# **Nearly field-independent in-plane vortex solid-to-liquid transition in the** *c***-axis resistivity of oxygen deficient single crystals of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-** $\delta$ **</sub>**

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The *c*-axis resistivity has been measured in oxygen deficient single crystals of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> (anisotropy  $\gamma \approx 24-29$ ) with the magnetic field *B* accurately aligned parallel to the *ab* plane. An almost field-independent vortex solid-to-liquid transition is observed at high fields in contrast to the usual field-dependent one, which is recovered for a small misalignment between *B* and *ab*. This observation challenges previous interpretations of the same effect in the *ab* resistivity as coming from a smectic vortex state or a decoupling of the superconducting planes.

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## **I. INTRODUCTION**

The mixed state of the high-temperature superconductors (HTSC) allows for the experimental observation of a large number of fascinating phenomena.<sup>1,2</sup> Most importantly, there is a separation of the mixed state into a low-temperature superconducting vortex solid with a nonzero critical current density and a high-temperature dissipative vortex liquid. In clean single crystals it is possible to observe a first-order transition between the two states. $3,4$  However, when disorder is introduced this transition turns into a second-order one and the low-temperature phase is in a glassy state, whose properties depend on the kind of disorder.<sup>5–8</sup> The extent of the dissipative vortex liquid in the magnetic phase diagram depends strongly on the anisotropy of the superconductor, and is larger for materials with higher anisotropy.

The layered structure of the HTSC gives rise to a strong modulation of the order parameter in a direction perpendicular to the superconducting  $CuO<sub>2</sub>$  planes. For well aligned in-plane magnetic fields this results in a *c*-axis periodic pinning potential, since the free energy of the superconductor is minimized when the vortices are located in the interlayer spacings between the  $CuO<sub>2</sub>$  layers.<sup>9</sup> This intrinsic pinning has been shown to be of significance for the vortex dynamics with  $I||ab$ , where the Lorentz force acting on the vortices tries to push them perpendicularly to the planes.<sup>10,11</sup> For a *c*-axis current, however, the Lorentz force is directed parallel to the *ab* plane and consequently the intrinsic pinning mechanism should be of less importance. In strongly anisotropic  $Bi_2Sr_2CaCu_2O_{8-\delta}$  single crystals, an enhanced dissipation has actually been observed for this configuration as compared to the case with a small misalignment between *B* and *ab*. 12,13 In layered superconductors with *B* parallel and nearly parallel with the layers it has been predicted theoretically that the vortex liquid can condense into a smectic state  $(with a small nonzero resistivity)$  as an intermediate phase between a solid and an uncorrelated liquid.<sup>14</sup> By varying the field strength various incommensurate and commensurate smectic and solid phases were found, mapping out a complex

magnetic phase diagram. Other studies on highly anisotropic HTSC have suggested that strong enough in-plane fields should penetrate as if the superconducting layers were completely decoupled.<sup>15</sup>

Oxygen deficient single crystals of  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-\delta</sub>$ (YBCO) provide a powerful tool for studies of the vortex physics, since the electrical anisotropy  $\gamma=(m_c / m_{ab})^{1/2}$  can be varied systematically over a considerable range by changing the oxygen content.<sup>16</sup> When measuring the in-plane resistivity of such crystals we observed a nearly fieldindependent vortex solid-to-liquid transition at high magnetic fields accurately aligned parallel to the *ab* plane.  $17-19$  This effect was attributed to a decoupling of the superconducting planes, which then suggested a twodimensional  $(2D)$  system.<sup>19</sup> Recently, the nearly fieldindependent transition was confirmed by Gordeev *et al.,* who also showed an oscillatory behavior of the solid-to-liquid line at high fields and took their findings as evidence for a smectic vortex phase. $20$  Here, we present measurements of the *c*-axis resistivity of oxygen deficient YBCO crystals. Remarkably, an almost field-independent finite temperature vortex solid-to-liquid transition was found also for this configuration. This observation is in contrast to what one would expect from models based on decoupling of superconducting planes or smectic vortex phases.

### **II. EXPERIMENT**

Single crystals of YBCO were grown by a self-flux method in yttria-stabilized zirconia crucibles as described elsewhere. $21$  Twinned crystals of varying oxygen content were obtained by annealing for about one week at different temperatures. Electrical contacts were prepared by applying silver paint, followed by heat treatment under the same conditions as for annealing, giving contact resistances below 1.5  $\Omega$ . Three different contact configurations shown in Fig. 1 were used in order to test the rigidity of the results with respect to possible *ab* components and inhomogeneities in the *c*-axis current. Typical dimensions of the crystals were  $(l \times w \times t)$  0.4 $\times$ 0.4 $\times$ 0.05 mm<sup>3</sup>. Sample 3 had a larger



FIG. 1. The three contact configurations used in this study in order to test the rigidity of the results with respect to possible *ab* components and inhomogeneities in the *c*-axis current. Sample 3 has the most convenient contact configuration for *c*-axis measurements and should give a minimal contribution from the *ab* plane.

thickness of 0.16 mm. The anisotropy of samples 1 and 2 was estimated from fitting the equation  $B_g(T) = B_0[(1$  $-T/T_c$ )/(*T*/*T<sub>c</sub>*)]<sup> $\alpha$ </sup>, with  $B_0 \propto 1/\gamma^2$  (and  $\alpha \approx 1$  for YBCO), to the vortex solid-to-liquid transition line for the  $B||c$  axis as previously described.<sup>22</sup> This method was originally developed for in-plane resistive measurements, but gives also good estimates of the anisotropy in *c*-axis measurements, as observed from comparisons between in-plane and *c*-axis samples prepared in the same way. Since sample 2 and 3 were annealed under similar conditions it is likely that they have approximately the same oxygen content and anisotropy. Properties of the samples are summarized in Table I.

The samples were mounted in a rotatable sample holder with an angular resolution of 0.01°, allowing for accurate alignment with respect to the magnetic field. Resistive measurements in the Ohmic regime were made with a four-probe dc technique using a current of 0.1 mA for samples 1 and 2, and 0.01 mA for sample 3. A picovoltmeter was employed as a preamplifier and the effect of thermopower was reduced by averaging over voltages measured for positive and negative currents. This method routinely gives a voltage resolution of better than 300 pV. The samples were cooled in field and the data were recorded during increasing temperatures.

#### **III. RESULTS AND DISCUSSION**

Figure 2 shows the angular dependence of the *c*-axis resistance close to the  $B||ab$  plane for sample 2. At high tem-

TABLE I. Oxygen deficient single crystals of YBCO used in this study.  $T_c$  was taken as the temperature for onset of resistivity (our resolution limit) and  $\Delta T_c$  are transition widths (10–90 %).  $\gamma$  of samples 1 and 2 was estimated from the vortex glass line for the  $B\|c$  axis as described in Ref. 22. The errors are below 10%. Since samples 2 and 3 were annealed under the same conditions it is likely that they have approximately the same  $\gamma$ .

Sample	$T_c$ (K)	$\Delta T_c$ (K)	Annealing	$\gamma$
	70.6	2.6	air, $550^{\circ}$ C	24
	59.2	2.4	air, $600^{\circ}$ C	29
3	60.6	1.8	air, $600^{\circ}$ C	$\approx$ 2.9



FIG. 2. Angular dependence of the *c*-axis resistance for sample 2 at  $B=10$  T close to parallel with the *ab* plane. The peak observed at high temperatures turns into a dip when approaching the vortex solid-to-liquid transition.

peratures there is a sharp peak for  $B||ab$ . Similar peaks have previously been observed for  $Bi_2Sr_2CaCu_2O_{8+\delta}$  (Bi-2212) single crystals, $12,13$  and interpreted as resulting from increased mobility of Josephson vortices along the layers as the additional pinning from pinned pancake vortices in the planes is lost for  $B||ab$ . However, at lower temperatures we observed that the peak was gradually replaced by a dip, similar to the one usually seen for  $B||ab$  with  $I||ab$ . The behavior shown in Fig. 2 is also representative for sample 3, whereas sample 1 (the less anisotropic one) only showed dips.

To determine the vortex solid-to-liquid transition temperature, the data were in most cases analyzed within the framework of theories for glassy vortex states,  $5.7$  which for a 3D superconductor predicts that the Ohmic resistivity should go to zero at the glass temperature  $T_g$  as  $\rho \propto (T - T_g)^e$ , where *e* depends on the static and dynamic critical exponents. Consequently,  $T<sub>g</sub>(B)$  can be extracted by a linear extrapolation when applying the relation

$$
(d\ln \rho/dT)^{-1} \propto (T - T_g) \tag{1}
$$

to the resistive tail. This method for finding  $T_g$  has previously been shown by in-plane transport measurements to be experimentally consistent with a glass scaling of the currentvoltage  $(I-V)$  characteristics.<sup>23,24</sup>

Figure 3 shows the resistive transition curves for sample 3 in magnetic fields aligned parallel to the *ab* plane. For fields below 6 T the usual broadening is observed. Here, good linear agreement with Eq. (1) was found when extracting  $T_g$ . More interesting, however, is the high field region where a kinklike feature develops, below which the resistance sharply drops to zero. In this regime the resistance does not follow the behavior predicted by Eq.  $(1)$ , and instead the solid-to-liquid transition had to be evaluated as the temperature for the onset of resistance (our resolution limit). Due to the sharp resistance drop, the errors can be estimated to be rather small (less than  $0.2$  K). The general trend is therefore that the transition temperature is nearly field independent at high magnetic fields. On the scale of Fig. 3 it is also seen that the 12 T curve goes to zero at a slightly higher temperature than the 10 T curve. This indicates that we have a similar



FIG. 3. Arrhenius plot of the resistive transition curves for sample 3. In the upper part of the graph (above 0.1  $\Omega$ ), the resistance grows monotonically with increasing field  $B||ab$  plane of (from left to right)  $0, 1, 2, 4, 6, 8, 10,$  and  $12$  T. In the lower part of the graph the 12 T curve falls to zero at a higher temperature than the 6, 8, and 10 T curves. Inset: The same data plotted as resistance versus temperature.

oscillatory behavior of the vortex solid-to-liquid transition in the *c*-axis resistivity as observed by Gordeev *et al.* in the *ab* resistivity.20 The above description of the resistive behavior is qualitatively the same for sample 2. For sample 1, the lower parts of the transition curves for the highest fields go more rapidly towards zero than for the lower fields as observed from the stronger downward curvature in an Arrhenius plot. For this sample good agreement with Eq.  $(1)$  is found for all studied fields, and for  $B \ge 8$  T the extrapolation lines practically merge on a single temperature, apparently leading to a saturation in the field dependence of the temperature where the Ohmic resistivity goes to zero. The same behavior has recently been reported by us from measurements of the in-plane resistance of similar crystals (see Figs. 3 and 4 in Ref. 19).



FIG. 4. Arrhenius plot of the resistive transition curves for sample 3 at  $B=12$  T accurately aligned parallel to the *ab* plane and also with a small misalignment of 1°. The zero-field transition has been included for comparison.



FIG. 5. Vortex solid-to-liquid transitions for the  $B||ab$  plane of oxygen deficient single crystals of YBCO. Open symbols: This work with  $I||c$  axis. Filled symbols: Results from in-plane resistive measurements  $(B \perp I)$  on another set of crystals (Ref. 19). For both current directions a nearly field-independent solid-to-liquid transition is observed above an apparently anisotropy-dependent field.

The described phenomena are very sensitive to the accuracy of the field alignment. As shown for sample 3 in Fig. 4 the usually observed behavior is recovered when a small misalignment between *B* and *ab* is introduced. This shows that the observed effect is intimately related to the layered structure of the material.

The magnetic field dependence of the solid-to-liquid transitions for *B*|| $ab$  plane and *I*|| $c$  axis is shown as open symbols in Fig. 5. For all samples the transition is almost field independent above an apparently anisotropy-dependent magnetic field  $B^*$ . For sample 1,  $B^* \approx 8$  T, and for samples 2 and 3,  $B^* \approx 5-6$  T. From an anisotropic Ginzburg-Landau theory using the anisotropies in Table I, one finds that *B*\* corresponds to a *c*-axis distance between vortex planes of 1.5 times the  $c$ -axis lattice parameter of the material.<sup>19</sup> This leads to a frustrated vortex state at  $B^*$ , which at higher fields approaches a commensurate one, when the vortex plane distance is equal to the *c*-axis lattice parameter. Since the vertical transition is observed for all samples, possible in-plane current components, that might be expected from some of the contact configurations shown in Fig. 1, do not seem to have any significant impact on the final result.

Saturation in the field dependence or even insensitivity to all studied magnetic fields have previously been reported for superconducting transitions in YBCO/PrBCO multilayers<sup>25,26</sup> and also for  $J_c$  in Bi-2212 thin films<sup>27</sup> and decoupled Bibased multilayers.28 In these experiments the in-plane transport properties were probed and the field independence was interpreted as being a result of the 2D character of the studied materials. Similarly from measurements of the in-plane resistivity we observed a field-independent vortex solid-toliquid transition for the  $B||ab$  plane in oxygen deficient single crystals of YBCO.<sup>17–19</sup> The transitions for this configuration are included as filled symbols in Fig. 5. It is interesting to note the similarities in the behavior for *c*-axis and *ab*-plane samples. This is most evident for the two samples with approximately the same  $T_c$ , for which the data almost collapse onto each other. This suggests that the same physical mechanism is responsible for the vertical transition for both in- and out-of-plane measurements.

In the case of in-plane measurements this transition has been proposed to either be connected to a field induced decoupling of the planes leading to a two-dimensional vortex system $1<sup>9</sup>$  or to a smectic vortex state where thermal fluctuations perpendicular to the planes are suppressed while thermal fluctuations along the layers will destroy any order along this direction.14,20 However, in both these cases one would expect the *c*-axis resistivity to stay nonzero for all temperatures in contrast to our observations. For untwinned and weakly twinned clean YBCO single crystals, an extra tail has recently been observed and interpreted as the result of a twostep transition from the vortex solid to the liquid with the smectic phase as an intermediate step. $^{29}$  In our oxygen deficient crystals, possible pinning centers are coming from twinning and oxygen disorder where the latter probably is

- <sup>1</sup>G. Blatter, M. V. Feigel'man, V. B. Geshkenbein, A. I. Larkin, and V. M. Vinokur, Rev. Mod. Phys. 66, 1125 (1994).
- 2L. F. Cohen and H. J. Jeldtoft Jensen, Rep. Prog. Phys. **60**, 1581  $(1997).$
- 3A. Schilling, R. A. Fisher, N. E. Phillips, U. Welp, D. Dasgupta, W. K. Kwok, and G. W. Crabtree, Nature (London) 382, 791  $(1996).$
- 4U. Welp, J. A. Fendrich, W. K. Kwok, G. W. Crabtree, and B. W. Veal, Phys. Rev. Lett. **76**, 4809 (1996).
- 5D. S. Fisher, M. P. A. Fisher, and D. A. Huse, Phys. Rev. B **43**, 130 (1991).
- <sup>6</sup>R. H. Koch, V. Foglietti, W. J. Gallagher, G. Koren, A. Gupta, and M. P. A. Fisher, Phys. Rev. Lett. **63**, 1511 (1989).
- ${}^{7}$ D. R. Nelson and V. M. Vinokur, Phys. Rev. B **48**, 13 060 (1993).
- <sup>8</sup>T. Giamarchi and P. Le Doussal, Phys. Rev. B **52**, 1242 (1995); **55**, 6577 (1997).
- 9M. Tachiki and S. Takahashi, Solid State Commun. **70**, 291  $(1989).$
- 10W. K. Kwok, U. Welp, V. M. Vinokur, S. Fleshler, J. Downey, and G. W. Crabtree, Phys. Rev. Lett. **67**, 390 (1991).
- <sup>11</sup> W. K. Kwok, J. Fendrich, U. Welp, S. Fleshler, J. Downey, and G. W. Crabtree, Phys. Rev. Lett. **72**, 1088 (1994).
- <sup>12</sup>K. Kadowaki and T. Mochiku, Physica B **194-196**, 2239 (1994).
- <sup>13</sup>L. N. Bulaevskii, M. Maley, H. Safar, and D. Domínguez, Phys. Rev. B 53, 6634 (1996).
- $14$ L. Balents and D. R. Nelson, Phys. Rev. Lett. **73**, 2618 (1994); Phys. Rev. B 52, 12 951 (1995).
- 15P. H. Kes, J. Aarts, V. M. Vinokur, and C. J. van der Beek, Phys. Rev. Lett. **64**, 1063 (1990).
- 16T. R. Chen, W. R. Datars, B. W. Veal, A. P. Paulikas, P. Kostic,
- Chun Gu, and Y. Jiang, Physica C 229, 273 (1994). <sup>17</sup>B. Lundqvist, Ö. Rapp, and M. Andersson, Physica B 284-288,
- 705 (2000).<br><sup>18</sup>B. Lundqvist, Ö. Rapp, and M. Andersson, Physica C **341-348**,
- 1335 (2000).<br><sup>19</sup>B. Lundqvist, Ö. Rapp, and M. Andersson, Phys. Rev. B **62**, 3542
- $(2000).$
- 20S. N. Gordeev, A. A. Zhukov, P. A. J. de Groot, A. G. M. Jansen,
- R. Gagnon, and L. Taillefer, Phys. Rev. Lett. **85**, 4594 (2000). <sup>21</sup>Yu. Eltsev, W. Holm, and O<sup>¨</sup>. Rapp, Phys. Rev. B **49**, 12 333
- (1994).<br><sup>22</sup>B. Lundqvist, A. Rydh, Yu. Eltsev, Ö. Rapp, and M. Andersson, Phys. Rev. B 57, R14 064 (1998).
- <sup>23</sup> Lifang Hou, J. Deak, P. Metcalf, M. McElfresh, and G. Preosti,
- Phys. Rev. B **55**, 11 806 (1997). <sup>24</sup>B. Lundqvist, J. Larsson, A. Herting, Ö. Rapp, M. Andersson, Z. G. Ivanov, and L.-G. Johansson, Phys. Rev. B 58, 6580 (1998).
- $25$  J. M. Triscone,  $\phi$ . Fischer, O. Brunner, L. Antognazza, A. D. Kent, and M. G. Karkut, Phys. Rev. Lett. **64**, 804 (1990).
- <sup>26</sup>O. Brunner, M. G. Karkut, L. Antognazza, L. Miéville, P. van der Linden, J. A. A. J. Perenboom, J. M. Triscone, and  $\emptyset$ . Fischer, Physica C 185-189, 2079 (1991).
- 27P. Schmitt, P. Kummeth, L. Schultz, and G. Saemann-Ischenko, Phys. Rev. Lett. **67**, 267 (1991).
- 28S. Labdi, S. F. Kim, Z. Z. Li, S. Megtert, H. Raffy, O. Laborde, and P. Monceau, Phys. Rev. Lett. **79**, 1381 (1997).
- <sup>29</sup>S. A. Grigera, E. Morré, E. Osquiguil, G. Nieva, and F. de la Cruz, Phys. Rev. B 59, 11 201 (1999).

the most important here since twin boundaries make a 45° angle with the flux lines. A possible interpretation of our results is thus that a smectic vortex state is destroyed by pinning. However, these issues have to be further clarified.

In summary, the vortex solid-to-liquid transition for accurate field alignment  $B||ab$  plane in oxygen deficient single crystals of YBCO has been studied from the vortex liquid side by measurements of the *c*-axis resistivity. A nearly fieldindependent transition similar to the one previously reported by us from in-plane measurements was observed at high magnetic fields. A satisfactory physical explanation for the vertical transition remains to be found.

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