

Magnetization jump and the vortex-lattice melting transition in $\text{YBa}_2\text{Cu}_3\text{O}_y$

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Magnetization measurements in high-quality twinned $\text{YBa}_2\text{Cu}_3\text{O}_y$ single crystals are performed as a function of temperature, magnetic field, and its direction. We find an anomalous magnetization jump just below the irreversibility line, which provides the thermodynamic evidence of the first-order vortex-lattice melting transition in $\text{YBa}_2\text{Cu}_3\text{O}_y$ system, following a reasonable Lindemann criterion with $c_L=0.13$. In addition, we demonstrate that a peak effect in the critical current density just below the melting line is associated with the enhanced vortex pinning due to the premelt softening of the vortex lattice and this peak is different from a so-called second peak.

Recent studies of the magnetic phase diagram in high- T_c superconductors have proved that a vortex liquid state exists above an irreversibility line $H_{\text{ir}}(T)$ due to strong thermal fluctuations.¹⁻⁵ The vortex liquid state in highly disordered systems is predicted to undergo a second-order phase transition into a vortex glass state¹ or a Bose glass state,² depending on the type of the disorder. The transition, on the other hand, changes to a first-order phase transition into the Abrikosov vortex lattice in case of clean systems with very weak flux-pinning centers.³⁻⁵

Experimental investigations of the first-order melting transition were performed in untwinned and twinned single crystals of $\text{YBa}_2\text{Cu}_3\text{O}_y$.⁶⁻¹⁰ Farrell, Rice, and Ginsberg⁶ observed a sharp damping peak in the low-frequency torsional-oscillator measurement at the temperature T_m , and pointed out that the obtained magnetic-field dependence, $H(T_m) \propto (T_c - T_m)^2$, agreed well with the vortex-lattice melting theory.^{4,5} A discontinuous resistive kink with hysteresis was also interpreted as first-order melting transition.⁸⁻¹⁰ However, more recently, Jiang *et al.*¹¹ claimed that the resistive hysteresis is not direct evidence of the first-order transition, and they proposed that the hysteresis may result from a current-induced nonequilibrium effect which is related to the vortex dynamics. Since the resistivity is not thermodynamic property, measurements of thermodynamic quantities such as a specific heat or a magnetization is required in order to make sure the first-order transition. Contrary to the above case of $\text{YBa}_2\text{Cu}_3\text{O}_y$,⁶⁻¹⁰ the discontinuous change of the magnetization was observed at the vortex-lattice melting transition in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ single crystals.¹²⁻¹⁴

In this paper, we present an anomalous magnetization jump just below $H_{\text{ir}}(T)$ in high-quality twinned $\text{YBa}_2\text{Cu}_3\text{O}_y$ single crystals. We show the thermodynamic evidence that the abrupt magnetization jump at a temperature $T_j(H)$ is closely related to the first-order vortex-lattice melting transition. We also report that a peak effect in the critical current density $J_c(H, T)$ just below the $T_j(H)$ line is caused by the

enhanced pinning due to the softening of the vortex lattice at the melting transition. These results indicate that the vortex pinning region in the H - T phase diagram is very close to the vortex melting transition line for $\text{YBa}_2\text{Cu}_3\text{O}_y$.

High-quality twinned $\text{YBa}_2\text{Cu}_3\text{O}_y$ ($y=6.9$) single crystals were grown by a slow cooling of a solid-melt mixture in yttria crucibles under the peritectic condition. The as-grown crystals were annealed at 450–500 °C for 7 days in a flowing oxygen. Samples used in this study showed superconducting transition temperature $T_c=89$ K with the sharp width of $\Delta T_c(10-90\%)<500$ mK in both resistive and magnetic measurements. The electrical resistivity in the a - b plane is $\rho_n=28 \mu\Omega \text{ cm}$ at the onset temperature of the transition. Sample dimensions were typically $1.3 \times 1.5 \times 1.4 \text{ mm}^3$. Details of the crystal-growth technique were described elsewhere.¹⁵ The magnetization measurements were performed by using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design). In order to avoid an influence of the field inhomogeneity, magnetization data were taken with a short scan length of 10–20 mm. In our SQUID system, a magnetic-field variation in a solenoid was smaller than a typical value of the magnetization jump and magnetization data did not depend on the various scan length in the range of 10–20 mm. Furthermore, following the discussions in Ref. 16, special attention was paid to the scan wave form to determine the scan length and it was confirmed that there were no irregular scan wave forms with asymmetry during measurements. The sample was rotated around the axis parallel to the a or b axis, and the rotated angle θ was defined as the angle between the c axis and the direction of the magnetic field.

Figure 1 shows the magnetization M as a function of the temperature in zero-field-cooled (ZFC), field-cooled on cooling (FCC), and field-cooled on warming (FCW) modes at $H=30$ kOe parallel to the c axis. The dependence of M on T shows an anomalous magnetization jump at a certain temperature $T_j(=81.8$ K) with a narrow width (~ 0.5 K) just

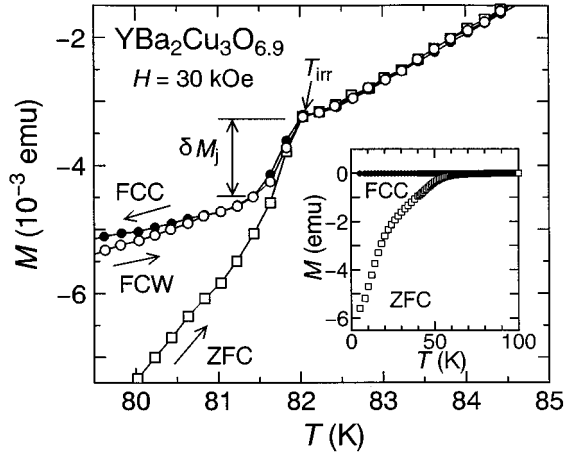


FIG. 1. Temperature dependence of the magnetization in ZFC, FCC, and FCW processes at $H=30$ kOe parallel to the c axis. The inset shows an overall view in whole temperature region studied.

below the irreversibility line. Except for above small jump, data of ZFC and FCC modes in a wide range of T change gradually and continuously with no additional anomaly as shown in the inset of Fig. 1. Since the magnetization jump is related to the discontinuous change of the vortex density in the sample, the jump would result from the thermodynamic property of the first-order transition or the vortex pinning behavior. Pastoriza *et al.*¹² reported a similar magnetization anomaly in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ and they related the anomaly with the first-order vortex melting transition.

Figures 2(a) and 2(b) show the temperature dependence of the magnetization in ZFC and FCC processes in various

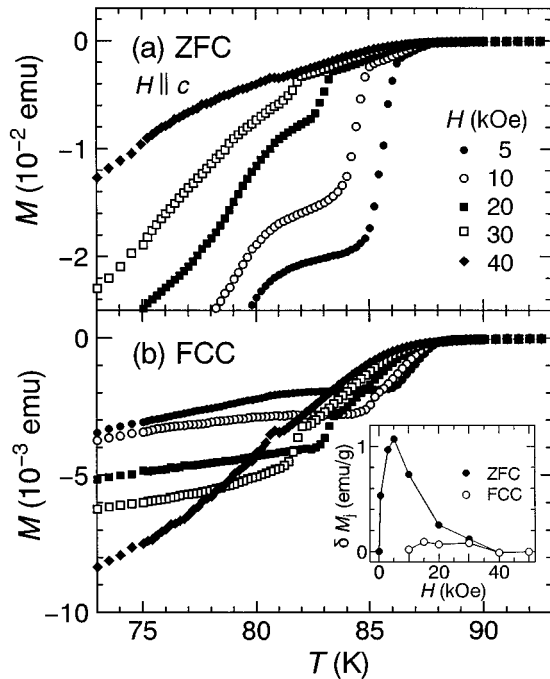


FIG. 2. Magnetic-field dependence of $M(T)$ in (a) ZFC and (b) FCC scans. The inset shows the magnetic-field dependence of a height of the magnetization jump δM_j .

fields parallel to the c axis, respectively. In the ZFC magnetization, a height of the magnetization jump, δM_j , depends largely on the magnetic field as shown in the inset of Fig. 2(b). However, in the FCC magnetization, the distinct jump is observed only above 10 kOe and has small δM_j . The magnetization at the onset of jumps M_j^{on} has almost constant value (about -0.003 emu), independently of H and the cooling process. It is similar to resistivity measurements for the vortex-liquid transition with the constant value ($\sim 10\%$ ρ_n) at the onset of the resistive kink.⁸⁻¹⁰ On the contrary, the endpoint value of the magnetization jump M_j^{end} depends on H in the ZFC magnetization. The result may suggest that the low- T region of the jump is significantly affected by the vortex pinning property as shown later and the value of δM_j would be enhanced. However, the magnetization jump in the FCC mode cannot be explained by the vortex pinning behavior, because the jump means a steep decrease of the vortex density with decreasing temperature, contrary to the usual T dependence of the vortex pinning force. Therefore, the discontinuous change of the magnetization indicates a thermodynamic first-order melting transition of the vortex lattice. The vortex liquid above the transition is denser than the vortex solid due to the strong thermal fluctuations in the liquid phase.

According to the Clapeyron-Clausius equation, the entropy change per vortex per CuO layer at the transition is given by $\Delta s = -d\phi_0(\delta M_j/B_j)(dH_j/dT)$, where δM_j is defined by $M_j^{\text{on}} - M_j^{\text{end}}$, d is the interlayer distance and ϕ_0 the flux quantum. Since T_j decreases with increasing H (i.e., $dH_j/dT < 0$) and δM_j indicate a positive value below $H=30$ kOe, the magnetization jump is consistent with the first-order vortex-lattice melting transition. From the FCC magnetization jump, the latent heat per vortex per layer, $l = T_j \Delta s$, is estimated to be $6 k_B T$ at 10 kOe and $25 k_B T$ at 30 kOe. The obtained value at 10 kOe is several times as large as that in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$,¹⁴ and is about an order of magnitude larger than that obtained by the Monte Carlo simulations of vortex lattice melting.¹⁷ These differences may be caused by the different characteristic parameters between them.

On the contrary, in the high-field region above $H=35$ kOe, the sign of δM_j changes to negative for both the ZFC and FCC measurements. A similar sign reversal of δM_j was also reported in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$.¹³ The negative sign of δM_j is inconsistent with the first-order phase transition, however H dependence of T_j for both positive and negative regions of δM_j is almost connected continuously. Although the origin of the negative jump is not clear, the transition above $H=35$ kOe may be a second-order transition from glass state to liquid state.¹³

Figure 3 shows the critical current density J_c estimated from the $M-H$ hysteresis curve as a function of the reduced temperature in various constant fields parallel to the c axis. With increasing T , J_c decreases rapidly, and a sharp peak is observed at a temperature $T_{\text{ps}} = 0.98 T_{\text{irr}}$ independent of H . Shown in the inset of Fig. 3, $J_c(H)$ exhibits remarkable peak structures at an intermediate field region [$H_{p(2\text{nd})} \sim 0.4 H_{\text{irr}}$], which correspond to a so-called second peak of the $M-H$ curve.¹⁸ In the vicinity of H_{irr} , an additional shoulder structure also appears at a field H_{ps} , which would be more clear if the second peak did not exist. The peaks at T_{ps} in $J_c(T)$ (Fig. 3) and the shoulders at H_{ps} in $J_c(H)$ (the inset of Fig. 3) can

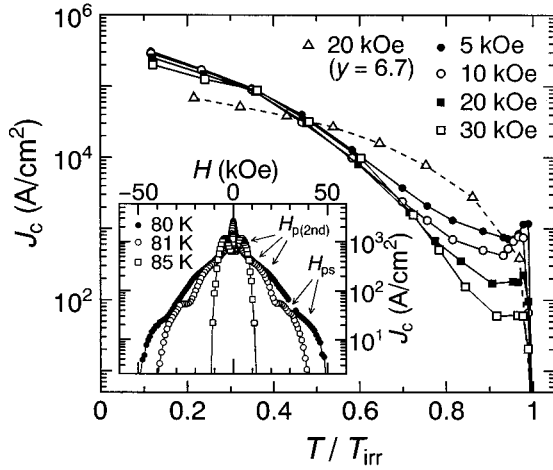


FIG. 3. Critical current density estimated from the M - H hysteresis curve as a function of the reduced temperature. The open triangles connected by the dashed curve show data of the reduced crystal with $y=6.7$ at $H=20$ kOe for comparison. The inset shows critical current density as functions of the magnetic field.

be expressed by the same line on the H - T phase diagram, which are located at higher magnetic-field region than the second peak $H_{p(2nd)}$ as shown later (see Fig. 4). Therefore the vortex pinning mechanism of the peak and the shoulder is different from that of the second peak.

The peak effect obtained by the magnetization measurements in this study is similar to that obtained by the measurements of vibrating reed,¹⁹ susceptibility,²⁰ and transport critical current.²¹ It is suggested that the enhanced vortex pinning at H_{ps} and T_{ps} is connected with the softening of the shear modulus C_{66} of the vortex lattice near the melting transition.^{20,21} With decreasing C_{66} , vortex-vortex interaction decreases compared with the vortex-pinning interaction, so this effect enables the vortices to adjust into the more suit-

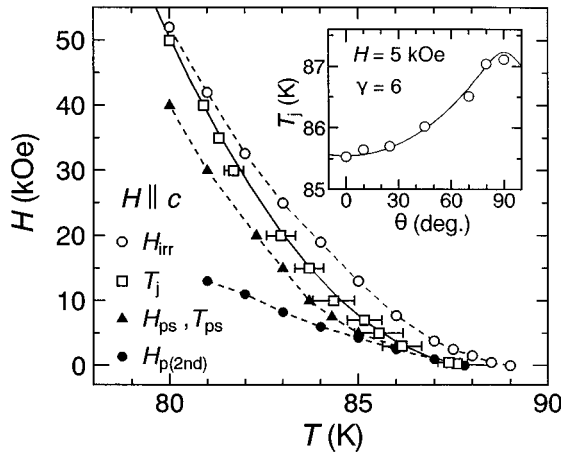


FIG. 4. T_j versus H and the temperature dependence of the various characteristic fields such as $H_{p(2nd)}$, H_{ps} , and H_{irr} . The solid curve is a fit to a power law $H=H_0(1-T_j/T_c)^n$ with $n=2$. The dashed curves are drawn as a guide to the eye. The inset shows T_j at $H=5$ kOe as a function of the angle θ between the c axis and the magnetic-field direction. The solid curve in the inset is a fit by Eq. (1) with the parameters $\gamma=6$ and $A=0.52$ K/kOe^{1/2}.

able pinning configuration and the volume pinning force increases. This explanation is analogous to the synchronization mechanism for conventional superconductors,²² which is applied near the upper critical field H_{c2} , thus $H_{ps}(T)$ line [$T_{ps}(H)$ line] is referred to as the synchronization line. Moreover, according to the collective-pinning theory,²³ the volume pinning force is given by $F_p=J_c B=(W/V_c)^{1/2}$, where V_c is the correlation volume and $W(\approx N_p f_p^2/2)$ is the pinning strength with the concentration of pinning centers N_p and the elementary pinning force f_p . The enhancement of J_c is related to the decrease of V_c when C_{66} approaches zero at the melting transition. Therefore the existence of the peak effect just below $T_j(H)$ also strongly suggests the appearance of the vortex-lattice melting transition.

The open triangles in Fig. 3 show $J_c(T)$ of the reduced crystal with $y=6.7$ at $H=20$ kOe for comparison. With increasing temperature, $J_c(T)$ decreases monotonously and does not show any anomalies such as the peak effect. At the high-temperature region $T/T_{irr}=0.7-0.9$, the J_c value of the reduced sample is about an order of magnitude larger than that of the oxygenated one. Furthermore, the magnetization jump is not observed in M - T characteristics for the reduced sample. These results indicate that the first-order vortex melting transition would disappear due to the strong vortex pinning by the oxygen deficiency, suggesting the second-order transition from the vortex glass into the vortex liquid.

Figure 4 shows T_j versus H and the temperature dependence of the various characteristic fields such as $H_{p(2nd)}$, H_{ps} and H_{irr} . $H_{irr}(T)$ in the figure is the irreversibility line obtained from M - H curves, which is slightly larger than that obtained from M - T curves, because of the different time scale of measurements or a small inhomogeneity of the magnetic field in the magnet.¹⁶ On the contrary, the $T_j(H)$ line is independent of measured processes such as M - T and M - H . As shown in Fig. 4, the irreversibility line almost agrees with the onset of the vortex-lattice melting line $T_j(H)$, which is continuously connected with the synchronization line $H_{ps}(T)$ with decreasing T or H . Contrary to the case of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$,¹⁴ the vortex pinning becomes dominant just below the vortex-lattice melting line in $\text{YBa}_2\text{Cu}_3\text{O}_y$. This fact may be related to the enhancement of δM_j in the ZFC scan as mentioned above.

In order to further examine the vortex-lattice melting scenario, let us compare the dependence of T_j on both H and θ with the vortex-lattice melting theory. According to the theory of the first-order vortex-lattice melting,^{4,5,24} the field dependence of the transition temperature T_j for the anisotropic case is given by

$$T_j = T_c - A \sqrt{H} (\sin^2 \theta + \gamma^2 \cos^2 \theta)^{1/4}, \quad (1)$$

with $A = \pi^2 k_B T_c^2 \lambda_{ab}^2(0) \gamma^{1/2} / \phi_0^{5/2} c_L^2$. Here, $\lambda_{ab}(0)$ is the in-plane penetration depth at zero temperature, $\gamma^2 (\equiv m_c/m_{ab})$ is the effective mass ratio, and c_L is the Lindemann criterion number. As shown by the solid curve in Fig. 4, a power-law fit to the experimental data yields $H(T_j) = H_0(1 - T_j/T_c)^n$ with $n=2$ and $H_0 = 6 \times 10^3$ kOe which almost agrees with the phase boundary obtained by Farrell, Rice, and Ginsberg,⁶ and the exponent is consistent with the value expected for the vortex-lattice melting theory.^{4,5} Moreover, the very rea-

sonable value of the Lindemann number $c_L=0.13$ is obtained, using parameters of $\lambda_{ab}(0)=1400 \text{ \AA}$, $T_c=88.5 \text{ K}$ and $\gamma=6$ (Ref. 18).

The inset of Fig. 4 shows the angular dependence of $T_j(\theta)$ at $H=5 \text{ kOe}$. Using the same values above used, the data could be fitted by Eq. (1) with the parameter $A=0.52 \text{ K/kOe}^{1/2}$ which is close to the value ($0.41 \text{ K/kOe}^{1/2}$) obtained by torsional-oscillator measurement.⁷ Thus, the Lindemann criterion is also derived to be $c_L=0.13$ which is in excellent agreement with the value obtained from the T_j - H characteristic as mentioned above. Furthermore, the determined value of c_L by two methods in this study is consistent with the values of 0.16 and 0.15 obtained from the torsional-oscillator measurements⁷ and from transport resistive measurements,⁹ respectively, and is also consistent with the value ($c_L=0.1\sim 0.4$) obtained by Monte Carlo simulations.²⁵

Although the results in this study are very suggestive of the first-order vortex-lattice melting transition, it remains a question whether the melting transition is observable in the twinned $\text{YBa}_2\text{Cu}_3\text{O}_y$ single crystal or not. It was suggested that the first-order transition is suppressed by the strong vortex pinning caused by the planar pinning centers⁹ such as the twin plane and the intrinsic pinning center and point defects introduced by the electron irradiation.²⁶ The planar pinning centers become most effective when the vortices are parallel to the plane and the vortices move across the planes. These configurations are realistic in the resistive measurements under the transport current.⁹ However, in the case of the magnetization measurements, planar defects do not necessarily work as the strong pinning centers but act as the vortex channeling along the plane,²⁷ so the pinning effect by the twin plane would be small. On the contrary, point defects due to the oxygen deficiency and the irradiation defects would work

as collective-pinning centers, independent of the field direction and the measurement technique. We believe that our single crystals contain a small number of point defects, similar to the case of the untwinned single crystals. The oxygen-deficient sample with $y=6.7$, as described above, has large value of F_p at high-temperature region and shows no anomaly corresponding to the vortex-lattice melting transition.

In conclusion, we have presented the discontinuous magnetization jump in T dependence of $M(H, \theta)$ in high-quality twinned $\text{YBa}_2\text{Cu}_3\text{O}_y$ single crystals. We have found the thermodynamic evidence that the magnetization jump is caused by the first-order vortex lattice melting transition, although the obtained latent heat at the transition is slightly larger than the value previously reported. The dependence of the transition temperature T_j on both H and θ is consistent with the vortex-lattice melting theory and a reasonable value of Lindemann number $c_L=0.13$ is obtained. Furthermore, we have demonstrated that the peak effect in J_c just below the melting line is associated with the enhanced vortex pinning due to the softening of the vortex lattice and this peak is different from the so-called second peak. The anomalous magnetization was not observed for the reduced sample with $y=6.7$, indicating the melting transition is suppressed by the strong disorder by the oxygen deficiency.

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