Onset of the fishtail peak in an untwinned $YBa_2Cu_3O_{7-\delta}$ **crystal**

M. Pissas, E. Moraitakis, and G. Kallias

Institute of Materials Science, NCSR Demokritos, 15310, Aghia Paraskevi, Athens, Greece

A. Bondarenko

Kharkov State University, 310077 Svoboda square 4, Kharkov, Ukraine (Received 1 November 1999; revised manuscript received 17 February 2000)

Isothermal global magnetic measurements in a detwinned $YBa₂Cu₃O_{7-\delta}$ single crystal reveal a jump in the slope of the increasing part of the virgin magnetization curves. This feature is broadened or even disappears at low temperatures (T <15 K). At high temperatures (T >75 K) and below the $H_{sp}(T)$ curve in the obtained vortex-matter phase diagram, it is also hard to discern. The magnetic field at which this anomaly occurs (H_3) decreases as the temperature increases. The observed feature may be attributed to a recently proposed disorderinduced transition (or crossover) from a relatively ordered vortex lattice to a highly disordered vortex solid.

I. INTRODUCTION

Extensive experimental studies in the past, including transport,^{1–4} magnetization,^{5–7} ac-susceptibility,^{8–11} and calorimetric measurements $1^{2,13}$ have established the existence of a first-order melting transition of the Abrikosov vortex lattice in YBa₂Cu₃O_{7- δ} (YBCO), Bi₂Sr₂CaCu₂O_{8+ δ} (Refs. 14 and 15) (BSCCO), and $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (Ref. 16) high-quality single crystals.

In parallel with these studies, many theoretical and experimental efforts have been devoted to the properties of the solid state of the vortex lattice, in the presence of quenched random disorder.^{17–21} The most widely discussed feature is the increase of the magnetization with the magnetic field occurring in isothermal magnetization measurements (the socalled fishtail or second peak H_{sp}). Depending on the density and strength of pinning centers, the temperature, and anisotropy of the compound, the irreversible magnetization near the second peak displays different characteristics. For example, the onset of the increase in the magnetization is very sharp and sometimes is divided into two peaks.²² The second peak has been observed in YBa₂Cu₃O_{7- δ} (Refs. 23–30), $YBa_2Cu_4O_8$ (Ref. 31), $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Refs. 32–39), Tl based compounds (Ref. 40), HgBa₂CuO_{4+ δ} (Ref. 41–43), $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ (Ref. 44), and in $La_{2-x}Sr_xCuO₄$ (Ref. 45) high-quality single crystals. Consequently, the second peak is a generic feature of anisotropic and relatively clean high-temperature superconducting crystals. Local magnetization measurements of Zeldov *et al.*⁴⁶ on high-quality $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals revealed that the onset of the peak is very sharp, suggesting that it may indeed mark a phase transition. The smoothness of the peak in global measurements may be a consequence of spatially averaging the inhomogeneous induction inside the sample.

The expectation of such a transition is further supported by theoretical studies of Nattermann,⁴⁷ Giarmarchi and Le Doussal, 48 Gingras and Huse, 49 Ertas and Nelson, 50 Vinokur et al.,⁵¹ and Koshelev and Vinokur,⁵² which predict a fieldinduced transition from a relatively ordered vortex lattice to a highly disordered entangled vortex solid.

At low magnetic fields, in the presence of weak random-

point disorder, there is a transition that separates two distinct solid phases: a weakly disordered quasilattice phase without topological defects, named Bragg glass phase, and a highly disordered entangled solid present at higher fields. These two phases, together with the liquid phase, connect to a multicritical point. The transition lines between these phases are determined by the interplay between three energy scales: the vortex elastic energy, the energy of thermal fluctuations, and the pinning energy.^{53,51} It is the competition between the elastic and pinning energies that determines this new transition, provided that thermal energy can be neglected. The particular line in the phase diagram that is related to this new transition must be lowered in field upon adding impurities. $48,49$ Increasing the magnetic field increases the effective strength of random pinning through the reduced interlayer vortex interaction, making the system more twodimensional and consequently more susceptible to disorder. Such a field-driven transition corresponds to the destruction of the Bragg glass by proliferation of topological defects upon raising the field, which is equivalent to increasing the effective disorder, which favors dislocations.

In the present work, we studied the region where the irreversible magnetization starts to increase with the magnetic field in the temperature regime $0 \le T \le T_c$. For this purpose we used magnetization and magnetic relaxation measurements for a detwinned YBa₂Cu₃O_{7- δ} single crystal. Our major finding is a change in the slope of the increasing part of the $m(H)$ curve, which can be attributed to a field-driven transition to a highly disordered vortex matter. The dc magnetization was measured with a commercial superconducting quantum interference device (SQUID) magnetometer (Quantum Design MPMS II). The measurements were performed using a 2-cm scan length in order to keep the sample in a highly homogeneous magnetic field.

II. EXPERIMENTAL RESULTS

Our sample is an YBa₂Cu₃O_{7- δ} single crystal with dimensions \sim 1.25 \times 1.15 \times 0.035 mm³, which was grown with the self-flux method in a gold crucible. Thermal treatment was made at 450 °C, in oxygen flow and ambient pres-

FIG. 1. Virgin magnetization curves as a function of the applied magnetic field of an YBa₂Cu₃O_{7- δ} single crystal at several temperatures. The inset shows an expanded view of the hysteresis loop at $T=40$ K in the field regime where the new feature is observed. Also, the inset shows the hysteresis loop at $T=83$ K. The arrows indicate the location of this feature.

sure. Detwinning was performed at 400 °C in air under a pressure \approx 25 MPa, followed by additional annealing in oxygen flow at 400°C and ambient pressure for three days. The transition temperature was determined to be T_c \approx 91.6 K (ΔT_c =0.3 K).

Figures 1 and 2 show the virgin magnetization curves for $5 \le T \le 87$ K. Figure 2 is an expanded view of Fig. 1 that displays clearly the new feature. The magnetization curve at 5 K shows a large peak at the field H_{fp} , known in the literature as the first peak. The first peak corresponds to the state where the flux penetrates completely in the interior of the single crystal. For $H > H_{\text{fp}}$, |m| decreases up to approximately 30 kOe, where it starts increasing slowly with a rate of 1×10^{-6} emu/Oe. Since our SQUID magnetometer is limited to 55 kOe, we cannot explore further the magnetization curve at this temperature. The 10 K measurement is similar to that at 5 K. At 20 K, the magnetization curve clearly shows a new feature: a drastic change in the slope in the increasing part of the magnetization $|m(H)|$ curve. It is followed by a monotonic increase of $|m|$ up to the field $H_{\rm sn}$, beyond which a further increase of the magnetic field leads to a decrease of $|m|$. As the temperature is raised, the magnetization curves continue to present the new feature with $H_{\rm sp}$ and $H_{\rm fp}$ moving to lower values, and |m| decreasing rapidly with temperature for constant magnetic field. The overall behavior described above has been reported in sev-

FIG. 2. An expanded view of the virgin magnetization curves as a function of the applied magnetic field, covering the field region where the $m(H)$ curve changes slope abruptly. The arrows indicate the location of the feature that is at the intersection of the dashed lines.

eral publications, $32-38$ except from the change of slope in the $m(H)$ curve.

The new feature can be defined as the point where the slope changes. However, this method introduces uncertainty in determining the location of this feature in the temperature range where it is hardly detectable. Consequently, we prefer to use the derivative dm/dH . Figure 3 shows the $m(H)$ curve and the corresponding *dm*/*dH* versus *H* curves at 50 K. *dm*/*dH* first increases from negative values up to zero,

FIG. 3. An expanded view of the virgin magnetization curve and its derivative *dm*/*dH* as a function of the applied magnetic field at $T = 50$ K.

FIG. 4. (a) Magnetic phase diagram of the studied YBa₂Cu₃O_{7- δ} single crystal. Open circles denote the field *H*₃, defined as the field where $m(H)$ changes slope abruptly. Open squares represent H_{\min} , defined as the field where $|m(H)|$ displays a local minimum. The solid lines represent a fit of H_3 and H_{min} (see main text). The solid squares denote the irreversibility line, while open dotted circles show the melting line. Filled circles represent the second magnetization peak. T^* is the temperature where the $H_{\text{sn}}(T)$ curve exhibits an anomaly. The inset shows the $H_{sp}(T)$ line near the T_c , clarifying the anomaly in the $H_{sp}(T)$ curve. The solid line in the inset is a fit with the relation $H_{sp}(T) = 60[1 - (T/T_c)^4]^{1.4}$. (b) Representative temperature variations of the magnetic moment under 55 and 40 kOe. The arrows mark the irreversibility and melting points.

where the first peak of the virgin magnetization curve occurs. Subsequently, it increases further and then drops to zero again. At this point, we define H_{\min} . For $H > H_{\min}$, dm/dH decreases until point *A* and then, in an interval ΔH \approx 1.8 kOe, jumps to a nearly field independent value at point B (Fig. 3). We locate the new feature at the middle of the interval *AB*. The curves at 5 and 10 K do not show this feature. We observe clearly this feature only above 15 K and up to 75 K.

Figure $4(a)$ is the magnetic phase diagram of the studied crystal showing the H_{min} and H_3 lines, the second peak, and the irreversibility and melting lines. Figure $4(b)$ shows representative *m* vs T measurements that illustrate the way the irreversibility and melting points were defined. The irreversibility line is defined by the locus of points where the $m(T)$ curve changes slope abruptly. At this point, the irreversible screening current drops about two orders of magnitude and seems that for $T \geq T_{irr}$ the magnetic moment is reversible. Of course such an estimation of the irreversibility line depends on the resolution of magnetometer. The melting line is defined by the locus of points (H, T_m) where the $m(T)$ curves, in the reversible regime (according to the previous definition) show a jump after subtracting a linear $m(T)$ variation below T_m . We note that the anomaly at the melting temperature is difficult to be traced in our experimental curves for $H \le 10$ kOe, probably due either to resolution limitations of our SQUID, or to the existence of a lower critical point.^{54,55}

The curve defined from the points (T, H_3) decreases slowly with increasing temperature as $H_3(T) = H_3^0 T^{\alpha}$ with $H_3^0 = 40.4 \pm 5$ and $\alpha = -0.48 \pm 0.03$, while the curve $H_{\text{min}}(T)$ decreases as $H_{\text{min}} = H_{\text{min}}^0 T^\beta$ with $H_{\text{min}}^0 = 176 \pm 2$ kOe and β $=$ -1.09 \pm 0.01. The second magnetization peak curve $H_{\text{sn}}(T)$ displays a change in the slope at $T^* = 84 \pm 1$ K, implying a change of temperature dependence. The second magnetization peak curve $H_{sp}(T)$ for $T \leq T^*$ can be described with a power-law relation, $H_{\rm sp}(T) = H_{\rm sp}^0 (1 - T/T_c)^{\nu}$, with ν = 1.23 and H_{sp}^0 = 76.45 kOe. For $T > T^*$, the relation $H_{sp}(T)$ =60[1 – $(T/T_c)^4$]^{1.4} proposed by Abulafia *et al.*⁵⁶ is more appropriate. It is interesting to note that the $H_3(T)$ curve tends toward the point at which the $H_{\text{sn}}(T)$ curve changes its temperature dependence. This means that the change of the vortex matter marked by $H_3(T)$ should disappear at this point.

In order to understand better the physical origin underlying this change of slope occurring in the $m(H)$ curve at H_3 , we employed relaxation measurements of the irreversible magnetization. Specifically, we performed relaxation measurements for several fields at 10 K, where the new feature is not present, and at 40 K, where it is. In all the relaxation measurements the crystal was first cooled in zero field to the desired temperature, and then the magnetic field was raised to the desired H_i with a ramp rate $\mathcal{R} = H_0 \approx 100$ Oe/s. After the field was stabilized to H_i , the relaxation of $m(t)$ was measured within the time window $t_i = 10^2 \le t \le t_f \approx 10^4$ s. From the normalized relaxation rate $S = d \ln[-m(t)]/d \ln t$, we calculated the pinning potential as a function of the magnetic field at constant temperature. Figures 5 and 6 are semilogarithmic plots of the $m(t)$ variation (relaxation of magnetization) at $T=10$ and 40 K, with $H\|c$ axis and for 6.5 $\leq H_i \leq 50$ kOe, and $1 \leq H_i \leq 50$ kOe, respectively. The inverse relaxation rate, which in the framework of the interpolation formula is equal to $S^{-1} = U_c / k_B T + \mu \ln(t/t_0)$ at small time intervals, can give an estimation of U_c/k_BT . For very long periods of time, the slope of S^{-1} can lead to an estimation of μ .

At 10 K, the S^{-1} vs ln *t* curves (not shown) are nearly constant and increase as the corresponding magnetic field increases. This means that S^{-1} increases monotonically as the field increases (see Fig. 7). Only in small fields ($6 \leq H$ ≤ 9 kOe), S^{-1} is nearly field independent. In this field range, S^{-1} versus ln *t* curves display a small positive slope, which means that μ is small and positive. A rough estimation of μ gives 0.14 \pm 0.05. For higher fields, μ increases up to \approx 1.5.

Contrary to the relaxation measurements at 10 K, the measurements at 40 K show a different behavior in the various field ranges. For $H_i \ge 5$ kOe, a nearly linear variation of

FIG. 5. Semi-logarithmic plot of the magnetic moment *m* vs time (relaxation of magnetization) for an YBa₂Cu₃O_{7- δ} single crystal at $T=10$ K and for $H=6.5, 7.5, 9, 12, 14, 16, 18, 20, 22,$ 24, 26, 30, 40, and 50 kOe with the applied field $\mathbf{H}\|c$ axis.

 $m(t)$ is observed in a semi-logarithmic plot. Second, $m(H)$ for a given time follows the shape of the virgin magnetization curve at the same temperature only for $t_i \le t \le 800$ s. After that time, the second peak shifts to lower fields (see Fig. 6). Indeed, for $t > 10^3$ s the maximum in |m| occurs at 30 kOe, while for $t < 10^3$ s, it is at 40 kOe. Third, the slope of the m vs $ln(t)$ curves exhibits a pronounced crossover for $H \leq 3$ kOe. This feature is readily discernible in the measurement at $H=1$ kOe.

Figure 7 also shows the variation of $S^{-1}(t=10^3 \text{ s})$ as a function of the magnetic field at 40 K. $S^{-1}(t=10^3 \text{ s})$ increases abruptly with the field and displays a shoulder in the vicinity of H_3 . This shoulder could mean that the shape of the $m(H)$ curve does not change with time close to H_3 . Indeed, the open squares in Fig. 7 (representing the relaxed moment at 40 K after 10^4 s) demonstrate that the location of the H_3 field does not depend on time. For higher fields, S^{-1} continues to increase up to a maximum located below the second peak at this temperature. Above this maximum, S^{-1} decreases monotonically.

The enhancement of the relaxation barrier above H_{min} has been recently discussed by Koshelev and Vinokur⁵² for the case of the transition from an ordered quasilattice to an amorphous vortex configuration. In addition, with the estimation of the pinning potential, we can ascertain, that below the maximum in U_c , μ is positive, whereas above the maximum it is negative. The negative value of μ , in combination with the decrease in the pinning potential with the field, are in favor of the idea of plastic deformation of the flux lattice near the second peak.

It is worth noting that at high temperatures, below H_3 , the

FIG. 6. Semi-logarithmic plot of the magnetic moment *m* vs time (relaxation of magnetization) for an YBa₂Cu₃O_{7- δ} single crystal at $T=40$ K and for $H=1, 2, 3, 4, 5, 6.5, 8, 9, 11, 13.5, 15,$ 16, 18, 20, 24.8, 30, 34.83, 40, 45.2, and 50 kOe with the applied field $H\|c$ -axis. The upper panel shows an enlarged view of the measurement performed for the 1–5 kOe field range.

hysteresis loops are asymmetric (comparing the increasing and decreasing field branches), a behavior indicative of Bean-Livingston surface barriers. At lower temperatures, e.g., 40 K, there is a small asymmetry, but it is a small fraction of the total m at this field value (see inset of Fig. 1). This remark would suggest that at high temperatures (*T* >80 K) and low fields, the Bean-Livingston surface barriers are essential, and bulk pinning is present only in higher fields. Instead, at low temperature and fields, both bulk pinning and Bean-Livingston surface barriers are present.

III. DISCUSSION AND POSSIBLE INTERPRETATION

The change in the slope dm/dH at H_3 might correspond to the recently proposed field-driven disordering transition from Bragg glass, where the vortex lattice is weakly disordered without topological defects, to a highly disordered entangled solid at higher fields. Giamarchi and Le Doussal⁴⁸ introduced the idea of Bragg glass to distinguish from the glassy state without any crystalline order. Further, Ertas and Nelson,50 Vinokur *et al.,*⁵¹ and Koshelev *et al.*⁵² proposed that the onset of the second magnetization peak may correspond to the destruction of the Bragg glass by proliferation of topological defects upon raising the field. This is equivalent to increasing the effective disorder that favors dislocations. It is interesting at this point to note that the collective pinning theory¹⁹ suggests that at low field, the lattice is less ordered than in higher fields. This conclusion does not hold

FIG. 7. $S^{-1}(t=10^3 \text{ s})$ as function of magnetic field at 10 K (solid circles) and 40 K (open circles). Also shown is the magnetization curve at $40 K$ (solid line). The open squares denote the relaxed magnetic moment at 40 K after a time of $10⁴$ s. The peak of $S^{-1}(t=10^3 \text{ s})$ curve at 40 K is located below the second magnetization peak of the magnetization curves. Note that the curve *m*(*T* $=40$ K, $t=10^4$ s) reveals that the location of H_3 does not change with time.

when the distance between local minima of the random potential is much smaller than the typical intervortex spacing, as pointed out by Koshelev and Vinokur.⁵² In such a case (typical for high-temperature superconductors), the vortices have a wide choice of minima, which gives them an extra possibility to minimize their interaction energy and restore the lattice.

An important point that should be made is that our result is not a peculiarity of our crystal. The same feature, with some worth-mentioning differences, has also been observed by Nishizaki *et al.*⁵⁷ and Giller *et al.*⁵⁸ in an untwinned $YBa₂Cu₃O_{7-\delta}$ single crystal using local magnetization measurements. More specifically, the differences lie in the following points: (a) In our results the feature is not observed for $T < 20$ K, while Nishizaki and co-workers observed it for *T* > 53 K, and in Giller's data the feature appears at 40 K; ~b! Both Nishizaki *et al.* and Giller *et al.* show an increase of the characteristic magnetic field with temperature, whereas in our data, H_3 decreases slowly with increasing temperature; and (c) In the present measurements, the H_3 field is difficult to be defined when H_3 is close to H_{sp} . In the measurements of Nishizaki *et al.*, the H_3 line is terminated at the critical point, where the melting transition changes from first to second order. In Giller's *et al.* data, it is not clear where the H_3 line terminates. These differences may originate from the different amount of disorder in the samples, i.e., our crystal is more disordered than those of Nishizaki *et al.* and Giller *et al.*

Although the decrease of H_3 with temperature is predicted within a ΔT_c pinning mechanism, the curvature is not that predicted in the simplified model of Refs. 51,44. Probably, due to the simple character of the particular model, only the direction of change of H_3 with temperature and not the exact temperature variation can be predicted. Since a Δl (Refs. 19 and 58) type pinning gives an increase of $H_3(T)$ with temperature, the larger disorder of our crystal might lead to a combination of ΔT_c and Δl pinning mechanism, thereby producing the observed behavior. An open question is the physical reason for the disappearance of the feature at low temperatures. One could suppose that at low temperatures the pinning force is too strong to permit a regular lattice. As the temperature increases, the thermal fluctuations of the vortex lines smooth the quenched disorder potential that produces the pinning. As the amplitude of the thermal fluctuations increases beyond the extent of the vortex core, the vortex will experience an averaged disorder potential that corresponds to a reduced pinning force.

In conclusion, we have found an abrupt change in the slope of the irreversible magnetization in a detwinned $YBa₂Cu₃O_{7-\delta}$ single crystal for the temperature range 20 $\leq T < T^* \approx 83$ K. The *H*₃(*T*) line deduced from the sudden change of the slope in the $m(H)$ data might correspond to the field-driven disordering transition from the ''Bragg glass'' to the high-field topologically disordered phase. The second magnetization peak curve $H_{\text{sn}}(T)$ is governed by the relation $H_{\text{sp}}(T) = H_{\text{sp}}^0 (1 - T/T_c)^{\nu}$ from ≈ 30 K up to T^* . For $T>T^*$, the $H_{\text{sp}}(T)$ curve changes functional form and can be described by the relation $H_{\text{sp}}(T) = 60[1-(T/T_c)^4]^{1.4}$ proposed by Abulafia *et al.*⁵⁶

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