History Effects and Phase Diagram near the Lower Critical Point in $YBa_2Cu_3O_7$ Single Crystals

A. A. Zhukov,* P. A. J. de Groot, S. Kokkaliaris, and E. di Nicolo
Department of Physics and Astronomy, University of Southampton, Southampton SO17 1BJ, United Kingdom

A. G. M. Jansen, E. Mossang, G. Martinez, and P. Wyder
Grenoble High Magnetic Field Laboratory, MPIF-CNRS, BP 166, 38042 Grenoble Cedex 09, France

T. Wolf and H. Küpfer
Forschungszentrum Karlsruhe, Institut für Technische Physik, D-76021 Karlsruhe, Germany

H. Asaoka
Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki 319-11, Japan

R. Gagnon and L. Taillefer
Department of Physics, McGill University, Montreal, Canada H3A 2T8
(Received 7 November 2000; published 19 June 2001)

Using a sensitive torque magnetometer we have studied magnetization curves for untwinned overdoped $YBa_2Cu_3O_7$ single crystals in fields of up to 28 T. We demonstrate the existence of history effects below the lower critical point and provide a full demarcation of the Bragg-glass phase. A pronounced symmetry is observed in the behavior of the phase lines, irreversible magnetization, and value of the magnetization jump near both critical points.

DOI: 10.1103/PhysRevLett.87.017006 PACS numbers: 74.60.Ge, 74.60.Jg, 74.72.Bk

The vortex phase diagram of high temperature superconductors (HTSC) has been a subject of intense interest in recent years [1–9]. Its most prominent feature is the first-order solid-liquid melting transition [1–4]. The transformation from the vortex solid into the liquid state induces sharp anomalies in the resistance [1], magnetization [2,3], and specific heat [4] of the superconductor. Similar to melting of ice into water, the vortex system also shrinks with increasing temperature at the melting point. At low temperatures the first-order transition terminates at the upper critical point [5]. This results in the appearance of a pronounced peak effect [6–9] in critical current closely related to an order-disorder transition. Because of the presence of pointlike defects the ordered state is a Bragg glass [10], which does not possess the perfect translational order of the hexagonal lattice but retains full topological order with six nearest neighbors for every vortex. Above the transition the disordered phase can better adjust to the pinning system and this results in an increase of the critical current. Prominent history effects [11] appear at the onset of the peak and mark a destruction of the Bragg-glass phase. These effects were shown [11,12] to offer an excellent method to locate this boundary deep in the solid vortex state. Previous research of the phase diagram [5–7] has focused mostly on the upper critical point B_{umc}. However, transport and specific heat measurements demonstrate that the first-order melting transition also terminates at high temperatures, for fields below a lower critical point B_{lmc} [13–15]. In this paper we report the existence of history and peak effects near B_{lmc} and provide a full mapping of the Bragg-glass phase. We observe a pronounced symmetry in the behavior of the phase lines, irreversible magnetization, and value of the magnetization jump near both critical points.

We have studied three flux-grown $YBa_2Cu_3O_y$ single crystals [16]. They were prepared by three different groups using Y stabilized Zr and Y_2O_3 crucibles. Crystal ZY was in an as-grown untwinned state. The two others, named AS and D3, were detwinned by applying a uniaxial pressure. Using high (370–480 bars) oxygen pressure annealing at low (400–500 °C) temperature for a long time (140–180 h) the crystals were oxidized to the state $y = 7.00$ with deficiency less than 0.003 according to the isotherms [17].

Magnetic hysteresis measurements have been performed using a cantilever capacitance torque meter [18] in magnetic fields up to 28 T. For the conventional presentation of the peak effect, the measured torque values τ were used to calculate the magnetic moment, using the relation $m = \tau / B \sin\theta$, where θ is the angle between the magnetic field and the magnetic moment. Because of the geometrical confinement of the shielding currents in thin samples [19] the direction of the moment essentially coincides with the c axis.

Figure 1 shows typical behavior of the magnetization curves for the fully oxidized $YBa_2Cu_3O_y$ single crystal AS. The results clearly demonstrate the existence of three different temperature intervals related to the two critical points. Approaching from above $B_{\mathrm{umc}} = 24$ T the peak in the irreversible magnetization decreases in width with increasing temperature and transforms to an extremely narrow peak that vanishes at the upper critical point. This

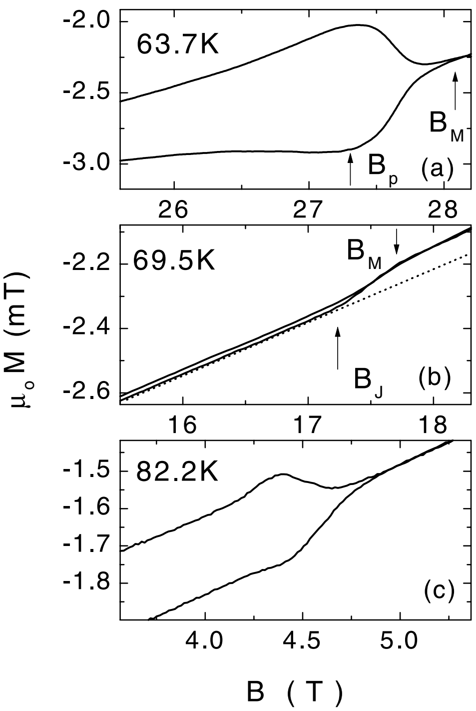

FIG. 1. Magnetization curves for crystal AS at different temperatures: (a) above the upper critical point $B_{\rm umc} = 24$ T, (b) between these points, and (c) below the lower critical point $B_{\rm lmc} = 5.5$ T.

extremely sharp peak in the current has a width even smaller than that observed in conventional superconductors [20]. The decreasing branch of the peak $(B_M - B_p)$ essentially has the same width as the first-order jump [Fig. 1(a)].

In the intermediate temperature region [Fig. 1(b)] we observe essentially reversible magnetization with a jump at the first-order melting transition. As was shown previously by structure sensitive measurements [21], an ordered vortex Bragg glass transforms to a disordered vortex liquid at this point. We find two different types of behavior in this region: either with a very small irreversibility or with an irreversibility close to a jump value [22]. To characterize the first-order melting transition we use the field parameters B_j and B_M corresponding to the beginning and the end of the jump [Fig. 1(b)].

At high temperatures [Fig. 1(c)] the irreversible magnetization starts to increase drastically and a very narrow peak effect develops again, for $B \leq B_{\rm lmc} = 5.5$ T. The peak position B_p essentially coincides with the onset of the jump B_j extrapolated from the first-order region. Going away from the critical points outside the region of the first-order transition, the irreversible part increases drastically and the peak broadens. Such broadening is already known for the upper critical point [8,9]. We find a similar behavior for the lower critical point. Close to T_c the peak transforms to a broad fishtail.

The disappearance of a sharp jump in the reversible magnetization was associated with a transition to a second-order melting [14]. It is accompanied by the appearance of a narrow peak effect. So we can use this behavior to detect critical points in our measurements. For crystal AS the transition fields of $B_{\rm umc} = 24$ T and $B_{\rm lmc} = 5.5$ T determine the upper and lower critical points, respectively, where the first-order transition terminates. Although the value of the upper critical field shows significant variations in different samples (15, 19, and 24 T for ZY, D3, and AS, respectively), the lower critical point for all studied samples has the same value.

It is already well established that the upper critical point is a locus for several phase lines separating vortex liquid, ordered Bragg glass, and disordered solid states. Recently we observed large history effects in the transition region between ordered Bragg glass and disordered phases [11]. These effects clearly mark the order-disorder transition. In this Letter we report for the first time a similar behavior below the lower critical point. This experiment is based on the partial magnetization loop technique [23]. In this technique a series of magnetization loops are generated with the maximum applied field increasing by a small step for every consecutive cycle. As can be seen from Fig. 2 the partial magnetization loops coincide for field excursions, which remain below 0.15 T. For hysteresis curves with higher maximum field a significant difference extending for a wide field range is observed. It is induced by supercooling of a disordered phase as the result of hysteretic behavior of dislocations in the vortex system. The disordered phase experiences a stronger pinning and shows a larger irreversible magnetization. The difference between the magnetization loops is quantified in the inset in Fig. 2 and demonstrates a sharp onset of history effects above

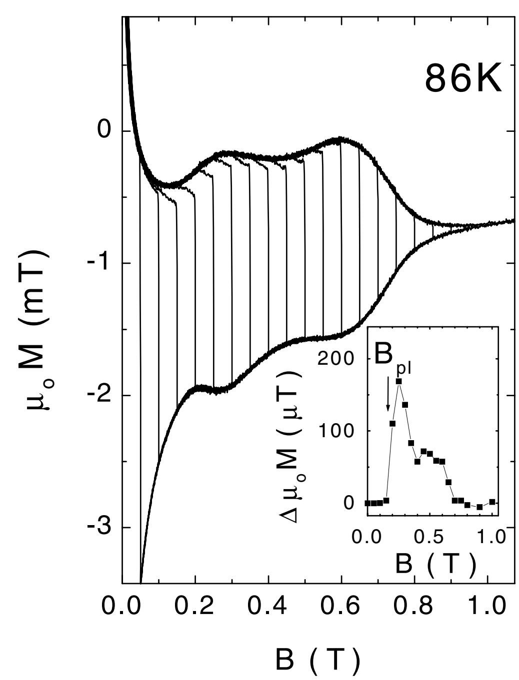

FIG. 2. Partial magnetization loops for crystal ZY at 86 K. The inset demonstrates the difference between the consecutive partial loops [11] after reversal of the critical state. The onset field of the history effects $B_{\rm pl}$ is shown by the arrow.

B_{pl}. The history effects persist until 0.6 T when a completely disordered phase is created [11].

It should be noted that for a significant temperature interval a splitting of the peak effect was observed. Such splitting was already discussed in Ref. [8] and, we suggest, originates from the intermixture of different vortex states: liquid, disordered, and ordered Bragg-glass phases [22,24–26]. Recently, strong evidence was obtained that the transition from a Bragg glass to a disordered state is realized via an intermediate state which corresponds to a mixture of both states [24,25]. The characteristic scale of the mixture would be either microscopic or macroscopic depending on the purity and pinning in the sample. Also a broad intermediate region is expected for the solid to liquid transition [26]. The overlapping of these two regions is a probable reason for the peak splitting. In this case the first peak may be related to the melting of the Bragg glass and the second peak reflects a transition to the liquid state. Of course this problem requires further detailed investigation and is outside the scope of this Letter.

As can be seen from Fig. 3 the behavior of the peak effect is highly symmetrical for both critical points. The symmetry of the vortex phase diagram becomes even more pronounced after normalization by the melting field, as demonstrated in the inset in Fig. 3. This suggests a similarity in the mechanism of the peak effect in both regions. Similar to the behavior in the low temperature region [11] the onset of history effects B_{pl} essentially coincides with the onset of the peak effect B_{on}. This provides a full demarcation of the Bragg-glass phase near both critical points.

An important characteristic of the melting transition is the value of the jump in the reversible magnetization δM_j.

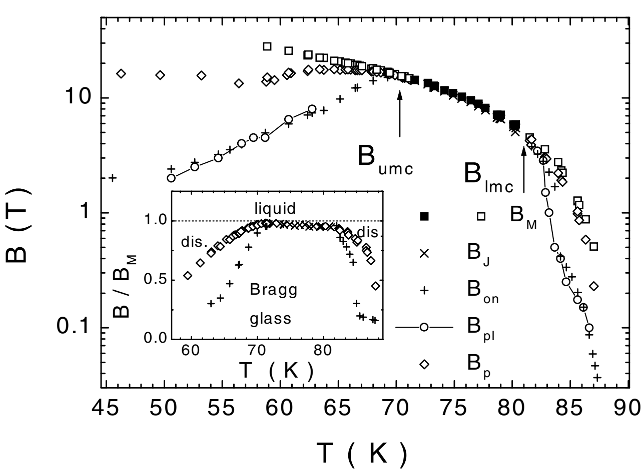

FIG. 3. Phase diagram for crystal ZY. This figure represents the characteristic fields: melting transition B_M, position of the peak B_p, of the onset of the peak effect B_{on}, history effects B_{pl}, and of the melting jump B_j. For the region of the first-order transition B_M was determined by the end of melting jump in the reversible magnetization (closed squares), and outside this region B_M was related to the vanishing point of the irreversibility (open squares). The inset shows the characteristic fields normalized by the melting field B_M value and the regions of liquid, Bragg glass, and disordered vortex phase (dis.).

As can be seen in Fig. 4 this jump vanishes on approaching both critical points. Simultaneously the irreversible magnetization at the peak $\Delta M_p \propto j_c$ starts to increase drastically. Again, the curves above and below the corresponding critical points are quite symmetrical. It is natural to relate this behavior to an increasing influence of the disorder in both cases.

The previous analysis of the vortex phase diagram was concentrated mainly on the behavior near the upper critical point [10]. The transition between different vortex phases is governed by the relation between the energy of vortex-vortex interaction $E_{v\text{-}v}$, pinning energy E_p, and temperature k_BT. For pure crystal the $E_{v\text{-}v}$ energy is dominant at sufficiently low fields and drives the vortex system into the ordered Bragg-glass phase. Therefore, at low magnetic fields B the relation between $E_{v\text{-}v}$ and k_BT determines melting from an ordered Bragg-glass (dominant vortex-vortex interaction) to a disordered liquid (dominant temperature disorder). With increase of B the vortex-vortex interaction energy drops faster ($\propto B^{-1/2}$) than the pinning energy by pointlike disorder ($\propto B^{-1/10}$). Then, at the upper critical point, all three energies become equal. At lower temperatures, two transitions take place. First the Bragg-glass phase transforms to a disordered glass at the point where the pinning interaction becomes dominant and only after the decreasing pinning energy ($\propto B^{-1/10}$) reaches k_BT, a transition to a liquid state is realized. The experimental data near the upper critical point are in agreement with such a scenario.

The lower critical point may also be understood in the framework of such an analysis [10]. One should take into account that only in very low magnetic fields the vortex-vortex interaction energy changes to exponentially decreasing with field behavior. Then again we may expect that the pinning energy becomes dominant (especially in the presence of strong pinning centers), and the vortex lattice may melt to a glass state [27]. Another possible interpretation of the lower critical point could be reentrant melting caused by the entropy factor [28]. However, both

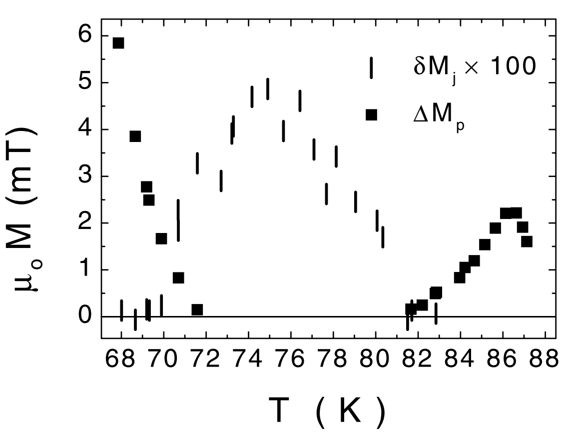

FIG. 4. Temperature dependence of the melting jump value in reversible magnetization δM_j and of the irreversible magnetization at the peak ΔM_p for crystal ZY.

cases correspond to very small fields below $\mu_0 H_{c1}$. Therefore, this cannot explain our high value of $B_{\rm lmc} = 5.5$ T.

The presence of strong defects may cause the appearance of the lower critical point. In this case when the energy $E_{\rm sp}$ of strong pinning is larger than the vortex-vortex interaction energy of the vortex bundle, then a low-field disordered phase will form. This explains the existence of a disordered phase below the lower critical point in columnar irradiated samples [15]. However, this mechanism cannot be realized in our untwinned samples.

We should stress the anomalous behavior of the lower critical point, which increases with the decrease of disorder in the crystal lattice caused by oxygen vacancies [14]. In crystal AS we find $B_{\rm lmc} \approx 0$ for $y = 6.94$, and $B_{\rm lmc}$ increases to 5.5 T in the fully oxidized state. In addition, there is yet another anomaly. The similar values of the lower critical point in all the samples studied, the consistency with previous results for a fully oxidized sample by specific heat measurements [14] (5 T $< B_{\rm lmc} <$ 6 T), and very recent magnetic measurements [29] ($B_{\rm lmc} = 5$ T) suggest that the $B_{\rm lmc}$ value is rather independent of the sample quality. This consequently excludes other extrinsic defects because it is quite unlikely that the samples with considerably different $B_{\rm umc}$ have the same defect concentrations, especially as they were prepared in different groups. Taking into account that $B_{\rm lmc}$ strongly increases with the decrease of the oxygen disorder we may exclude the influence of oxygen clusters on $B_{\rm lmc}$ in similarly annealed samples. Our results also disagree with numerical studies of Ref. [30], which predicts the existence of a low-field critical end point. According to these calculations, the first-order line and critical point should exist for any oxygen content that also differs from the experiment.

Therefore, our results suggest the existence of a disorder with an intrinsic origin, whose influence increases with oxygen contents approaching $y = 7$. This may be caused by the exotic nature of superconductivity in HTSC. It is known that a pseudogap [31], which is suggested to be related to charge or spin excitations, is present in these materials. In the overdoped state the gap energy E_g falls below $k_B T_c$. So at sufficiently high temperatures, $T \geq E_g/k_B$, a spin/charge disordered state appears, which introduces "intrinsic" pinning of vortices causing the appearance of a critical point at low fields.

In summary, using a sensitive torque magnetometer we have studied magnetization curves for three untwinned overdoped $YBa_2Cu_3O_7$ single crystals in magnetic fields up to 28 T. Our results reveal clearly the transformation of the first-order jump to the peak effect near both critical points. We have demonstrated the existence of history effects below the lower critical point and have provided a full mapping of the Bragg-glass phase. A pronounced symmetry is observed in the behavior of the phase lines, irreversible magnetization, and jump value. Our results suggest an intrinsic origin for the lower critical point.

This work was supported by the U.K. Engineering and Physical Sciences Research Council, by the TMR Program of the European Community, and by INTAS Grant No. 97-1717. The authors are grateful to J.R. Cooper, M.A. Moore, and E. Zeldov for useful discussions and comments.

*On leave from the Chemistry Department, Moscow State University, 117234 Moscow, Russia.

[1] H. Safar *et al.,* Phys. Rev. Lett. **69**, 824 (1992); W.K. Kwok *et al., ibid.* **69**, 3370 (1992).
[2] H. Pastoriza *et al.,* Phys. Rev. Lett. **72**, 2951 (1994).
[3] E. Zeldov *et al.,* Nature (London) **375**, 373 (1995).
[4] A. Schilling *et al.,* Nature (London) **382**, 791 (1996); M. Roulin *et al.,* Science **273**, 1210 (1996).
[5] H. Safar *et al.,* Phys. Rev. Lett. **70**, 3800 (1993); D. Lopez *et al., ibid.* **80**, 1070 (1998).
[6] W.K. Kwok *et al.,* Phys. Rev. Lett. **73**, 2614 (1994); H. Safar *et al.,* Phys. Rev. B **52**, 6211 (1995).
[7] B. Khaykovich *et al.,* Phys. Rev. Lett. **76**, 2555 (1996).
[8] K. Deligiannis *et al.,* Phys. Rev. Lett. **79**, 2121 (1997).
[9] H. Küpfer *et al.,* Phys. Rev. B **58**, 2886 (1998); T. Nishizaki *et al.,* Phys. Rev. B **58**, 11 169 (1998).
[10] T. Giamarchi and P.L. Doussal, Phys. Rev. Lett. **72**, 1530 (1994); Phys. Rev. B **55**, 6577 (1997); D. Ertas and D.R. Nelson, Physica (Amsterdam) **272C**, 79 (1996); V. Vinokur *et al.,* Physica (Amsterdam) **295C**, 209 (1998); A. van Otterlo *et al.,* Phys. Rev. Lett. **81**, 1497 (1998).
[11] S. Kokkaliaris *et al.,* Phys. Rev. Lett. **82**, 5116 (1999).
[12] M. Gaifullin *et al.,* Phys. Rev. Lett. **84**, 2945 (2000); D. Giller *et al., ibid.* **84**, 3698 (2000); K. van der Beek *et al., ibid.* **84**, 4196 (2000).
[13] U. Welp *et al.,* Phys. Rev. Lett. **76**, 4809 (1996).
[14] M. Roulin *et al.,* Phys. Rev. Lett. **80**, 1722 (1998).
[15] W.K. Kwok *et al.,* Phys. Rev. Lett. **84**, 3706 (2000); L.M. Paulius *et al.,* Phys. Rev. B **61**, R11 910 (2000).
[16] Th. Wolf *et al.,* J. Cryst. Growth **96**, 1010 (1989); R. Gagnon *et al.,* J. Cryst. Growth **121**, 559 (1992); H. Asaoka *et al.,* Jpn. J. Appl. Phys. **32**, 1091 (1993).
[17] D.J.L. Hong and D.M. Smith, J. Am. Ceram. Soc. **74**, 1751 (1991).
[18] M. Willemin *et al.,* Phys. Rev. Lett. **81**, 4236 (1998).
[19] F. Hellman *et al.,* Phys. Rev. Lett. **68**, 867 (1992); A.A. Zhukov *et al.,* Phys. Rev. B **56**, 2809 (1997).
[20] R. Meier-Hirmer *et al.,* Phys. Rev. B **31**, 183 (1985); R. Wördenweber and P. Kes, Phys. Rev. B **34**, 494 (1986); S. Bhattacharya and M. Higgins, Phys. Rev. Lett. **70**, 2617 (1993).
[21] K. Harada *et al.,* Phys. Rev. Lett. **71**, 3371 (1993); S.T. Johnson *et al.,* Phys. Rev. Lett. **82**, 2792 (1999).
[22] A.A. Zhukov *et al.,* Physica (Amsterdam) **341–348C**, 1027 (2000).
[23] S.B. Roy and P. Chaddah, J. Phys. **9**, L625 (1997).
[24] A.A. Zhukov *et al.,* Phys. Rev. B **61**, 886 (2000).
[25] Y. Paltiel *et al.,* Nature (London) **403**, 398 (2000).
[26] A. Soibel *et al.,* Nature (London) **406**, 282 (2000).
[27] J. Kierfeld, Physica (Amsterdam) **300C**, 171 (1998).
[28] D.R. Nelson, Phys. Rev. Lett. **60**, 1973 (1988).
[29] T. Nishizaki *et al.,* Physica (Amsterdam) **341–348C**, 957 (2000).
[30] A.K. Kienappel and M.A. Moore, cond-mat/9804314.
[31] T. Timusk and B. Statt, Rep. Prog. Phys. **62**, 61 (1999).