Vortex lattice melting into a disentangled liquid followed by the 3D-2D decoupling transition in YBa₂Cu₄O₈ single crystals

X. G. Qiu,* V. V. Moshchalkov, and Y. Bruynseraede

Laboratorium voor Vaste-Stoffysica en Magnetisme, Katholieke Universiteit Leuven, Celestijnenlaan 200D, B-3001 Leuven, Belgium

J. Karpinski

Laboratorium für Festkörperphysik, ETH-Hönggerberg, CH-8093, Zürich, Switzerland

(Received 21 April 1998)

A sharp resistance drop associated with vortex lattice melting was observed in high quality $YBa_2Cu_4O_8$ single crystals. The melting line is well described by the anisotropic Ginzburg-Landau theory. Two thermally activated flux flow regions, which were separated by a crossover line $B_{cr}(T) = 1406.5(1 - T/T_c)/T(T_c)$ = 79.0 K, B_{cr} in unit of Tesla), were observed in the vortex liquid phase. Activation energy for each region was obtained and the corresponding dissipation mechanism was discussed. Our results suggest that the vortex lattice in YBa₂Cu₄O₈ single crystal melts into disentangled liquid, which then undergoes a two- to threedimensional decoupling transition. [S0163-1829(98)03237-8]

The vortex dynamics in the mixed state of high- T_c superconductors remains a subject of intense research because of its fundamental importance for physics of the vortex matter.^{1–3} It was predicted theoretically^{4–6} and confirmed experimentally^{7–15} that the vortex system undergoes a second-order transition from a high-temperature vortex liquid to a low-temperature vortex glass or Bose glass in superconductors with strong disorders. In a clean superconductor a first-order transition from vortex liquid to a well-ordered vortex lattice occurs.

Vortex melting in high-quality high T_c superconducting single crystals has been detected by various techniques in-cluding torsion oscillator,⁸ transport,^{9,10} magnetization,^{11,12} neutron scattering,¹³ μ spin rotation,¹⁴ and calorimetric measurements.¹⁵ Central to the question of flux melting is: does the vortex lattice melt via a first-order transition? How does disorder influence the melting transition?¹⁶ It was found by Safar et al.9 and by Kwok et al.10 that the resistive transition of high-quality YBa₂Cu₃O₇ (Y123) single crystal showed a sharp discontinuity and that the transition exhibited a hysteric behavior. They attributed this phenomenon to flux melting. Although their explanation was questioned by Jiang et al.¹⁷ who found that the hysteresis was not necessarily an indication for a melting transition, it could be resulted from a nonequilibrium behavior seen in the resistive transition. Subsequent magnetization^{11,12} and calorimetric¹⁵ measurements convincingly proved that the melting transition was indeed a first-order one, i.e., discontinuity in the internal energy at the melting temperature was observed. By doing simultaneous transport and magnetization measurements, Welp et al.¹² and Fuchs et al.¹⁸ found that the temperatures, where the resistance jumped, coincided with the melting temperature obtained from magnetization measurements on Y123 and Bi₂Si₂CaCu₂O₈(Bi2212) single crystals, respectively. The role of disorder in the melting transition has been checked by Kwok et al.¹⁰ and by Fendrich et al.¹⁹ They found that strong pinning from twin boundaries or artificially introduced defects drove the first-order melting transition into a second order glass transition.

Another important question related to the flux melting is how the melting propagates. Will the vortices lose their coherence along the *c* direction during the melting transition or will they keep the *c*-axis integrity and then lose it afterwards, at higher temperature, when thermal fluctuations destroy long-range correlations along the *c* direction?²⁰ This question has been recently pursued by employing the flux transformer configuration. The results remain still controversial. Lopez *et al.* and Fuchs *et al.* found a coincidence of the melting transition and decoupling one in Y123 and Bi2212 single crystals, respectively.²¹ However, Wan *et al.* and Keener *et al.* did similar measurements on Bi2212 single crystals and concluded that the melting took place in a twostage fashion.²²

Previous studies of the vortex melting transition have been concentrated on Y123 and Bi2212 single crystals. Up to now, no detailed measurements on YBa₂Cu₄O₈ single crystals have been reported. Meanwhile, the dissipation in the liquid state after the melting transition still remains to be clarified. The main purpose of this work is to show that in YBa₂Cu₄O₈ single crystals, vortex lattice melts into liquid via a first-order transition. It was also found that the liquid state after the melting transition could be separated into two regions that were governed by different dissipation mechanisms. Our results support the picture that *the vortices first lose their transverse coherence at the melting temperature, this loss being followed by a disappearance of correlation in longitudinal direction at higher temperature.*

YBa₂Cu₄O₈ (Y124) single crystals were grown by traveling solvent floating-zone method. The details for the single-crystal growth were published elsewhere.²³ T_c of the single crystals was about 76–78 K. Three single crystals were used for similar measurements. Each crystal was carefully cleaved to obtain optically flat surfaces with the *c* axis normal to the sample surface. Gold wires were attached to the surface of the crystal by using Pt epoxy. Then the crystal was heated in air at 100 °C for 1 h, yielding a typical contact resistance below 0.5 Ω . The resistance was measured by

8826



FIG. 1. Arrhenius plot of resistance vs 1000/*T* at different applied magnetic fields. Solid lines are experimental data. From left to right: H=0.2, 0.5, 1, 2, 3, 4, 5, 6, 7, 8, 10, 12 T. The dashed lines are the fits with TAFF theory. *h* and *l* are used to identify the two TAFF regions. T_{cr} shows how the decoupling transition temperature is determined. Inset: determination of T_m from the dR/dT vs *T* plot derived from the *R*-*T* curve for H=0.5 T.

four-probe low-frequency ac lock-in technique with an excitation current of 0.3 mA at 17 Hz in the *ab* plane. The magnetic field was generated by a 15-T Oxford superconducting magnet. The direction between the crystalline *c* axis and the magnetic field was adjusted by rotating the sample holder with an accuracy of 0.2° . The crystal we report on here had dimensions of $1.2(l) \times 0.3(w) \times 0.05(t)$ mm³. It had a T_c of 77.6 K and transition width of about 1.2 K at zero field.

Figure 1 shows the Arrhenius plot for the temperature dependence of the resistance measured at different fields up to 12 T applied perpendicular to the *ab* plane. A sharp jump in the resistance R with a magnitude of $R/R_n \sim 10\%$ is clearly visible at each magnetic field. The magnitude of the resistive drop lies between those of Y123 ($\sim 20\%$) and Bi2212 ($\sim 0.5\%$), suggesting a possible dependence of the R(T) jump on the anisotropy of the materials since Y124 has an anisotropy between those of Y123 and Bi2212. We also observed small hysteresis in the resistive jump upon warming up and cooling down. We measured resistive transition at 4 T with different excitation currents of 0.1 mA, 0.3 mA, and 1 mA, respectively. The *R*-*T* curves measured at 0.1 mA and 0.3 mA were essentially identical. However, that of 1 mA deviated from those of 0.1 mA and 0.3 mA. Similar hysteretic resistance jumps were earlier observed on Y123 and Bi2212 single crystals and were attributed to a first-order flux melting in clean superconductors.9,10 To quantify the melting transition, we determine the melting temperature T_m as the temperature where a dR/dT vs T plot shows a sharp peak. The inset in Fig. 1 demonstrates how the melting points T_m are determined. The obtained melting temperatures are plotted against the corresponding magnetic fields in Fig. 2. The melting line can be described by the empirical formula $B_m(T) \sim (1 - T/T_c)^{\alpha}$ with the exponent α between 1



FIG. 2. The *H*-*T* phase diagram for vortices in YBa₂Cu₄O₈ single crystal. Solid line and dashed line are best fittings to the experimentally obtained melting line (filled circles) and the three-to two-dimensional decoupling line (open circles) with $B_m(T) = 31.5(1 - T/77.6)^{1.47}$ (solid line) and $B_{cr}(T) = 1406.5(1 - T/79.0)/T$ (dashed line), respectively.

and $2^{.8-10}$ We find that the melting line is best fitted by $B_m(T) = 31.51(1 - T/T_c)^{1.47}$, $T_c = 77.6$ K being the transition temperature at zero field. This result agrees well with that obtained by Kwok *et al.*¹⁰ who found $B_m(T) = 103(1 - T/92.33)^{1.41}$ on Y123 single crystal when the field was applied parallel to the *c* axis.

To check further an interpretation of the $B_m(T)$ line in the framework of the vortex melting scenario, we measured the resistive transition with the magnetic fields applied at different angles to the *ab* plane, while fixing the field magnitude at 4 T. The angular dependence of the melting transition is shown in Fig. 3, where clear resistance jumps can be seen at all the angles. It is interesting to note that at large angles $(\theta > 70^{\circ}, \theta$ being the angle between the c axis and the applied magnetic field), the resistive transition shows an anomalous behavior above the kink. This anomaly was more clearly visible in our flux flow measurements (R-B curves). Such kind of anomalous behavior could be due to the vortex tilting instability at large angles and under a magnetic field of a few Tesla, which induces a competition between intrinsic pinning and vortex interaction. Following the same procedure, we have obtained the angular dependence of the melting transition temperatures at 4 T as shown in the inset of Fig. 3.

For a three-dimensional vortex lattice, the melting transition occurs when the shear modulus c_{66} goes to zero, which is determined by using a phenomenological Lindermann criterium, i.e., when the mean square-root amplitude $\sqrt{\langle u^2 \rangle}$ of the displacement of the vortex from its equilibrium position is larger than a certain portion of the lattice constant, $\sqrt{\langle u^2 \rangle} > c_L a_0$, where c_L is the Lindermann number, a_0 being the vortex spacing. According to the theory for an anisotropic superconductor, $T_m(\theta)$ is given by⁶

$$k_B T_m = \frac{\Phi_0^{5/2} c_L^2}{4 \pi^2 \lambda_{ab}^2 (T_m) B^{1/2} \varepsilon^{1/4}(\theta)},$$
 (1)



FIG. 3. Angular dependence of the resistive transition at a fixed magnetic field of 4 T. From left to right: $\theta = 0, 20, 30, 40, 50, 60, 70, 80, 85, 88^{\circ}$. Inset: Angular dependence of the melting transition in YBa₂Cu₄O₈; solid line is fitting by using anisotropic Ginsburg-Landau theory, Eq. (1), with a Lindermann number $c_L = 0.14$.

where $\varepsilon(\theta) = \sin^2(\theta) + \gamma^2 \cos^2(\theta)$, $\Phi_0 = 2.07 \times 10^{-7}$ G cm² is the flux quantum, λ_{ab} is the penetration depth in the *ab* plane, and $\gamma = \lambda_c / \lambda_{ab}$ the anisotropy parameter. A best fit to the obtained data by using Eq. (1) with $\lambda_{ab} = 2000$ Å (Ref. 24) is achieved when $\gamma = 13.6$, $c_L = 0.14$, which is shown as the solid line in the inset of Fig. 3. Clearly, a quite satisfactory result has thus been obtained. The c_L value agrees very well with those reported by Safar *et al.*⁹ and by Kwok *et al.*¹⁰ Considering the crystalline structures of Y123 ($\gamma = 7.7$),¹⁰ Y124, and Bi2212 ($\gamma = 50 \sim 170$),²⁵ we think that the anisotropy of $\gamma = 13.6$ is a reasonable value.

As mentioned at the beginning of this paper, in a clean layered superconductor, the melting line separates the vortex lattice phase at low temperatures from the vortex liquid phase above $B_m(T)$. Depending on the temperature, magnetic field, and strength of disorder, the vortex can be melted into a disentangled three-dimensional line liquid or an entangled liquid.^{1,26} It is important to note that the melting transition is not necessarily a depinning transition, which means that vortex liquid can be pinned. The pinning force can arise from the residual pinning or from viscosity of the vortices. Due to the different dimensionality and vortex configurations involved in these various liquid states, the dissipation mechanisms should also be quite different. Useful information about the liquid state can be extracted by carefully analyzing the activation behavior of vortex liquid.

From Fig. 1 it is clearly seen that above the melting temperature, i.e., in the liquid state, there are two distinct parts where ln *R* shows a linear dependence upon 1/T with different slopes. This kind of behavior ($\rho = \rho_0 e^{-U/kT}$) is typical for thermally activated flux flow (TAFF) with an activation energy $U(B,T) = U(B,0)(1-T/T_c)$.²⁷ Such two separate TAFF regions with different activation energies have previously been observed by us on oxygen-deficient Y123 thin films.²⁷



FIG. 4. Activation energies derived from the slopes in Fig. 1. Filled circles and open circles are experimental data. Solid line and dashed line are the corresponding best fits.

Using the same notations as in Ref. 27, we define the TAFF regions as region l and region h shown in Fig. 1. For each TAFF region, we extract the activation energies from the slopes of the Arrhenius plot. The obtained results are shown in Fig. 4. We find that the activation energy can be best fitted by $U_0^l = -84.3 + 497.5/\sqrt{B}$ and $U_0^h = 550$ - 423.6 ln *B*, respectively (*U* in units of K and *B* in units of Tesla). Moreover, from a low-temperature extrapolation of TAFF fit to region *h* and a high-temperature one for region *l*, we obtain the temperatures $T_{cr}(B)$ that define the crossover from region *l* to *h*.

It was argued^{1,26} that the Abrikosov lattice could melt into a disentangled liquid under certain circumstances. As discussed by Geshkenbein *et al.* and by Vinokur *et al.*, the dissipation in a disentangled liquid can be developed via plastic deformations of the vortices.²⁸ The activation energy is associated with the energy required to create a double kink in the vortex that is the free energy of two vortex segments along the *ab* plane with the length of $a_0 = (\Phi_0/B)^{1/2}$, the average distance between the vortices. Using anisotropic Ginzburg-Landau theory, the barrier for this plastic movement can be estimated as

$$U_{pl} = 2E_v a_0 \simeq \frac{\Phi_0^2}{8\pi^2 \gamma \tilde{\lambda}^2} \left(\frac{\Phi_0}{B}\right)^{1/2},$$
 (2)

where E_v is the vortex energy per unit length along the ab planes and $\tilde{\lambda}^2 = \lambda_{ab}\lambda_c$. With $\lambda_{ab}^2 = \lambda_{ab}^2(0)/(1-t)$ (where $t = T/T_c$), an activation energy $U_0 = U_{pl} \propto (1-t)/\sqrt{B}$ is predicted which is in qualitative agreement with our experimental data. To make an order of magnitude estimation of U_{pl} , we have used the above value for $\gamma(=13.6)$ and $\lambda_{ab} = 2000$ Å, thus obtaining $U_0 \approx 1500$ K at 1 T, which agrees well with the experimental data.

As the temperature increases, due to the enhanced thermal fluctuations, an entanglement-disentanglement or three- to two-dimensional decoupling transition should happen. Considering the high temperature, we think a three- to twodimensional decoupling is more likely. Daemen *et al.*²⁰ have calculated self-consistently the decoupling line for a Josephson-coupled superconducting system. When the renormalization of the Josephson coupling by thermal fluctuations and static disorder are taken into account, the decoupling field is given by

$$B_{cr}(T) = \frac{\Phi_0^3}{16\pi^3 k_B T se \lambda_{ab}^2(T) \gamma^2},$$
 (3)

for moderate anisotropy when $\xi_{ab} \ll \gamma s \ll \lambda_{ab}$, where *s* is the distance between the superconducting Cu-O planes ($s \sim 10$ Å for Y124).²⁴ Since $\lambda_{ab}^2 = \lambda_{ab}^2(0)/(1-t)$, we have $B_{cr}(T) \propto (1-T/T_c)/T$. We find that the $B_{cr}(T)$ line we obtained above can be nicely fitted by Eq. (3). The fitted result is $B_{cr} = 1406.5(1-T/T_c)/T$ as shown in Fig. 2 by the dashed line.

As discussed in Ref. 27, the dissipation in the twodimensional liquid state is controlled by the plastic motion of pancake vortices which can be visualized as the generation of dislocation pairs. The typical energy for creating such a pair is

$$U_e = \frac{\Phi_0^2 s}{16\pi^3 \lambda_{ab}^2(T)} \ln(B_0/B)$$
(4)

with $B_0 \approx \Phi_0 / \xi^2(T)$. We note that Eq. (4) gives a $(1 - t) \ln B$ dependence of U_e as we have just observed. Inserting the values for s (~10 Å) and λ_{ab} (~2000 Å), we find $U_e = 146.5(\ln B_0 - \ln B)$, in excellent agreement with our

- *Present Address: Research Institute for Engineering Materials, Tokyo Institute of Technology, Nagatsuta 4259, Yokohama 226, Japan.
- ¹G. Blatter et al., Rev. Mod. Phys. 66, 1125 (1994).
- ²E. Brandt, Rep. Prog. Phys. 58, 1465 (1995).
- ³L. Cohen and H. Jensen, Rep. Prog. Phys. **60**, 1581 (1997).
- ⁴M.P.A. Fisher, Phys. Rev. Lett. **62**, 1415 (1989); D.S. Fisher, M.P.A. Fisher, and D.A. Huse, Phys. Rev. B **43**, 130 (1991).
- ⁵D.R. Nelson and V.M. Vinokur, Phys. Rev. Lett. **68**, 2398 (1992); Phys. Rev. B **48**, 13 060 (1993); A.I. Larkin and V.M. Vinokur, Phys. Rev. Lett. **75**, 4666 (1995).
- ⁶D.R. Nelson, Phys. Rev. Lett. **60**, 1973 (1988); A. Houghton, R. Pelcovits, and A. Sudbø, Phys. Rev. B **40**, 6763 (1989); D.R. Nelson and H.S. Seung, *ibid.* **39**, 9153 (1989); M.V. Feigelfman, V.B. Geshkenbein, and A.I. Larkin, Physica C **167**, 177 (1989); R.E. Hetzel, A. Sudbø, and D.A. Huse, Phys. Rev. Lett. **69**, 518 (1992).
- ⁷R.H. Koch *et al.*, Phys. Rev. Lett. **63**, 1511 (1989); C.J. van der Beek *et al.*, *ibid.* **74**, 1214 (1995).
- ⁸R.G. Beck *et al.*, Phys. Rev. Lett. **68**, 1594 (1992).
- ⁹H. Safar et al., Phys. Rev. Lett. 69, 824 (1992); 70, 3800 (1993).
- ¹⁰W.K. Kwok *et al.*, Phys. Rev. Lett. **69**, 3370 (1992); **72**, 1088 (1992); **72**, 1092 (1994).
- ¹¹E. Zeldov *et al.*, Nature (London) **375**, 373 (1995); R. Liang, D.A. Bonn, and W.N. Hardy, Phys. Rev. Lett. **76**, 835 (1996).
- ¹²U. Welp *et al.*, Phys. Rev. Lett. **76**, 4809 (1996).

experimental data. The large prefactor ρ_0 derived from TAFF analysis for region *h* suggests that the liquid in region *h* is very viscous. Since a two-dimensional pancake vortex liquid is just an extreme case for an entangled liquid, i.e., the entanglement length is equal to *s*, the entanglement increases the viscosity of vortex liquid and results in a larger activation energy, and thus a large prefactor.

Therefore, our results suggest feasibility of the following scenario: below $B_m(T)$, the vortices form a regular Abrikosov lattice. As the temperature increases, the lattice melts into a disentangled liquid and loses its transverse correlation while keeping the longitudinal one. The dissipation is governed by the generation of double kinks. Upon further warming, a three- to two-dimensional decoupling transition sets in and the vortices lose their longitudinal coherence. The two-dimensional vortices form very viscous liquid and the activation energy increases following a $(1-t)\ln B$ relationship.

In conclusion, we have shown that vortex lattice melting can also been observed in Y124 single crystals. The melting line can be well described by using anisotropic Ginsburg-Landau theory with a Lindermann constant $c_L=0.14$. Two different liquid phases were observed that we think correspond to the disentangled liquid and the two-dimensional pancake vortex liquid, respectively.

We thank E. Rossel and P. Wagner for their help during the measurements. This research has been supported by the Belgian IUAP and Flemish GOA, FWO, and BIL 97/35 programs.

- ¹³R. Cubitt et al., Nature (London) 365, 407 (1993).
- ¹⁴S.L. Lee et al., Phys. Rev. Lett. 75, 922 (1995).
- ¹⁵A. Schilling *et al.*, Nature (London) **382**, 791 (1996).
- ¹⁶E.A. Jagla and C.A. Balseiro, Phys. Rev. Lett. **77**, 1588 (1996);
 D. Domínguez, N. Grønbech-Jensen, and A.R. Bishop, *ibid*. **78**, 2644 (1997).
- ¹⁷W. Jiang et al., Phys. Rev. Lett. 74, 1438 (1995).
- ¹⁸D.T. Fuchs *et al.*, Phys. Rev. B **54**, 796 (1996).
- ¹⁹J.A. Fendrich et al., Phys. Rev. Lett. 74, 1210 (1995).
- ²⁰L.I. Glazman and A.E. Koshelev, Phys. Rev. B 43, 2835 (1991);
 L. Daemen *et al.*, Phys. Rev. Lett. 70, 1167 (1993).
- ²¹D. Lopez *et al.*, Phys. Rev. Lett. **76**, 4034 (1996); D.T. Fuchs *et al.*, Phys. Rev. B **55**, 6156 (1997).
- ²²Y.M. Wan, S.E. Hebboul, and J.C. Garland, Phys. Rev. Lett. **72**, 3867 (1994); C.D. Keener *et al.*, *ibid.* **78**, 1118 (1997).
- ²³J. Karpinski et al., Nature (London) 336, 660 (1988).
- ²⁴W.C. Lee and D.M. Ginsberg, Phys. Rev. B 45, 7402 (1992); M. Xu *et al.*, *ibid.* 53, 15 313 (1996).
- ²⁵R. Doyle *et al.*, Phys. Rev. Lett. **75**, 4520 (1995).
- ²⁶D.R. Nelson, in *Phenomenology and Applications of High Temperature Superconductors*, edited by K. Bedell *et al.* (Addison-Wesley, New York, 1992).
- ²⁷X.G. Qiu et al., Phys. Rev. B 52, 559 (1995).
- ²⁸ V. Geshkenbein *et al.*, Physica C **162-164**, 239 (1989); V.M. Vinokur *et al.*, Phys. Rev. Lett. **65**, 259 (1990).