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Melting of the vortex lattice in $YBa_2Cu_4O_8$ in parallel fields

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The mixed state c-axis resistivity of $YBa_2Cu_4O_8$ has been measured with the magnetic field applied parallel to the a, b, and c axes. For all orientations of the magnetic field, a ''kink'' is observed in the resistive transition, associated with the first-order melting of an anisotropic three-dimensional vortex lattice. While the melting lines for $H\|b$ and $H\|c$ obey the expected relation $H_m=H_0(1-T/T_c)^n$ with $n=1.5$, $H_m(T)$ for $H\|a$ follows a different T dependence with a lower exponent. This result is consistent with a reduction in the dimensionality of $YBa_2Cu_4O_8$ in this field geometry, as observed in normal-state magnetoresistance measurements. [S0163-1829(99)50418-9]

The existence of a first-order melting transition of the vortex lattice in high-T_c superconductors is now well established for the field geometry $H\|c$.[1] For $H\|ab$, however, the situation is more controversial. When a magnetic field penetrates between the CuO_2 layers, the vortices can be subject to an intrinsic pinning mechanism due to the periodic variation of the order parameter along the c axis,[2] in which case, the melting transition is shown to become second order.[3,4] This suppression of the first-order transition was confirmed in beautiful experiments by Kwok *et al.*,[5] who measured the in-plane resistivity of $YBa_2Cu_3O_7$ (Y-123) with high angular resolution and found that the ''kink'' in resistivity at the melting temperature T_m was smeared out when the field was aligned exactly within the ab planes.

This observation alone, however, does not preclude the possibility that a vortex lattice between the planes can also melt via a first-order transition, since the above pinning mechanism comes into effect only when the Lorentz force acts to move vortices across the CuO_2 planes. Charalambous and coworkers, for example, measured the resistive transition in high quality Y-123 single crystals with $I\|c$ and $H\|ab$ and observed a discontinuity in the resistivity that was hysteretic.[6] In this geometry, the Lorentz force is aligned parallel to the CuO_2 layers, and the intrinsic pinning mechanism from the layered structure will be inactive. (In the experiment of Kwok *et al.*, the Lorentz force was acting along the c axis.) Although the angular resolution of the experiments of Charalambous *et al.* is unknown, recent thermodynamic measurements[7,8] support the view that in the absence of intrinsic pinning, the first-order transition in Y-123 is a three-dimensional (3D) phenomenon. In contrast, for highly anisotropic $Bi_2Sr_2CaCu_2O_8$ (Bi-2212), the angular dependence of the melting transition is found to obey 2D scaling over a wide range of angles, and the entropy jump associated with the transition is insensitive to the presence of Josephson vortices within the plane.[9]

Flux lattice melting has been studied almost exclusively for Bi-2212 and Y-123, due primarily to the availability of high quality single crystals with low densities of pinning centers. Since Y-123 ($\gamma\approx7.5$) and Bi-2212 ($\gamma\approx250$) lie at the extremes of the anisotropy spectrum for high-T_c cuprates (γ is defined as the ratio of the in- and out-of-plane penetration depths), it is instructive to look for evidence of melting in other cuprates of intermediate anisotropy, particularly in the geometry $H\|ab$. $YBa_2Cu_4O_8$ (Y-124) offers an attractive possibility since it is a stoichiometric compound with very few oxygen vacancies, and, unlike Y-123, is naturally free of twin-plane defects. Indeed, sharp features in the in-plane resistivity have already been observed in Y-124 for $H\|c$.[10,11] Unfortunately, Y-124 crystals are grown under high-pressure conditions and are typically an order of magnitude smaller than Y-123 or Bi-2212, making high-resolution bulk thermodynamic measurements extremely difficult.

Here, we report measurements of the mixed state c-axis resistivity $\rho_c(T)$ of Y-124, where the intrinsic pinning mechanism due to the periodicity of the CuO_2 planes is inactive. We find a ''kink'' in the resistive transition for all field orientations, that shows many features associated with a first-order melting transition. Moreover, in contrast to the data of Kwok *et al.*,[5] the kink actually becomes *more* pronounced as the field is tilted into the ab plane. The melting lines for $H\|b$ and $H\|c$ obey the expected relation $H_m=H_0(1-T/T_c)^{1.5}$. The anisotropy of the melting line $H_m^b/H_m^c\approx15$, in good agreement with the anisotropy parameter determined by torque magnetometry.[12] We also observe in-plane anisotropy of the melting line, which increases as the melting field increases. This result is consistent with a reduction in the dimensionality of Y-124 when $H\|a$, as deduced from normal-state magnetotransport measurements.[13]

Single crystals of Y-124 were grown by a flux method in Y_2O_3 crucibles and an Ar/O_2 mixture at 2000 bars with a partial O_2 pressure of 400 bars. Details are reported elsewhere.[14] Several crystals from two batches were used in this study. T_c (midpoint) values were all 82 K$\pm$1 K with a sharp transition width of 0.5 K (90–100 %) of the resistive transition), followed invariably by a small foot in the transition of width 0.5–1.0 K. For $\rho_c(T)$ measurements, large current and voltage contacts were placed on the top and bottom faces of thick crystals in a quasi-Montgomery configuration

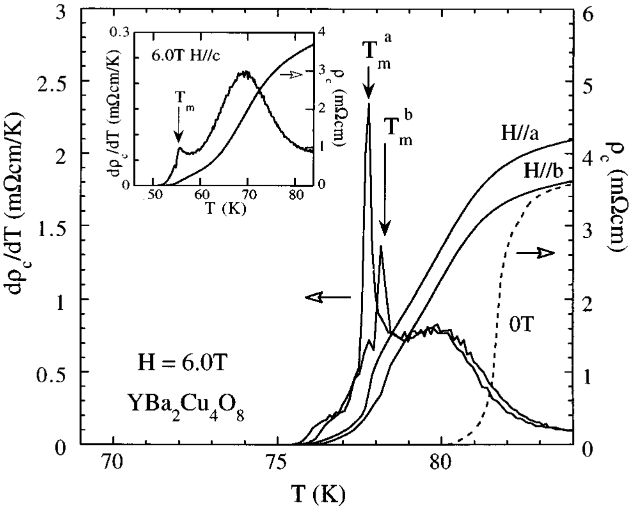

FIG. 1. $\rho_c(T)$ sweeps, and their temperature derivatives, for Y-124 for $H\|a$ and $H\|b$ (H=6 T). The zero-field transition is also shown as a dashed line. The arrows labeled T_m indicate the position of the resistive kink. Inset: 6 T sweep for $H\|c$.

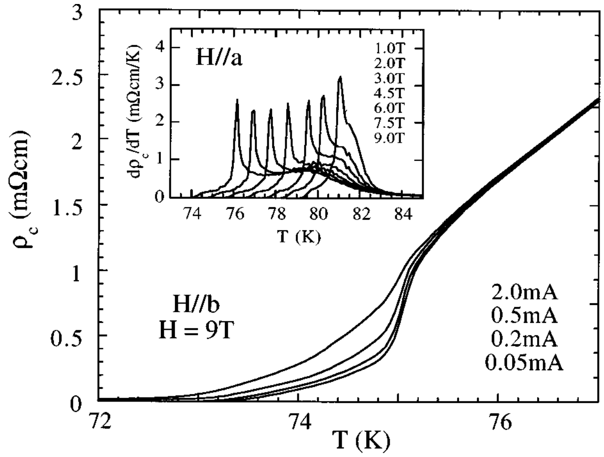

FIG. 2. $\rho_c(T)$ sweeps for $H\|b$ (H=9 T) at different excitation voltages, showing the nonohmic resistivity behavior below T_m. (These data were taken on a second crystal with a lower T_c.) Inset: Derivatives of $\rho_c(T)$ sweeps for $H\|a$ at different fields. Note that both the height and width of the derivative peak are independent of the value of H.

using Dupont 6338 silver paint. Typical dimensions of the crystals used were 200 μm×100 μm×100 μm. Zero-field $\rho_c(T)$ data were all consistent with previous work.[15] For other crystals from the same batches, the in-plane anisotropy in the normal-state resistivity was found to be extremely large ($\rho_a/\rho_b \approx 6$ at T_c), confirming the highly conducting nature fo the CuO chains in these samples and the high quality of the crystals.

Constant field temperature sweeps were carried out in three separate cryostats. The bulk of the measurements were made using a commercial Quantum Design physical properties measurement system (PPMS) with a field capability of 9 T. Higher field measurements up to 18 T were made using an Oxford Instruments superconducting magnet cryostat. For these two experiments, the field alignment with the crystal axes was estimated to be ±2°. For fine angular measurements with $H\|ab$, we used a 5 T split coil magnet with a two-axis rotation stage and an angular resolution of ±0.1°.

Figure 1 shows a series of constant field $\rho_c(T)$ sweeps and their derivatives $d\rho_c/dT(T)$ plotted for $H\|a$, $H\|b$, and $H\|c$ (inset) at H=6 T. The zero-field transition is also shown as a dashed line. Note the large normal-state magnetoresistance for $H\|a$. This large magnetoresistance was described in our earlier work[13] and is attributed to the large Lorentz force on the carriers in the highly conducting CuO chains that run parallel to the b axis. At each field, and for each field orientation, a kink is observed in the resistivity curve, as indicated by an arrow. Rather surprisingly, the kink is most pronounced for $H\|a$, and is only just discernible for $H\|c$ near the foot of the transition. This is probably due to the fact that for $H\|c$, the Lorentz force is absent. Recent in-plane resistivity measurements on Y-124 in the mixed state showed a much more pronounced kink for $H\|c$.[10] In fact, the derivative $d\rho_c/dT$ for $H\|c$ reported in Ref. 10 has a remarkably similar shape to that shown here for $H\|a$. In both cases, the Lorentz force on the vortices was acting along the b axis.

It has been shown for Y-123 that the sharp feature in the mixed state resistivity coincides with the step in magnetization at T_m,[16] and as such, is regarded as a clear signature of the first-order melting transition. Unfortunately, the small size of our crystals prevents a similar comparison being made here for Y-124. However, there are certain features of the resistive kink that reveals its relation to a first-order phase transition.

Figure 2 shows $\rho_c(T)$ curves in the vicinity of the kink for $H\|b$ with H=9 T, taken with a series of different excitation currents. (These measurements were taken on a second crystal with a lower T_c). The data show strongly nonohmic behavior below T_m, while above this temperature, it is clearly ohmic. Furthermore, the width of the resistive kink, and its step height were found to be independent of the strength of magnetic field. This is illustrated in the inset to Fig. 2, where the derivatives $d\rho_c/dT$ are plotted for a selection of different field strengths for $H\|a$. The width and height of the derivative peak are essentially constant for all fields up to 9 T; the peak position merely shifts to lower temperatures with increasing field.[17] Both these features were observed previously in Y-123 for $H\|ab$ (Ref. 6) and $H\|c$ (Ref. 18) and are regarded as key signatures of a first-order transition in the vortex lattice.[19] It should be added here that no clear evidence for hysteretic behavior around T_m was detected in our measurements within experimental resolution, though hysteresis in transport measurements itself is not necessarily direct evidence of a first-order melting transition.[20]

To confirm that the presence of the kink for $H\|ab$ is not an experimental artifact due to misalignment of the field with respect to the planes, we carried out a fine angular study of the kink feature at small angles relative to the planes using a two-axis rotation stage inside a 5 T split-coil magnet. In this experiment, we initially aligned the crystal within the plane by locating the sharp dip in resistivity for $H\|b$ (to within ±0.1°), rotated by 90° to $H\|a$, then measured constant field temperature sweeps with small angular increments of 0.9° on either side of the initial alignment.

Figure 3 shows a selection of $\rho_c(T)$ curves for different angles close to $H\|a$. As we approach parallel alignment, the resistive kink actually becomes more pronounced, i.e., the

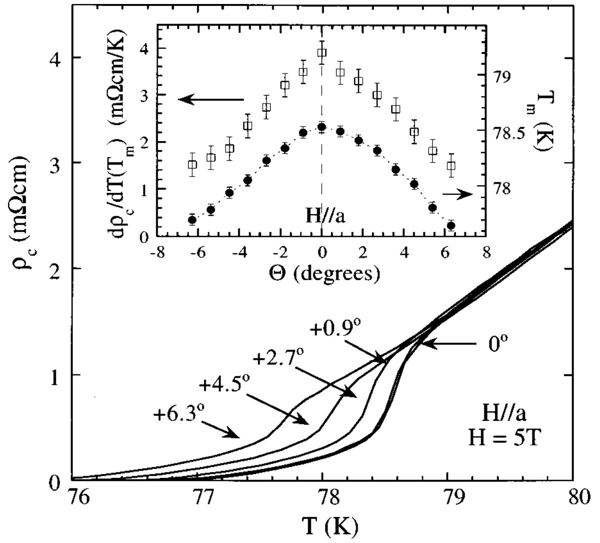

FIG. 3. $\rho_c(T)$ sweeps for fields close to $H\|a$ (H=5 T). The angles indicated refer to the direction of the field relative to the a axis. Inset: The melting temperature (solid circles) and the height of the derivative $d\rho_c/dT$ at T_m for fields close to $H\|a$.

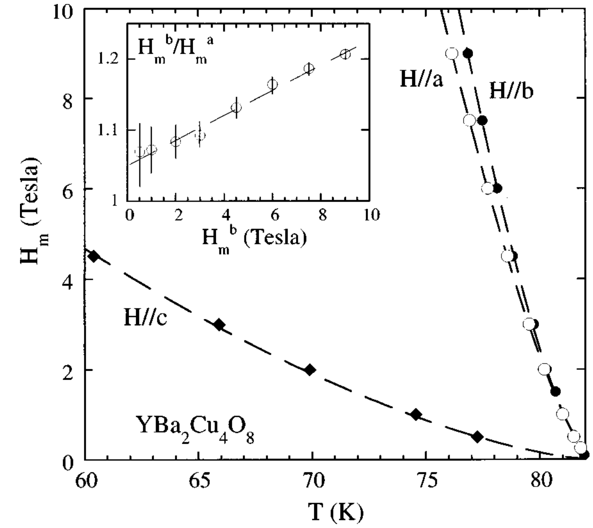

FIG. 4. $H_m(T)$ of Y-124 for $H\|a$ (open circles), $H\|b$ (closed circles), and $H\|c$ (closed diamonds). Inset: In-plane anisotropy of the melting line as a function of H_m^b.

drop in resistivity increases as $\theta\rightarrow 0$. The width of the kink remains unchanged, implying that the first-order transition is preserved for all angles. This result is in marked contrast to the behavior observed by Kwok *et al.* for Y-123 with both field and current in the plane,[5] and seems to confirm that the first-order melting transition for $H\|ab$ can be suppressed only when the periodic pinning mechanism is active.

In the inset to Fig. 3, we have plotted the melting temperature T_m and the height of the derivative peak at T_m as a function of field angle to illustrate how the transition evolves as we sweep through parallel alignment $H\|a$. The position of the peak is symmetric about our initial alignment to within our experimental accuracy of $\pm 0.1°$. Furthermore, the smooth variation of the peak position with angle gives us added confidence that the resistive kink for $H\|a$ is not due to any misalignment of the magnetic field. If the melting transition was due solely to the field component perpendicular to the planes, as expected for a two-dimensional superconductor for example, one would expect the peak temperature to show rapid changes (diverging as $1/\sin\theta$) for small increments in angle as $\theta\rightarrow 0$. As a further check of our alignment, we performed a similar series of sweeps while rotating the field in the *ab* plane (i.e., rotating the field towards the *b* axis). No noticeable changes in the peak structure were observed for $\theta\pm 10°$.

Perhaps the most compelling evidence however, is that the derivative peak has a maximum at θ=0. To our knowledge, this is the first observation of a first-order melting transition in the parallel field alignment in a cuprate system other than Y-123. This result is made more significant in that the anisotropy of the superconducting state in Y-124 is essentially twice that of Y-123. A recent theoretical study of the Lawrence-Doniach model with $H\|ab$ has suggested that even in a 2D Josephson-coupled superconductor, the melting transition can remain first-order for in-plane fields, in the absence of defects.[21] This contradicts numerical simulations carried out for Josephson-coupled superconductors that appear to show the transition is continuous when $H\|ab$.[4] Clearly, it is important to establish experimentally whether intrinsic Josephson coupling and a first-order melting transition in the plane can coexist. For optimally doped Y-123, there is no clear evidence for intrinsic Josephson coupling between the planes[22] and even for Y-124, it should be recalled that in the normal state, $\rho_c(T)$ is metallic,[15] and so it is not unrealistic to presume that Y-124 undergoes a transition from an anisotropic 3D metal to an anisotropic 3D superconductor. Hence we are not in a position to confirm this correlation at the present time. In this regard, it would be very interesting to look for evidence of intrinsic Josephson effects in Y-124, or alternatively, to investigate the mixed state *c*-axis resistivity in underdoped Y-123, where a crossover to nonmetallic $\rho_c(T)$ and a 2D superconducting state is known to take place.[23]

The temperature dependence of the melting lines for all three field orientations are given in Fig. 4. For $H\|b$ and $H\|c$, $H_m(T)$ obeys the expected relation $H_m=H_0(1-T/T_c)^{1.5}$. For $H\|c, H_0=33.1$, which agrees well with the values reported by Qiu *et al.*[10] $[H_m=32(1-T/T_c)^{1.47}]$ and Metlushko *et al.*[11] $[H_m=32(1-T/T_c)^{1.4}]$ with $I\|ab$. The anisotropy of the melting line $H_m^b/H_m^c\approx 15$, in good agreement with the anisotropy parameter determined by torque magnetometry[12] and from the angular dependence of the melting transition.[10] For $H\|a$, the melting line was found to follow a different T dependence; the best fit was obtained using the expression $H_m=H_0(1-T/T_c)^{1.38}$. This implies that the in-plane anisotropy of the melting line actually *increases* with decreasing T or increasing H_m. This can be seen more clearly in the inset to Fig. 4, where we have plotted the ratio H_m^b/H_m^a for each value of H_m^b. In-plane anisotropy of H_m has also been observed previously in Y-123,[7] though for Y-123 both melting lines were found to have the same T dependence with a constant anisotropy factor of $\approx$1.2.

As noted earlier, in the normal state, $\rho_c(T)$ increases rapidly with increasing field when $H\|a$.[13] By contrast, the in-plane resistivity stays relatively unchanged. Thus, the resistive anisotropy in Y-124 is strongly field dependent for $H\|a$.

We conclude, therefore, that the in-plane anisotropy of $H_m(T)$ in Y-124 is also field induced and reflects the correlation between the position of the melting line and the normal-state resistive anisotropy recently proposed by Sasagawa *et al.*[24] When the magnetic field ($H \| a$) exceeds 20 T, $\rho_c(T)$ in Y-124 becomes nonmetallic,[13] implying a 3D to 2D crossover in the normal state. It would be interesting to investigate what happens to the first-order melting transition in Y-124 in this field range to determine if there is a correlation between the dimensionality of the normal state and the first-order melting transition in the in-plane vortex lattice. Preliminary data reveal that the kind feature in $\rho_c(T)$ is still present up to 18 T. Higher field measurements are envisaged in the future.

In conclusion, we have shown evidence for a first-order melting transition in Y-124 for fields both perpendicular and parallel to the CuO_2 planes. This phenomenon has been observed previously only for optimally doped Y-123. Since both systems are anisotropic 3D metals in the normal state, we conclude that a first-order melting transition for $H \| ab$ does exist in clean layered superconductors in the presence of (relatively) strong interlayer coupling and when the strong pinning mechanism, imposed when in-plane vortices are forced to jump across the planes, is rendered inactive.

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