was revealed by torsional oscillator experiments [2] and transport measurements [3–7] on untwinned crystals in magnetic fields, where a sharp onset of the resistivity $\rho(T,H)$ was observed at low field. Hysteresis in the resistivity $\rho(T,H)$ at the transition [4,6,7] indicates that the transition is first order, as predicted by theory [8,9]. At fields higher than a critical value, the first order transition gives way to a continuous vortex glass transition [6].

There remains, however, an important issue to be resolved. A first order phase transition is necessarily characterized by a discontinuity of the first derivatives of the free energy G, such as entropy $S(T,H) = -\partial G/\partial T$ and magnetization $M(T,H) = -\partial G/\partial H$. A recent Monte Carlo simulation has suggested that the entropy change of the vortex melting transition is $\Delta S = 0.3k_B$ per vortex per CuO_2 double layer for YBCO in a magnetic field of 100 kOe [9]. Nevertheless, neither a latent heat nor a clear discontinuity of magnetization at the transition has been experimentally observed for YBCO. A recent torque measurement [10] suggested that the magnetization jump, if it exists, is less than 1% of the theoretical value [9]. However, it should be noted that the observation of the latent heat or magnetization jump at the transition is extremely difficult in the presence of large amounts of pinning which will broaden the transition. In fact, the sharp onset of the resistivity observed in YBCO indicates that a certain level of pinning remains in the samples, since for a perfect sample in the vortex lattice state the magnetization should be reversible, and the resistivity should be nonzero for finite current due to the energy dissipation of the vortex flow. For BSCCO, where the pinning is much weaker compared to YBCO and a sharp onset of the resistivity was not observed, a clear magnetization jump has recently been observed at the vortex melting transition [11].

In this paper, we present reversible dc magnetization data on clean and untwinned YBCO crystals with extremely low pinning level as indicated by an irreversibility line that lies well below the vortex melting transition. A discontinuity in the magnetization at the vortex melting transition is observed, confirming the first order nature of the transition.

The experiments were performed on two YBCO crystals grown by a flux method using yttria stabilized zirconia crucibles, as reported elsewhere [12]. They both have a size of $1 \times 1 \times 0.05$ mm^3. One of the crystals was naturally untwinned and the other was detwinned by applying a uniaxial pressure of 5 MPa along the a axis at 250 °C. The crystals were finally annealed at 450 °C in pure oxygen for one week to ensure oxygen homogeneity. The oxygen content in the crystals was found to be 6.95 by a titration analysis on a piece of ceramic sample annealed together with the crystals. The two crystals, both having $T_c = 93.1$ K and a transition width of 0.2 K as determined by field cooled magnetization measurement at 2 Oe, showed essentially identical magnetization results.

The magnetization data were obtained using a commercial SQUID magnetometer (Quantum Design) with second derivative pickup coils and a 5.5 T magnet. The samples were held in a homemade sample holder designed to eliminate vibration of the sample during the measurements, and to have a very low background. The raw SQUID signals were recorded and then analyzed using a custom program which subtracts the sample holder signal from the measured signal before calculating the magnetization. The magnetic field was always applied along the c axis, and the measurements were made at fixed fields with changing temperature. Both field cooled (FC) and zero field cooled (ZFC) measurements were made at each field to verify the reversibility.

0031-9007/96/76(5)/835(4)$06.00

The standard measurement technique used in this work involves moving the sample through the pickup coils. It is known that the spatial inhomogeneity of the applied field over the distance that the sample is moved may cause artifacts in the magnetization results [13]. To clarify this issue, we have made the measurements with the scan length varying from 1 cm, where the field inhomogeneity is 0.001% according to the manufacturers' specifications, to 5 cm, where the field inhomogeneity is 0.6%. We found that, while the magnetization data below the irreversibility line is indeed scan length dependent, the magnetization data above the irreversibility line, the region of interest of the present work, is not affected by changing scan length. In particular, different scan lengths yield essentially the same magnetization jump at the vortex melting point. The data shown in this paper were typically obtained with a scan length of 1.8 cm where the field inhomogeneity was about 0.003%.

Shown in Fig. 1 are the magnetization data of a crystal for both FC and ZFC measurements. The irreversibility temperature, defined as the temperature at which the FC magnetization and ZFC magnetization start showing a difference larger than the resolution limit (corresponding to $J_c \sim 5\ \mathrm{A/cm^2}$), is 80 K at 5 kOe and 75 K at 30 kOe. To our knowledge, this is the most reversible magnetization behavior reported for YBCO, and is even better than the BSCCO crystal in which a vortex melting transition was observed [14]. Since the effect of pinning decreases with increasing temperature, and the vortex melting point is located at about 10 K above the irreversibility temperature, it is reasonable to believe that the magnetization data around the vortex melting point is unaffected by pinning and represents equilibrium behavior. An ac susceptibility measurement on a similar crystal [15] also indicated very low pinning. It is worth mentioning that we only observed a magnetization jump at the vortex melting point in crystals with such a low irreversibility line. We have found that a relatively small level of added impurities (such as 0.15 at. % Zn or 0.3 at. % Ni) or a number (order of 10) of twin boundaries is sufficient to move the irreversibility line right up to the vortex melting line.

The magnetization around the vortex melting transition is shown in Fig. 2 where a straight line fit to the magnetization data right above the transition temperature is subtracted from the raw data. The magnetization shows a jump at the transition, with the size of the jump a few times larger than the experimental uncertainty. The direction of the jump indicates that the vortex liquid phase has a higher vortex density and a larger entropy than the vortex lattice phase. The magnetization discontinuity is what one expects for a first order vortex lattice melting transition. The hysteresis observed in resistivity measurements [4,6,7] was not clearly observed in this work, because our temperature resolution of 0.01 K is comparable to the hysteresis width [4,6,7].

The slope change of $M(T)$ below and above the transition leads to a large uncertainty in determining the size of the magnetization jump. Nevertheless, we estimated the size of the jump ΔM as the height at

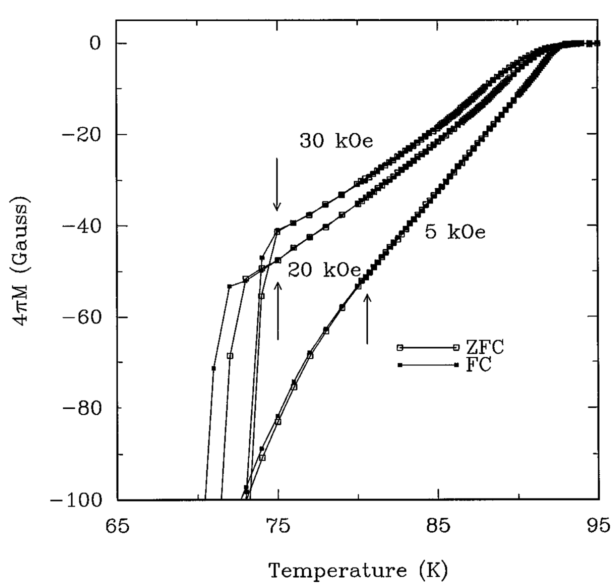

FIG. 1. FC and ZFC magnetization of a crystal at 5, 20, and 30 kOe. The arrows indicate the irreversibility temperature where the FC magnetization and ZFC magnetization start showing a difference larger than the resolution limit (corresponding to $J_c \sim 5\ \mathrm{A/cm^2}$). Note that the irreversibility temperatures are more than 10 K lower than the vortex melting temperatures observed in this work or reported in the literature [2–7], indicating very low pinning in the crystal.

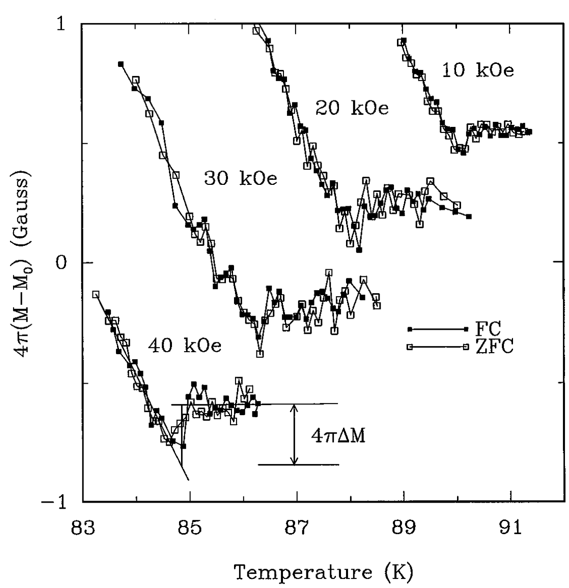

FIG. 2. Magnetization of a crystal near the vortex melting temperature. A straight line fit to the magnetization data right above the transition has been subtracted from the raw data. A discontinuity of the magnetization can be seen, which indicates a first order phase transition. The method of estimating the magnetization change at the transition is also demonstrated in the figure.

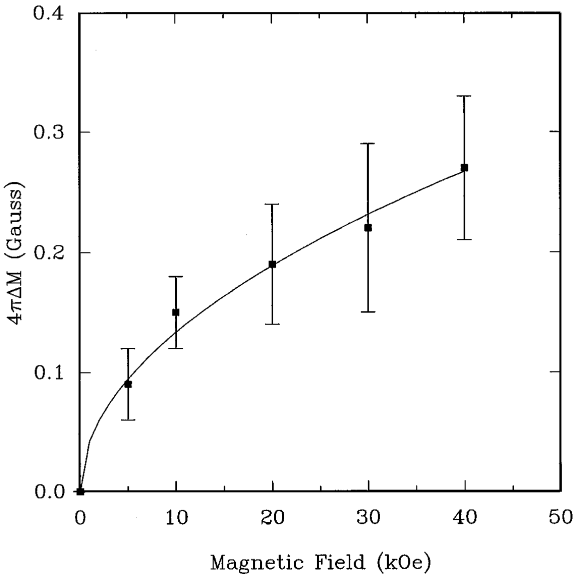

FIG. 3. The plot of the magnetization change ΔM vs vortex melting field H_m. The solid line is a fit of the data by $H^{0.5}$.

the midpoint of the transition from the straight line fit to the data right below the transition to straight line fit to the data right above the transition as demonstrated in Fig. 2. The results are shown in Fig. 3 as a function of magnetic field. We found that ΔM is roughly proportional to $H^{1/2}$.

The vortex melting and the irreversibility lines are shown in Fig. 4. The vortex melting line agrees reasonably well with the results of resistivity measurements [2–7]. We found that H_m is proportional to $(T_c - T)^{1.34\pm0.04}$ which is consistent with $(T_c - T)^{2\nu}$ [1], where $\nu = 0.669$ is the critical exponent expected for the 3D XY-type critical relations $\xi = \xi_0|t|^{-\nu}$ and $\lambda = \lambda_0|t|^{-\nu/2}$. The London penetration depth λ has been experimentally found to be $\propto |t|^{-\nu/2}$ in a 10 K wide temperature range below T_c [16].

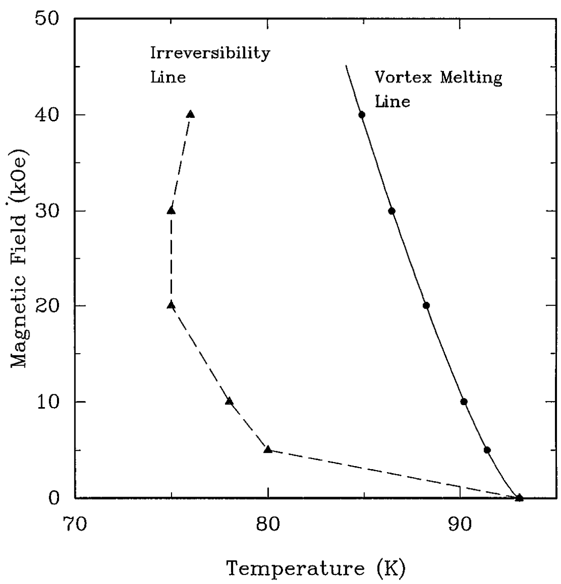

FIG. 4. Vortex melting and irreversibility fields of the crystals as a function of temperature. The solid line is a fit of the vortex melting point data by $H_m = 2.40 \times 10^3(T_c - T)^{1.34}$ (Oe).

According to the 3D XY critical model [1], the fluctuation magnetization has the scaling form

$$M = H^{0.5} f(t/H^{1/2\nu}), \tag{1}$$

where $t = (T - T_c)/T_c$ is the reduced temperature and f an unknown function. Using $T_c = 93.1$ K determined by FC magnetization measurements at 2 Oe, and without using any adjustable parameters, we found the magnetization data can be scaled perfectly by Eq. (1) from above T_c to somewhat below the vortex melting point, as shown in Fig. 5. Similar scaling was also reported by Salamon *et al.* [17]. We found that the vortex melting line collapses into a single point, and the magnetization jump can be seen clearly on the scaling curve, further confirming the temperature and field dependencies $H_m \propto |t|^{2\nu}$ and $\Delta M \propto H^{1/2}$.

It is of great interest to calculate the entropy change at the vortex melting transition. According to the Clausius-Clapeyron equation,

$$\Delta S = -\Delta M(dH_m/dT). \tag{2}$$

Putting the results of this work, $H_m = 2.40 \times 10^3(T_c - T)^{1.34}$ (Oe), $dH_m/dT = -3.22 \times 10^3(T_c - T)^{0.34} =$

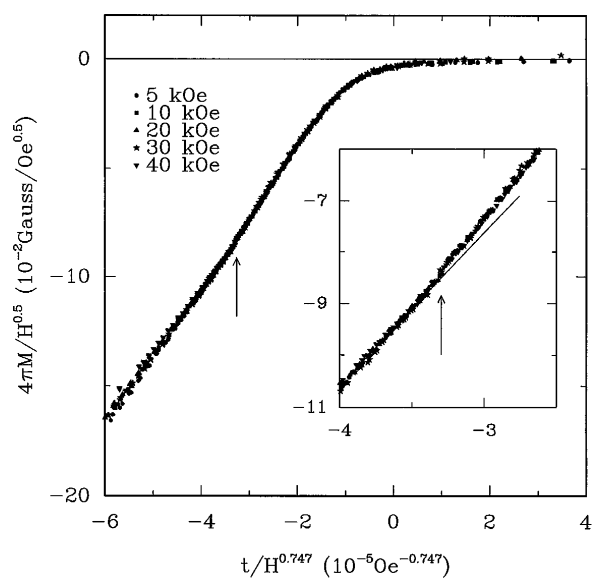

FIG. 5. 3D XY scaling of the magnetization data, where $t = (T - T_c)/T_c$. The inset shows the details of the data near the vortex melting transition. Using the independently measured $T_c = 93.1$ K and without any adjustable parameters, all of the data can be scaled into a single curve from above T_c to somewhat below the vortex melting transition. The vortex melting line collapses into a point on the scaling curve, and the magnetization discontinuity can be seen clearly, as indicated by the arrows.

$-4.47 \times 10^2 H_m^{0.254}$ (Oe/K) and $4\pi\Delta M = 1.3 \times 10^{-3} H_m^{0.5}$ (G) into the equation yields $\Delta S = 4.7 \times 10^{-2} H_m^{0.75}$ (erg K^{-1} cm^{-3}) with an uncertainty of $\pm 20\%$. At the vortex melting transition, $B \approx H$ and then we have $\Delta S = 8.2 H_m^{-0.25}$ (k_B per vortex per CuO_2 double layer) which gives $0.8k_B$ and $0.6k_B$ per vortex per CuO_2 double layer at 10 and 40 kOe, respectively. If one extrapolates the data to a higher field, the ΔS is found to be $0.48k_B$ per vortex per CuO_2 double layer at 100 kOe, in reasonable agreement with a recent Monte Carlo result of $0.3k_B$ per vortex per layer at 100 kOe [9].

In conclusion, we have measured the reversible magnetization on very clean, untwinned YBCO single crystals with an irreversibility line at exceptionally low temperatures. A discontinuity of the magnetization at the vortex melting transition was observed. This confirms the first order characteristic of the transition. We have calculated the entropy change of the transition from the magnetization data and found it agrees well with the Monte Carlo calculations of Hetzel, Sudbø, and Huse [9].

The authors would like to thank M. Charalambous for helpful comments on the manuscript. This work was supported by the Natural Science and Engineering Research Council of Canada and by the Canadian Institute for Advanced Research.

Note added.—Following submission of this paper, we have made measurements on a larger crystal and obtained data with 5 times better signal-to-noise ratio. The new data, in addition to being perfectly consistent with the results presented in this paper, show in detail that the magnetization data around the vortex melting transition are not affected by the inhomogeneity of the applied field.

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