## Onset of Plasticity and Hardening of the Hysteretic Response in the Vortex System of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>

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A method based on partial magnetization loops has been used to study memory effects in detwinned  $YBa_2Cu_3O_{7-\delta}$  single crystals. These measurements have revealed the transition from a dislocation-free Bragg glass to a disordered vortex phase. We have mapped this boundary in the *B-T* phase diagram and have found it to be in proximity to the onset of the second magnetization peak. For fields above the transition line, metastable topological disorder invades the vortex system leading to a pronounced dependence of the critical current on the formation history of the vortex lattice. [S0031-9007(99)09470-3]

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The structure of the vortex solid (VS) in type-II superconductors subject to quenched disorder and thermal fluctuations is an issue of intense scientific interest. Following earlier arguments that disorder destroys long-range order in the Abrikosov lattice [1], recent theoretical investigations have established that at low magnetic fields quasi-longrange translational order can be maintained in an elastic phase, termed the Bragg glass [2], where dislocations are energetically irrelevant [2-4]. However, upon raising the applied field above a critical value, proliferation of topological defects is favored [2-4], thus transforming the VS into a highly disordered vortex glass, which is characterized by the absence of spatial order [5]. This scenario is also supported by numerical simulations [6]. The existence of such a transition in the VS was conjectured in several experimental investigations on high temperature superconductors [7-9]. However, up to now the position of this transition in the *B*-*T* phase diagram has remained unclear.

A distinct difference between a dislocation-free elastic lattice and a highly disordered vortex glass should be the observation of prominent thermomagnetic history effects in the latter, similar to other disordered systems like spin glasses. Such history effects have been observed in the transport and magnetic properties of the low temperature superconductors, in the region of the conventional peak effect close to the  $H_{c2}(T)$  line [10–15]. It has been suggested that in this region plastic deformations occur in the VS, leading to a dependence of the hysteretic response on the past history of the superconductor [10,13-15]. In YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>, although magnetic measurements [9,16,17] together with the hysteretic behavior of the driven vortex solid seen in recent transport studies [18] have indicated the importance of plasticity in this material, as yet no detailed studies of similar history effects have been performed.

In this Letter we present magnetization measurements designed to study history effects on pure, detwinned YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals. By employing a partial loop technique, we have found clear evidence for the theoretically proposed transition between a dislocation-free Bragg glass and a disordered vortex phase [2], as indicated by the proliferation of topological defects in the VS. The position of this transition in the B-T plane is found to be close to the onset of the second magnetization peak. Memory effects become important above this transition. The critical current density  $(J_c)$ , obtained from the magnetic hysteresis width, depends strongly on the maximum field to which the VS is exposed, leading to increased values for  $J_c$  after excursions into higher fields. For the highest fields used in our studies, saturation in the density of topological defects occurs, eliminating the history effects.

We report results acquired on an optimally doped detwinned YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> ( $\delta$  = 0.07) single crystal with dimensions  $1.69 \times 1.08 \times 0.09 \text{ mm}^3$ , grown by a conventional self-flux method using yttria-stabilized zirconia crucibles [19], a process that gives crystals of excellent purity [20]. The superconducting transition occurs at 93.6 K, with a width of  $\Delta T_c < 0.3$  K. Magnetic hysteresis measurements were carried out using an Oxford Instruments vibrating sample magnetometer for applied fields up to 12 T. We used a novel measuring procedure that involves partial magnetization loops (PLs) realized in two different ways. In method I the sample is cooled in zero field that is subsequently swept up to a maximum value  $B_{\text{max}}$  before it is decreased back to zero. In a series of runs  $B_{max}$  is gradually increased with a fine step finally reaching values above the irreversibility field. In the last case we obtain a complete hysteresis loop (CL). In method II the previous procedure is inverted: After cooling the sample in zero field, the field is increased up to 12 T, and then after decreasing it down

to a certain value  $B_{\min}$ , it is swept again to 12 T. This procedure is repeated for several values of  $B_{\min}$ . In all cases, after completion of each run, the sample is heated up into the normal state and then cooled again to the required temperature in zero field. The sweep rate of the magnetic field was varied in the range of 5–20 mT/sec.

Figure 1 shows hysteretic magnetization loops for the single crystal at 74 K. The solid line represents the complete magnetization loop, whereas with the dashed and dotted lines we show PLs obtained by methods I and II, respectively, for  $B_{\text{max}} = B_{\text{min}} = 2$  T. Remarkably the PLs do not follow the CL, and  $J_c$  depends on the measuring procedure. We conclude that field cycling of the superconductor can lead to increased critical currents, i.e., a hardening of the hysteretic response. Since in the elastic theory  $J_c$  is a single valued function of B and T, the hardening effect points to the presence of plastic deformations in the VS, which lead to a dependence of the lattice structure on the magnetic history of the sample. The path dependent current further implies that description by a simple Bean model, where  $J_c$  is determined only by the value of B and T, is not applicable. We should also note that the deviation of the PLs from the CL, extended over a range of some teslas, cannot be associated with the in-



FIG. 1. Magnetic hysteresis curves for the detwinned YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystal at 74 K. The solid line is the CL, whereas the dashed and dotted lines show PLs obtained with methods I and II, respectively, for  $B_{\text{max}} = B_{\text{min}} = 2$  T. The arrows indicate the direction of the field sweep. The open circles represent magnetization values in the descending leg of several PLs (method I) directly after reversal of the field. The inset shows *R* (defined in the text), plotted as a function of  $B_{\text{max}}$  (triangles) or  $B_{\text{min}}$  (circles) at 74 K.

fluence of an incomplete reversal of the Bean profile. Indeed simple calculations following Ref. [21] demonstrate that for the sample under study at 74 K the Bean penetration field is less than 9 mT, a value much too small to account for our observations. The memory effects are shown quantitatively in the inset of Fig. 1, where we plot as a function of  $B_{\text{max}}$  or  $B_{\text{min}}$  the ratio R of the relative difference in the hysteresis widths between the PLs and the CL:  $R = (\Delta M_{\rm CL} - \Delta M_{\rm PL}) / \Delta M_{\rm CL}$  [22]. As can be seen at a certain field the magnetization on the decreasing or increasing field branches of PLs obtained by methods I or II, respectively, can deviate by as much as 25% from the CL magnetization width, suggesting that the current density on these branches can be varied by as much as 50%. R is maximum in the intermediate field region (1 < B < 3 T), whereas for higher or lower fields the effect becomes weak and only small traces of the memory effects are observed. The width of the complete loop,  $\Delta M_{\rm CL}$ , increases with decreasing temperature much faster than  $\Delta M_{\rm CL} - \Delta M_{\rm PL}$ leading to lower R for decreasing T.

Another interesting feature depicted in Fig. 1 is that for 1 < B < 3.5 T the upper and lower branches of the CL are highly asymmetric. This asymmetry has been observed for pure single crystals, but has remained unexplained. In this Letter we present a clarification of this effect. Note that in this field range the Bean-Livingston [23] surface barrier cannot explain the observed asymmetry [24]. Indeed if we consider the magnetization of various PLs obtained by method I directly after reversal of the field, the asymmetry disappears and the lower branch of the full loop is almost perfectly reproduced (see Fig. 1, open circles). This demonstrates that the asymmetry originates from the history dependent hysteretic response of the superconductor.

The main result of our investigation is illustrated in Fig. 2, where we display in detail PLs obtained by method I at 76 K for  $1.5 < B_{\text{max}} < 6.5$  T. For  $B_{\text{max}} \leq 3.5$  T the PLs follow the same universal curve. In contrast above 3.5 T they start to deviate significantly, and the magnetization attains much higher values, approaching the descending leg of the CL. This behavior continues up to 5.5 T, where the PLs fall again onto the same line. This observation becomes clearer in Fig. 3, where we display for two different field sweep rates the difference  $\Delta M_{\rm suc}$  in the magnetization widths of successive PLs [22] as a function of  $B_{\text{max}}$  of the lower PL. A sharp increase of  $\Delta M_{\text{suc}}$ is seen at a sweep rate independent characteristic field  $B_{p1}$ before it drops again below the level of experimental resolution at a second field  $B_{sat}$ , above which the PLs coincide independently of the value of  $B_{\text{max}}$ .

According to recent theoretical predictions due to the enhancement of effective quenched pinning disorder, above a threshold field topological defects such as dislocations appear resulting in a defective plastic VS [2-4,6]. A direct consequence of the introduction of topological defects is the enhancement of the critical current, since the VS



FIG. 2. Several PLs obtained by method I at 76 K for  $1.5 < B_{\text{max}} < 6$  T. Arrows indicate the direction of the field sweep. The inset shows the CL for this temperature with the box indicating the field region corresponding to the illustrated PLs.

can adapt more easily to the pinning potential [2]. Increasing the field during an isothermal magnetization loop in the regime where dislocations are present forces the VS into an increasingly disordered state. When the field is decreased dislocations remain trapped in local energy minima, thus exhibiting metastability (for a detailed discussion, see Ref. [10]). Hence the amount of topological



FIG. 3. The difference  $\Delta M_{\text{suc}}$  of the magnetization widths of successive PLs at T = 76 K is plotted as a function of  $B_{\text{max}}$  for two different field sweep rates as indicated on the graph. The arrows mark the positions of  $B_{\text{pl}}$  and  $B_{\text{sat}}$ .

disorder should not vary reversibly with the field, and one should expect a path dependent hysteresis as is evident from Fig. 1. The VS in the descending branch of the CL tends to retain the dislocation network from the high field state leading to higher critical currents as compared to the PLs. This effect persists until the field is reduced enough to fully expel out residual topological disorder thus erasing any memory on the formation history, as demonstrated by the coincidence of the PLs with the CL at low  $B_{\text{max}}$ and  $B_{\text{min}}$ .

The significance of our measuring procedure is that it allows us to determine the point where dislocations first invade the vortex system. Indeed when  $B_{\text{max}}$  is not high enough to give excursions of the VS beyond the elastic regime, the history effects are not observable due to the reversible structural properties of an elastically distorted lattice [25]. Consequently the PLs must follow the same universal line. This is exactly what we observe below the threshold field  $B_{p1}$  as can be seen in Fig. 2. For  $B_{max} >$  $B_{p1}$  and in a narrow field range the PLs strongly deviate from the universal curve exhibiting prominent history dependence. This clearly demonstrates the introduction of metastable topological disorder and the onset of plasticity in the VS. Increasing the field beyond  $B_{pl}$  enhances disorder in the lattice until  $B_{\text{sat}}$  is reached. At this point saturation in the density of dislocations occurs, and the amount of disorder is only negligibly affected by the changing field. For this reason at high B the PLs closely coincide with each other as well as with the CL.

The features characterizing the VS below  $B_{p1}$  (i.e., elastic behavior with undetectable topological disorder) are evidence for the existence of the Bragg glass phase proposed in Ref. [2]. Hence we interpret the  $B_{p1}(T)$  line as the transition from the Bragg glass to the vortex glass [2–4,6]. This line as well as the locus of the values for  $B_{sat}$  is shown in Fig. 4. Included are also the lines corresponding to the magnetization peak  $B_p$ , and the peak onset  $B_{on}$  [26] as well as the melting line, which was obtained from



FIG. 4. Position of  $B_{\rm p}$ ,  $B_{\rm pl}$ ,  $B_{\rm on}$ , and  $B_{\rm sat}$  in the *B*-*T* phase diagram. The corresponding melting line is also displayed.

resistivity measurements on an identical crystal from the same batch [27].  $B_{pl}$  is temperature independent for T < 76 K. However above 76 K it shows a sharp increase. This behavior is in support of theoretical predictions for the transition line separating the Bragg glass and vortex glass phases [2,3]. Topological defects appear when the pinning energy dominates over the elastic energy [3]. At elevated temperatures, thermal motion of the vortex cores reduces the pinning by point disorder. Therefore higher fields are required for the introduction of dislocations in the vortex solid resulting in the upward curvature of the  $B_{pl}$  line. Finally we find that the  $B_{sat}$  and  $B_p$  lines are located in the disordered plastic state of the VS. Further theoretical and experimental work is required to clarify the behavior in this regime.

We also studied the influence of time on the metastable disorder in the descending branch of the CL. In the decreasing field leg of a CL, the field was stopped at certain values and after waiting a given time, the field sweep was continued back to zero. Following this procedure with a waiting time of two hours, the magnetization on the decreasing field M(B) curve was found to be reduced by less than 4% of the CL magnetization width. This demonstrates that although annealing of dislocations does occur, the effect is rather weak illustrating that the dislocation network in the VS is stable over long time scales.

In conclusion, we studied history effects in the magnetic response of pure YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals. By using a special technique based on partial magnetization loops we were able to detect the transition from the Bragg glass to a disordered vortex phase. The position of this transition was identified in the *B*-*T* phase diagram, and it was found to lie in the vicinity of the onset of the second magnetization peak. For field excursions above this line a dependence of the current density on the past history of the superconductor is seen, which diminishes at high fields due to saturation in the density of dislocations.

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