Oscillatory Melting Temperature of the Vortex Smectic Phase in Layered Superconductors

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We report on transport measurements of $YBa_2Cu_3O_{7-\delta}$ single crystals with different oxygen contents in the geometry B, $J \| ab$ ($J \perp B$). Our data show that the vortices become confined between the Cu-O planes below a well-defined temperature at which the effective size 2ξ of the vortex core is approximately equal to the period of the Cu-O layers. This confinement strongly increases the vortex liquid freezing temperature. A new melting line is found separating a vortex liquid and a smectic phase, which shows an oscillatory field dependence reflecting differences between commensurate and incommensurate smectic states.

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In recent years a consensus has developed concerning the nature of the vortex phase diagram for magnetic fields applied perpendicular to the Cu-O layers ($B \parallel c$). The high-temperature part of the phase diagram is occupied by a mobile disordered state called the vortex liquid. With decreasing temperature this liquid freezes into a disordered glass [1] or an ordered Bragg glass [2] state, depending on the strength of random pinning. The situation is less clear for fields applied parallel to the Cu-O layers (the crystalline *ab* plane). Experiments by Kwok *et al.* [3,4], revealed that, in this geometry, pinning by the layered crystalline structure of the cuprate has a strong influence on the vortex properties. From theoretical studies [5,6] it has been predicted that when this pinning becomes strong enough the vortices become confined between the Cu-O planes resulting in a drastic change of the phase diagram. Several new phases [7,8] and different scenarios for the vortex liquid freezing [9] have been proposed for this case.

According to the scenario suggested by Balents and Nelson [8], upon lowering the temperature the confined vortex liquid first freezes into an intermediate "smectic" phase followed by a second freezing transition at lower temperatures into the vortex solid phase. The smectic/liquid melting line is predicted to have an oscillatory dependence on the applied field with maxima corresponding to the fields at which the vortex system is commensurate to the period of the Cu-O planes. Previous transport studies [3,4,10] which were performed on optimally doped $YBa_2Cu_3O_{7-\delta}$ (YBCO) single crystals in the field range 0–8 T, failed to observe such behavior. Instead the melting line showed a typical power-law dependence for 3D melting, $B \propto (T_c - T_m)^n$, where T_c and T_m are critical and melting temperatures, respectively, and $n \sim 1.28$ [4]. This observation suggests that for $B \leq 8$ T the intrinsic pinning within the vortex liquid phase in optimally doped YBCO single crystals is not strong enough to confine vortices between the Cu-O planes.

In this Letter, we show that the oscillatory melting line separating the vortex liquid from the vortex smectic state can be observed in YBCO single crystals with reduced oxygen content. We demonstrate that the confinement of the vortices between the Cu-O planes occurs below a characteristic (confinement) temperature $T_{\rm cf}$ at which the effective diameter $2\xi_c$ of the vortex core becomes equal to the period of the Cu-O planes. This confinement triggers the phase transition from the vortex liquid to the smectic state. Our observations of an abrupt appearance of commensurability oscillations below $T_{\rm cf}$ provides strong evidence for the transition from a disordered (liquid) to an ordered (smectic) phase at this temperature.

Experiments were performed on twinned YBCO single crystals grown by a conventional self-flux method [11] in yttria-stabilized zirconia crucibles, starting with powders of Y_2O_3 (99.9999%), $BaCO_3$ (99.999%), and CuO (99.9999%). To investigate freezing of the vortex liquid in the geometry $B \parallel ab$ and influence of intrinsic parameters on this process, we have prepared four samples with distinctly different critical temperature T_c, anisotropy Γ, and coherence length ξ_c (see Table I). This was achieved by annealing the crystals in flowing oxygen at different temperatures. We have measured the resistivity using a standard four-probe technique with a current applied parallel to the *ab* plane. The experimental arrangement is shown in inset (a) to Fig. 1. The crystal was rotated, with a resolution of 0.001°, around an axis along the current direction.

Inset (b) to Fig. 1 shows the dependence of the resistivity on the angle θ between the field and the Cu-O planes, $\rho(\theta)$, for sample T750 ($T_c = 60.1$ K). For temperatures below $T_{\rm cf} = 57.3$ K we observed a spectacular drop of resistivity, more than an order of magnitude, within a narrow angle range of about 0.5° centered at $B \parallel ab$. This strong reduction in resistivity is a signature of the vortex

TABLE I. Annealing temperature T_{an}, oxygen concentration 7-δ, critical temperature T_c, anisotropy Γ, coherence length at $T = 0$ K, $\xi_c(0)$; onset of smectic transition T_{cf}, coherence length at T_{cf}, $\xi_c(T_{cf})$; and ratio $s/\xi_c(T_{cf})$ for the samples studied.

Sample	T750	T700	T650	T500
T_{an} (°C)	750	700	650	500
7-δ[a]	6.55	6.63	6.72	6.93
T_c (K)	60.1	63.1	76.6	93.2
Γ[b]	15.5	13.5	12.9	7.2
$\xi_c(0)$, (nm)[c]	0.14	0.17	0.18	0.20
T_{cf} (K)	57.3	58.0	68.1	
$\xi_c(T_{cf})$ (nm)	0.66	0.62	0.55	
$s/\xi_c(T_{cf})$	1.8	1.9	2.1	

[a]Calculated from annealing temperatures using Eq. (6) in Ref. [18].
[b]Obtained from the period of commensurability oscillations (magnetization studies) [19,12].
[c]Obtained from measurements of the reversible magnetization as described in Ref. [20].

confinement between the Cu-O planes. The main panel of Fig. 1 shows the temperature dependence of the resistivity for the same sample for different field magnitudes in the geometry $B \parallel ab$ ($\theta = 0°$). All the curves are obtained with a low measurement current (0.1 mA) for which

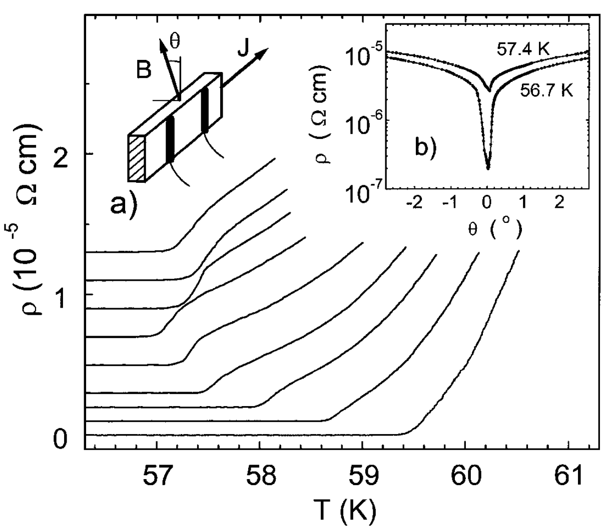

FIG. 1. Temperature dependence of the resistivity ($I = 0.1$ mA, Ohmic regime) for YBCO single crystal (sample T750) for fields 1, 2, 3, 4, 6, 8, 10, 12, 14 T. The field is applied accurately along the crystalline ab-plane. The curves are shifted along the y axis for clarity. Inset (a) shows contact geometry and the orientation of the magnetic field and applied current. Inset (b) shows the angular dependence of the resistivity in the vicinity of the ab-plane direction for temperatures above and below the lock-in transition ($B = 12$ T). The measuring current is 37 mA.

the voltage response is Ohmic. For fields $B \leq 4$ T the $\rho(T)$ curves are typical of high-temperature superconductors in the three-dimensional regime. For increasing applied fields the transition in $\rho(T)$ broadens, corresponding to the increase of the vortex liquid region. Freezing of the vortex liquid is accompanied by a decrease to zero of the Ohmic resistivity. This transition is continuous, in agreement with earlier observations on optimally doped YBCO [4]. Our measurements revealed dramatic changes of the $\rho(T)$ curves for all fields higher than 4 T. As shown in Fig. 1 for $B > 4$ T the $\rho(T)$ curves drop abruptly at approximately the same temperature, $T_{cf} \approx 57.3$ K.

We have found that at T_{cf} the character of the vortex dynamics changes as well. For $T < T_{cf}$ the voltage response becomes strongly non-Ohmic and as shown in Fig. 2, the resistivity has an oscillatory behavior as a function of the field. An important observation is that the positions of the minima in $\rho(B)$ are temperature independent. In previous investigations similar oscillations were observed in the magnetic hysteresis of YBCO [12,13] when the vortices are accurately aligned with the Cu-O layers. These oscillations relate to the fact that the energy of the vortex system has minima when the period of the layered structure of YBCO, $s = 1.17$ nm, matches the intervortex spacing in the direction normal to the layers $l = (\sqrt{3}\Phi_0/2\Gamma)^{1/2}B^{-1/2}$ (Φ_0 is the flux quantum). The oscillations shown in Fig. 2 closely correspond to this scenario: they are periodic in $B^{-1/2}$ and are present only when the field is accurately aligned with the ab plane ($\theta < 0.25°$). The positions of the resistivity minima correspond to the fields at which the ratio l/s has integer values (shown by arrows in Fig. 2). Hence our observation of commensurability oscillations is strong evidence that below T_{cf} the vortex system is locked between the Cu-O layers and is ordered in the direction normal to the layers.

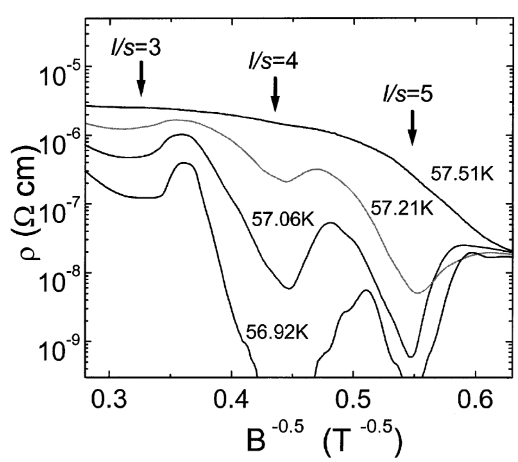

FIG. 2. Resistivity in the non-Ohmic regime ($I = 10$ mA) for various temperatures as a function of $B^{-0.5}$. The arrows indicate the fields for which the intervortex spacing l in the direction normal to the Cu-O layers is commensurate with the layer period s.

We have investigated the temperature at which the vortex liquid freezes for fields applied at different angles. It was shown both theoretically [6] and experimentally [14] that in anisotropic superconductors (i.e., neglecting the layered nature of the cuprate superconductors) the melting transition occurs at the same temperature for all angles θ if the effective field $B_{\rm eff} = B(\cos^2\theta + \Gamma^2 \sin^2\theta)^{1/2}$ is kept constant. Figure 3 displays dependences $B_{\rm eff}(T_{\rm m})$ for sample T750 for different angles. The melting temperature $T_{\rm m}$ was determined from $\rho(T)$ curves by using a constant resistivity criterion [15], $\rho(T_{\rm m}) = 0.03\,\rho(62{\rm K})$. As shown in Fig. 3 the normalized melting line for $\theta = 2.5°$ coincides with the corresponding line for $\theta = 90°$ and both follow the same power-law dependence $B_{\rm eff} \propto (T_{\rm c} - T)^n$ with $n \approx 1.4$. We observed such scaling behavior for all angles greater than 0.5°, which means that for $\theta > 0.5°$ YBCO behaves as an anisotropic 3-dimensional superconductor [6]. For smaller angles this behavior remains unchanged for $T > T_{\rm cf}$. However, for $T < T_{\rm cf}$, the melting line turns up abruptly (see Fig. 3). In the case of $\theta = 0°$ the steep part of the melting line is nearly vertical. Both the $\theta = 0°$ and the $\theta = 0.2°$ lines have an *oscillatory* field dependence with maxima, shown by arrows, near $B = 5.3$ T and $B = 9.5$ T. These fields correspond to the commensurate states $l/s = 4$ and $l/s = 3$ as determined from the data presented in Fig. 2. Such an oscillatory behavior of the melting line is a direct consequence of the difference between the confinement strengths in commensurate and incommensurate smectic vortex states. The commensurate states experience a stronger confinement potential and hence melt at higher temperatures than the incommensurate states. This observation provides strong support for the phase diagram predicted in the theoretical work by Balents and Nelson [8].

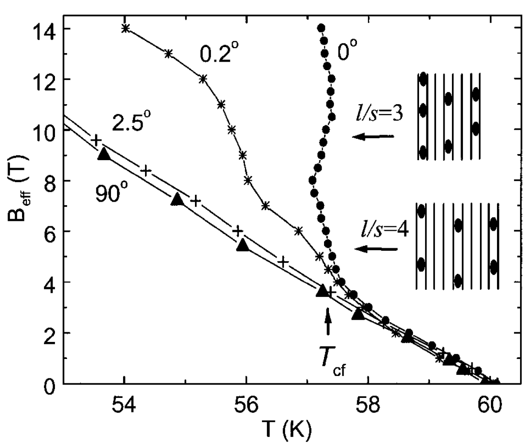

FIG. 3. Melting lines for sample T750 for different angles between the field and the crystalline *ab*-plane. Temperature $T_{\rm cf}$ indicates the onset of the transition from the vortex liquid to the smectic state for $\theta = 0°$. The upper and lower insets show schematically the vortex core positions between the superconducting layers for commensurate states corresponding to $l/s = 3$ and $l/s = 4$. The arrows indicate the fields for these states.

The abrupt appearance of commensurability oscillations for $T < T_{\rm cf}$ provides strong evidence for a change in the symmetry of the vortex system at this temperature, which is a signature of a phase transition. We conclude that the steep part of the melting line separates two different vortex phases: a disordered vortex liquid and a phase which is ordered in the direction of c axis. Simple arguments exclude the existence of in-plane order in the low-temperature phase. From Fig. 3 one can see that the unconfined vortex liquid ($\theta > 0.5°$) freezes into a solid for temperatures well below $T_{\rm cf}$. This means that in the 3-dimensional case thermal fluctuations at $T_{\rm cf}$ are so strong that the vortex lattice should melt. The confinement of the vortices between Cu-O layers suppresses thermal fluctuations *perpendicular to the layers* [8], encouraging ordering of the vortices in this direction. However, confinement does not reduce thermal fluctuations *along the layers.* According to the theoretical work by Balents and Nelson [8] these longitudinal fluctuations will destroy any order along the *ab* plane. Such phases, which have crystalline order in one direction but retain liquid properties in other directions, are well known in liquid crystals and are usually referred to as smectic phases [16]. Therefore the vertical melting line for $\theta = 0°$ manifests both the onset of the vortex confinement and freezing of the vortex liquid into a smectic phase. As shown in Fig. 1 this transition is accompanied by an abrupt drop of the Ohmic resistivity with a characteristic kink on $\rho(T)$ dependence. We have found that this kink also manifests a transition from the Ohmic to a strongly non-Ohmic regime. Such behavior of the resistivity was previously observed in optimally doped YBCO single crystals in the geometry $B \parallel c$ and was proved to be a result of a first order phase transition within the vortex system [17]. The apparent similarity in transport results suggests that freezing of the vortex liquid into smectic is also a first order transition. However, the final confirmation of this point requires measurements of thermodynamic parameters.

Figure 4 displays the melting lines (for $\theta = 0°$) for YBCO samples with different intrinsic parameters. The abrupt upturn which indicates the liquid-to-smectic transition shifts to lower relative temperatures, $T/T_{\rm c}$, and hence to higher fields with increasing oxygen content of the samples. Most measurements were performed using a 14 T superconducting magnet system and $T_{\rm cf}$ could not be determined for crystals T650 and T500. Further measurements on these crystals were performed at the Grenoble High Magnetic Field Laboratory for fields up to 28 T. These measurements allowed us to determine $T_{\rm cf}$ for crystal T650. However, for the optimally doped crystal T500 even this field range is not sufficient. The observed behavior of the melting line can be understood within the theoretical framework developed for vortex liquid-smectic transitions in layered superconductors [8]. This theory proposes that the confinement potential is determined by the ratio of the temperature dependent coherence

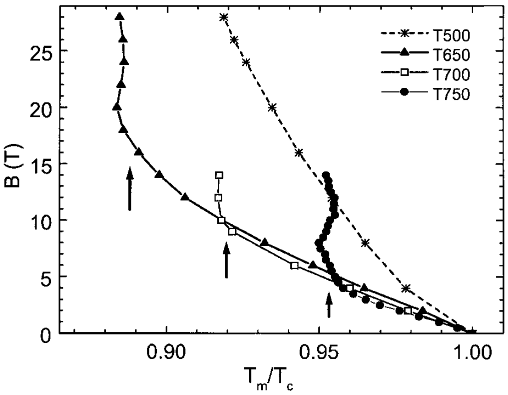

FIG. 4. Melting lines for YBCO single crystals with different intrinsic parameters (see Table I). The arrows indicate T_{cf} for the different crystals.

length, $\xi_c(T) = \xi_c(0)/(1 - T/T_c)^{1/2}$, to the distance between layers. At high temperatures this ratio is large, resulting in a weak confinement. With decreasing temperature the confinement potential increases rapidly and when this potential exceeds the thermal energy, vortices become locked between the superconducting layers. This confinement triggers the phase transition from the liquid to the smectic state. This approach suggests that even a small increase of $\xi_c(0)$ should significantly shift the smectic transition to lower relative temperatures. This is exactly what is found in our experiments. Moreover, as shown in Table I, the values of the coherence length at the point of the transition are close for different samples. It is remarkable that the ratio $s/\xi_c(T_{cf})$ is nearly 2, which means that at the transition the effective diameter, $2\xi_c$, of the vortex core is approximately equal to the period of the Cu-O layers. The lock-in transition occurs when the vortex cores fit between the layers. Based on these findings, we estimate T_{cf} for the optimally doped crystal T500 as $T_{cf} \approx 80$ K. Hence, fields of about 50 T are required to observe the liquid-to-smectic transition in optimally doped YBCO. Such high-static magnetic fields are as yet not available to experimentalists. This explains why all previous attempts to observe the liquid-smectic transition, which were performed on optimally $YBa_2Cu_3O_{7-\delta}$ crystals, have failed.

In summary, we have reported on transport measurements of YBCO single crystals with different oxygen contents in the geometry $B \parallel ab$. We find that the resistance drops abruptly at a certain temperature T_{cf} when the vortices become confined between the Cu-O planes. Our observations of commensurability oscillations below T_{cf} give strong evidence for the transition from a disordered (liquid) to an ordered (smectic) phase at this temperature. We show that the corresponding melting line has an oscillatory dependence on the applied field, in agreement with theoretical predictions by Balents and Nelson [8].

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