Absence of a Kosterlitz-Thouless transition in ultrathin $YBa_2Cu_3O_{7-\delta}$ films

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We have performed nanovolt-level dc measurements on single-unit-cell films of $YBa_2Cu_3O_{7-\delta}$. When only the low-sensitivity data are analyzed, they are roughly consistent with the occurrence of a Kosterlitz-Thouless transition. At the lowest currents and voltages, however, the samples are ohmic at temperatures quite far below the nominal Kosterlitz-Thouless transition temperature, contrary to the theory. The ohmic response at low currents implies that there are unbound vortices present well below the nominal Kosterlitz-Thouless transition temperature. [S0163-1829(96)52738-4]

In thin superconducting films there is believed to be a population of equal numbers of oppositely aligned vortices present even in a zero applied field. If these vortices were free to move, they would produce an ohmic response to an applied current. However, the theory of Kosterlitz and Thouless[1] (KT) predicts that for a large enough sample and a logarithmic attractive potential between the oppositely aligned vortices, the vortices should condense into a state of bound pairs at low temperatures. In this case there would be no ohmic response to the applied current, but instead there would be a nonlinear dependence of the voltage on current since the current can unbind more weakly bound pairs.[2]

This theory was not originally thought to be applicable to superconductors[1] because the potential between vortices only has the required logarithmic form for vortices separated by less than $\lambda_\perp = \lambda^2/d \sim 1/n_s$, where λ is the bulk penetration depth, d is the thickness of the superconductor, and $n_s = n_s^{3D} d$ is the areal superfluid density (3D is three dimensional).[3] For lengths greater than $\lambda_\perp$, the potential is bounded, so that pairs can unbind thermally at all temperatures. This means that for samples larger than $\lambda_\perp$, there will be a population of free vortices in the sample even below the nominal vortex unbinding transition temperature.

In dirty superconductors $\lambda_\perp$ is enhanced by a factor of ξ_0/ℓ, the ratio of the Pippard coherence length to the mean free path, and in some materials $\lambda_\perp$ is on the order of millimeters.[4,5] In fact, experiments in dirty low-T_c superconductors have shown evidence of this transition.[6,7] The cuprate superconductors are not in the dirty limit, so there is no such enhancement of the bulk penetration depth. It is, therefore, surprising that a number of papers have reported a KT transition in YBCO (Ref. 8), often in quite thick samples, where the penetration depth is apparently quite small. This suggests that either the KT theory applies in a regime where it is not expected to apply, or that the superfluid density in the cuprates is much lower than is generally believed.

To study this problem further, we have measured films of varying thickness. The data presented here are of the thinnest available samples, single-unit-cell thick films, $d = 12$ Å, of $YBa_2Cu_3O_{7-\delta}$ (YBCO). Ongoing work on the crossover from 2D to 3D behavior of the transition will be presented in a future publication.

The samples used were trilayer structures deposited by pulsed laser deposition onto $NdGaO_3$ substrates. The layers consisted of an eight-unit-cell buffer layer of $Pr_{0.6}Y_{0.4}Ba_2Cu_3O_{7-\delta}$ (PYBCO), a single unit cell of $YBa_2Cu_3O_{7-\delta}$, and a sixteen-unit-cell cap layer of PYBCO of the same composition as the bottom layer.[9,10] The following thickness calibration method was used for the single-unit-cell films. First 30 nm films of YBCO and PYBCO were deposited separately. Then a step was created by etching away part of the film, the height of which was measured using a profilometer. The thickness calibration is given by the ratio of this height to the number of laser pulses used to make the films. A cross calibration was performed using low-angle x-ray diffraction between the surface of the film and the interface between the film and the substrate. This result was consistent with the method above. The accuracy of this calibration is about 10%, with the uncertainty mainly due to fluctuations in the energy of the laser. An uncertainty of this magnitude is not believed to affect significantly the dc electrical transport properties of these films. In fact, measurements of the dc conductance of films made by this method show a steplike behavior as a function of thickness.[9] Our measurements of the normal-state resistivity of films of varying thickness show that the resistivity is independent of the thickness of the film to within the 10% error due to the correction for the buffer layers.[10] (The conductance of the YBCO layer was assumed to be the difference between the conductance of the entire trilayer structure and that of a separately prepared sample made of only PYBCO and with the same thickness as the PYBCO buffer layers of the trilayer samples.) We do not have enough samples of different thicknesses to detect these steps in the conductance. More details about the deposition method and structural information on these films can be found in Ref. 10.

The samples were patterned with a laser into a four-terminal geometry suitable for transport measurements, resulting in a rectangular stripe of material 1–2 mm in length and 100–400 μm in width. To reduce contact resistance, contacts were made to four 1000-Å-thick YBCO pads, which were deposited on the substrate prior to sample deposition.

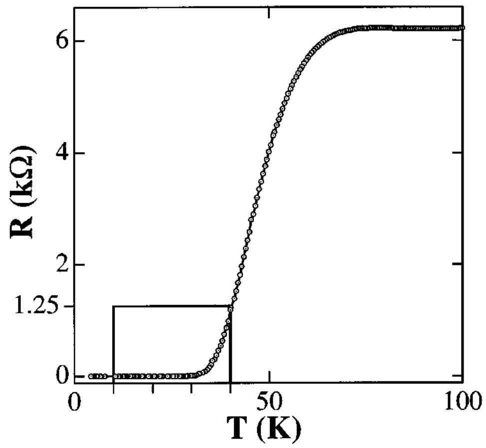

FIG. 1. dc resistance vs temperature at a current of 10 μA for a single unit-cell YBCO film with PYBCO buffer layers above and below. This sample was 200 μm wide by 2000 μm long. The data include the contributions to the conductivity from the PYBCO layers, which amount to about 35% of the total at 100 K. The box corresponds to the temperatures of the curves shown in Figs. 2 and 3.

Measurements were made of the longitudinal voltage of three of these films as a function of dc current and temperature. The data from the three samples are self-consistent. The current from the dc source was reversed for each point to eliminate any offset voltage. The subtracted points were then averaged, resulting in an rms noise of about 2 nV. The residual field in the sample environment was measured to be less than 0.1 mG.

The resistance of the entire trilayer structure as a function of temperature is shown in Fig. 1.

The voltage as a function of current (IV) data are shown in Fig. 2. At the lower temperatures, the IV curves have approximately a power law form. As the temperature is increased, the IV curves start to develop a region of decreasing slope for the lowest currents. As the temperature is increased further, the curves have an ohmic behavior for low currents. For the highest temperatures, the curves are ohmic for the entire current range.

For low temperatures, the KT theory predicts that the voltage varies as a power of the current, with the power greater than 3 and increasing as the temperature decreases. At the Kosterlitz-Thouless transition temperature T_{KT}, the voltage is predicted to vary as the third power of the current. Above the transition temperature, the IV curves are predicted to be linear, (ohmic) for low currents, and to follow a power law form, with power less than 3, for higher currents.[2,11] That is, for low currents, $V \sim I^{a(T)}$, where $a(T) \geqslant 3$ for $T \leqslant T_{KT}$ and $a(T)=1$ for $T > T_{KT}$. $a(T)$ is predicted to change sharply from 3 to 1 at T_{KT}.

In our data, the predicted sharp change in $a(T)$ from 3 to 1 at T_{KT} is not seen: there is no discontinuity in the behavior from higher-temperature IV curves which are linear for small currents ($a=1$) to lower temperature IV curves which are nonlinear at the lowest currents. This was true for our three single-unit-cell films as well as a four-unit-cell film. However, some rounding of this transition is expected in curves with $a(T) < 3$ due to the inability to measure the voltage at low enough currents.[11]

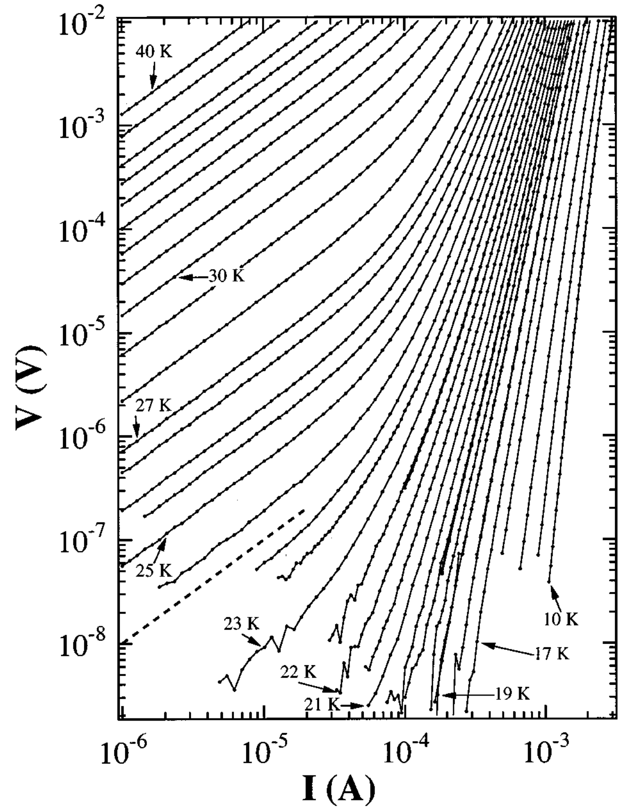

FIG. 2. Current-voltage characteristics at different temperatures for the sample which produced the data shown in Fig. 1. The temperature decreases from 40 K at the upper left to 10 K at the right. The solid lines are a guide to the eye. The dashed line at the bottom represents a slope of one, or ohmic behavior.

This rounding of the transition does not explain the ohmic low-current behavior of the curves with $a(T)$ greater than 3: the curve at $T=23$ K in Fig. 2 exhibits ohmic behavior for low currents, yet has a power of 4.6 for higher currents. Also, curves at slightly lower temperatures, which do not have an ohmic tail for low currents, still show that the voltage is a decreasing power of current for decreasing currents. This is easiest to see from Fig. 3, which represents the slopes of the $\log(V)$ vs $\log(I)$ curves calculated from the data shown in Fig. 2. It can be seen in this graph that the IV curves at lower temperatures have the same qualitative form as those at higher temperatures. The data suggest that a measurement with higher resolution would reveal ohmic behavior at even lower temperatures.

There are two effects not intrinsic to the sample which could produce an anomalous linear tail on the data: a magnetic field would create free vortices, and current noise in the sample would cause the averaged voltage to become linear as a function of current for currents smaller than the level of the current noise.

A detailed analysis of the effect of current noise on the measurement revealed the magnitude of the effect would be too small to explain our data.[12]

A magnetic field perpendicular to the sample would produce free vortices, and thus an ohmic response for low currents. To check for this, we applied a small magnetic field perpendicular to the sample and measured the voltage as a function of applied field. It was found that the voltage, which

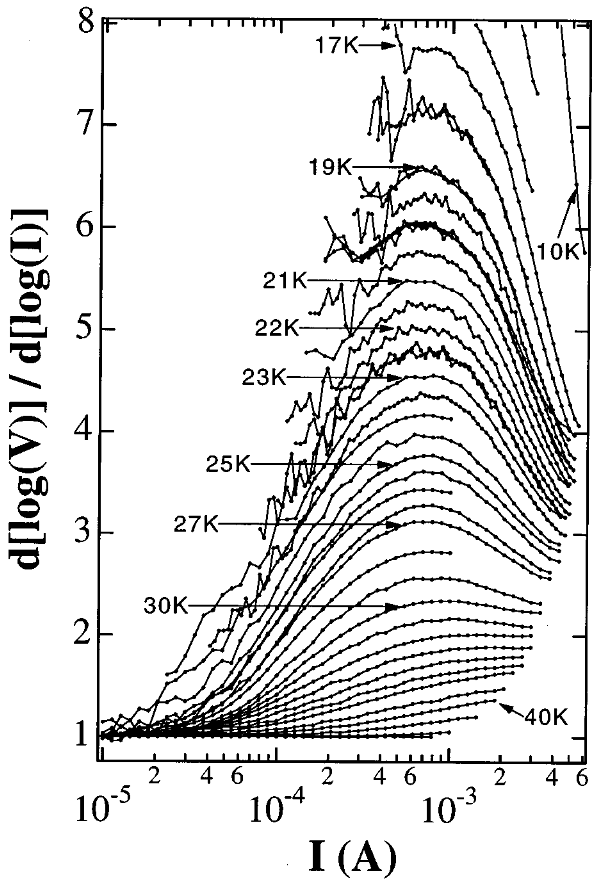

FIG. 3. $a(I,T)\equiv d(\log V)/d(\log I)$ as a function of current. The different curves represent different temperatures, with the temperature decreasing from 10 K at the extreme upper right to 60 K at the bottom. The solid lines are a guide for the eye.

was a linear function of the magnitude of the field, was a minimum at zero applied field, allowing us to conclude that stray or trapped fields were not the cause of the linear portion of the *IV* curves.

We can semiquantitatively account for our low-temperature *IV* curves by assuming that there is a thermal population of paired vortices, given approximately by the Kosterlitz-Thouless theory, plus a density of free vortices given by a Boltzmann factor,

$$n_{f0}(T)=C_T(2\lambda_\perp/\xi_c)^{-E_0/2k_BT}e^{-E_0/2k_BT}. \tag{1}$$

where ξ_c is the vortex core size, $E_0=2\pi n_s\hbar^2/m^*$, where n_s is the 2D superfluid density, L is the smallest of the length or width of the sample, and C_T is a function only of temperature. Equation (1) results from the fact that the energy required to unbind a pair is bounded in a sample which is much larger than $\lambda_\perp$. This density of free vortices accounts for the ohmic tail in the data below the nominal T_{KT}, as we will show below.

When an external current is applied to the sample, it creates additional free vortices by unbinding some of the pairs which are still bound. When the density of current-induced free vortices, n_{f1}, is much greater than the thermal background population given by Eq. (1), we expect to recover the Halperin-Nelson result,[2]

$$n_f(T)\approx n_{f1}(T)=C_p(j_s/j_0)^{E_0/2k_BT}, \tag{2}$$

where n_f is the total density of free vortices, $j_0=\hbar n_s e/2m^*\xi_c$, and C_p is analogous to C_T.

In general, it is expected that

$$V/I\sim n_f, \tag{3}$$

where n_f is given by Eq. (1) at low currents and by Eq. (2) at high currents. Thus, we expect that $V\sim I^{a(I,T)}$, where

$$a(I,T)=1, \tag{4a}$$

$$a(I,T)=1+E_0/2k_BT=1+\pi n_s\hbar^2/m^*k_BT$$
$$=1+\Phi_0^2/16\pi^2\lambda_\perp k_BT, \tag{4b}$$

where n_s and $\lambda_\perp$ are the fully renormalized quantities. Equation (4a) is expected to hold for low currents, and Eq. (4b) is expected to hold for high currents. From the above discussion, we see that the *IV* curves should be linear at small currents, where they are dominated by intrinsic thermally induced free vortices, crossing over to nonlinear power-law behavior at higher currents. This provides an explanation for earlier reports of the KT transition occurring in YBCO: if measurements are limited by voltage sensitivity, then the linear parts of the curves would not be detected, thus giving the impression the curves have a transition from 1 to 3. This simple model does not take into account the renormalization due to the population of free vortices.

There are some inconsistencies in this analysis, however. The $\lambda_\perp$ recovered from the data and Eq. (4b) was a good fit to the empirical equation $\lambda_\perp(T)=\lambda_\perp(0)/[1-(T/T_{C0})^4]$. This gives $\lambda_\perp(0)=160$ μm, which is about a factor of 4 higher than expected based on the bulk value measured from single crystals,[13] but still smaller than the sample size. The discrepancy in $\lambda_\perp$ cannot be explained by renormalization due to bound pairs, as the analysis of Davis *et al.*[14] has shown that the effect of this renormalization is negligible at low temperatures.

In summary, our data show that a Kosterlitz-Thouless transition from a low-temperature state with only paired vortices to a high-temperature state with free vortices does not occur in single-unit cell thick YBCO. We propose that because of the relatively high value of n_s (and thus the relatively small value of $\lambda_\perp$), there are free vortices present at temperatures much below the nominal KT transition temperature. In addition to this intrinsic population of free vortices, there is also a population of paired vortices which can be unbound by nonzero current. While data reported previously are consistent with our higher-current data, our interpretation differs: Based on more extensive lower current data, we believe that only current-induced vortex unbinding in the presence of intrinsic free vortices occurs, but a true KT transition does not occur.

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