Experimental Evidence of a Dimensional Crossover in $Y_1Ba_2Cu_3O_{7-\delta}$

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High-resolution torque-magnetometry data have been obtained on an untwinned single crystal of $Y_1Ba_2Cu_3O_{7-\delta}$ in the temperature range $63 < T < 88$ K ($T_c = 90.5$ K). At $T = 80$ K and above, the data are fitted extremely well with the accepted three-dimensional phenomenological theory, but below this temperature an anomalous torque develops when the magnetic field lies close to the Cu-O planes. The qualitative and quantitative features of our results provide strong evidence that breakdown of the three-dimensional description below 80 K is associated with a crossover to two-dimensional superconducting behavior.

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One of the main characteristics thought to distinguish high-T_c from conventional superconductivity is an extremely short zero-temperature coherence length, ξ_0. Along the c direction in $Y_1Ba_2Cu_3O_{7-\delta}$, for example, it has been estimated[1] that $\xi_0 \sim 3$ Å. This is significantly smaller than the spacing of the Cu-O planes that are considered to be mainly responsible for the superconductivity. In all superconductors, the coherence length should increase without limit as the temperature approaches T_c. An unavoidable consequence for all high-T_c materials is that some sort of crossover from two- to three-dimensional behavior should occur at an intermediate temperature. This temperature may be estimated by equating the Ginzburg-Landau coherence length, $\xi_0(1-T/T_c)^{-1/2}$, to an appropriate plane spacing. Choosing a value of 8 Å for the latter, one obtains[2] an estimate of 74 K for the crossover temperature in $Y_1Ba_2Cu_3O_{7-\delta}$. It is difficult to assign an uncertainty to this estimate, but a dimensional crossover somewhere in the region from 70 to 80 K seems to be a necessary consequence of our present understanding.[3] In principle, upper-critical-field anisotropy measurements[1,2] provide the most direct means of searching for this phenomenon. Unfortunately, at 74 K the magnitude of H_{c2} measured with the field parallel to the Cu-O planes is thought to be of the order of 100 T, making the direct approach impractical.

An alternative method for probing superconducting anisotropy, which does not require such large fields, is torque magnetometry.[4] In this technique, one measures the torque τ on a sample in the presence of a field H as a function of the angle θ which the field makes with the **c** axis. There is a field-strength requirement,[5] namely, $H \gg H_{c1}$, but this is well satisfied[6] in the present experiments with $H = 1$ T. For temperatures close to T_c, the angular dependence of the observed torque, $\tau(\theta)$, for both $Y_1Ba_2Cu_3O_{7-\delta}$ and $Bi_2Sr_2Ca_1Cu_2O_{8+\delta}$, is successfully described[4,7] by a London treatment,[5] which gives

$$\tau(\theta) = A\frac{\sin 2\theta}{\epsilon(\theta)}\ln\left[\frac{\gamma\eta H_{c2\parallel}}{H\epsilon(\theta)}\right],$$
$$\epsilon(\theta) = (\sin^2\theta + \gamma^2\cos^2\theta)^{1/2}. \tag{1}$$

In this equation, A is a parameter independent of angle, and $\gamma = (m_c/m_a)^{1/2}$, where m_c and m_a are the Ginzburg-Landau superconducting effective masses for pair motion along the **c** direction and in the **a**-**b** plane, respectively. $H_{c2\parallel}$ is the upper critical field measured along the **c** axis and η is a constant of order unity.[5] The theory leading to Eq. (1) is explicitly three dimensional in character, and no formal treatment of the (more difficult) two-dimensional case is available as yet. Nonetheless, we report here the observation of systematic departures from the 3D result in $Y_1Ba_2Cu_3O_{7-\delta}$. As the temperature is reduced, we find that the deviations emerge cleanly in the vicinity of the anticipated temperature, and exhibit a number of both qualitative and quantitative features expected to accompany a crossover to 2D behavior.

Torque measurements have been made on a total of ten $Y_1Ba_2Cu_3O_{7-\delta}$ single crystals. For a typical "large" twinned crystal, with a flat-plate geometry and edge dimensions of the order of 1 mm, we find irreversibility in the angular variation of the torque in a field of 1 T that is large enough to preclude a useful comparison with Eq. (1) below ~90 K. Such irreversibility, reflecting flux pinning, has been observed by others,[8] and appears to be a general feature of even the best twinned crystals. We observed a reduction in pinning for two large untwinned crystals; however, even for them, completely reversible

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data could be obtained only down to ~ 88 K. In conventional magnetization measurements, Bean's model[9] implies that the ratio of the hysteretic-to-reversible magnetization can be made as small as desired by using sufficiently small samples. A similar situation appears to hold for the transverse magnetization that is measured by torque magnetometry. We have found that the irreversibility is dramatically reduced in small crystals, for both untwinned and twinned material. In fact, with an important qualification that will be discussed later, the torque data that are reported here were reversible to within our measurement resolution over the complete angular and temperature range investigated.

We have made a detailed investigation of three small crystals. The data reported here are for one of these, which was untwinned and mounted so that when $\theta = 90°$ the field lay along the **b** axis. (Experimentally indistinguishable results were obtained in a separate run where the **a** axis was selected. Also, even though the other two crystals were twinned and displayed significant irreversibility, they demonstrated deviations from 3D behavior similar to those reported here.) Our measurements were made with a null-deflection torque magnetometer.[10] The major measurement uncertainty results from instrumental base-line instability over the measurement period. The time required to obtain a complete set of angular data at a single temperature is ~ 2 h, and the instability over this period was found to be $< 10^{-4}$ dyn cm, independent of temperature. The maximum torque observed varied between $\sim 10^{-2}$ dyn cm at $T = 88$ K and $\sim 10^{-1}$ dyn cm at $T = 63$ K. Our estimate of the uncertainty associated with a single measurement of $\tau(\theta)$ therefore ranges from 0.1% to 1%, at the lowest and highest temperatures, respectively. The temperature stability during a set of measurments was ± 0.01 K, and angular displacements of the field relative to the crystal were measured with an uncertainty of $\pm 0.03°$.

The parent of the untwinned crystal mentioned above was grown using the flux method as described elsewhere.[11] The oxygenation that was used is unusual; it produces untwinned crystals. Twinning is avoided by quenching the crystal to room temperature in the tetragonal phase, and then reheating in a stress-free environment, as previously discussed.[12] (The **a**, **b**, and **c** axes can be identified, as described elsewhere.[13]) The crystal used in our work was cut from this parent and was in the form of a square plate, of approximate dimensions $0.02 \times 0.17 \times 0.17$ mm^3. Its superconducting behavior was checked by measuring the torque as a function of temperature with the field H and field angle θ both held constant. The result, shown in Fig. 1(a), provides a direct measure of T_c ($= 90.5$ K). It is also demonstrates the high degree of temperature reversibility, extending down to the lowest temperature investigated ($T = 65$ K). Note also the linear behavior of the torque over a temperature range extending to within a fraction of a degree of T_c, testifying to the excellent sample homogeneity.

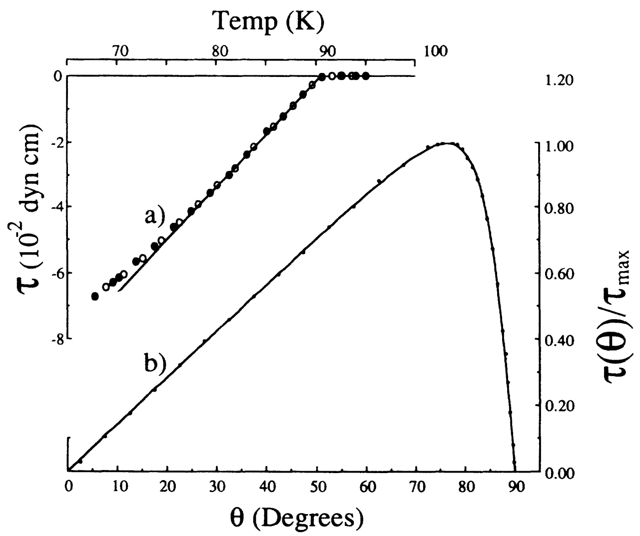

FIG. 1. (a) Temperature dependence of the torque for the untwinned single crystal of $Y_1Ba_2Cu_3O_{7-\delta}$ discussed in the text. The field was held constant at 1 T and the angle of the field to the **c** axis was 45°. Solid circles represent data taken with the temperature increasing; open circles with it decreasing. The straight line is drawn through the higher-temperature data to indicate the negligible rounding close to T_c. (b) The angular dependence of the torque, $\tau(\theta)$, normalized to its maximum value, τ_{max}, and measured at $T = 80$ K in a field of 1 T. The experimental uncertainty is represented by the size of the points. The solid line is a least-squares fit by Eq. (1), giving $\gamma = 7.9 \pm 0.2$ and $\eta H_{c2\parallel} = 23 \pm 4$ T.

Figure 1(b) displays the torque observed at $T = 80$ K in a field of 1 T, normalized to its maximum value τ_{max}, together with the curve representing the least-squares fit by Eq. (1), treating γ and $\eta H_{2c\parallel}$ as fitting parameters. The standard deviation of the points from the theoretical curve is 0.7%, comparable with the estimated uncertainty of the torque data itself. The value found for the anisotropy parameter at this temperature was $\gamma = 7.9 \pm 0.2$, where the indicated uncertainty represents 1 standard deviation. Similarly good fits with Eq. (1), with $\gamma = 7.7 \pm 0.2$, were obtained over the whole range $80 < T < 88$ K; i.e., over this temperature range the superconducting anisotropy is independent of temperature to within 3%.[14]

The three-dimensional expression continues to provide excellent fits to the data at lower temperatures, but only for angles up to the peak in the torque ($\theta \sim 75°$). At higher angles, a clear anomaly develops at lower temperatures. The high-angle portions of the overall fits, at $T = 85$, 80, and 75 K, are shown in Fig. 2. As the temperature is reduced from 80 to 75 K, a substantial anomalous torque has appeared. Not only is the high-angle fit at 75 K poor, but we also find that the fitting parameter $\eta H_{c2\parallel}$ is forced up to unphysically large values.[15] Both the sense of the anomaly and also the sense of its variation with angle are physically consistent with the emer-

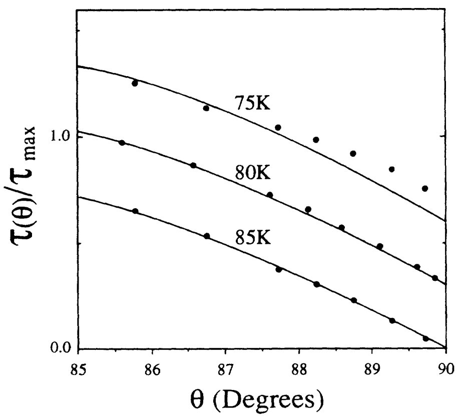

FIG. 2. Plots showing the anomalous high-angle torque that appears below $T=80$ K. The circles are normalized torque data, measured in a field of 1 T at the temperatures indicated. The lines are the high-angle portions of the least-squares fits of each data set by Eq. (1) over the whole angular range from 0° to 90°. (For clarity, the data and associated theoretical curves for $T=80$ K and $T=75$ K have been displaced upwards by 0.3 and 0.6 normalized torque units, respectively.)

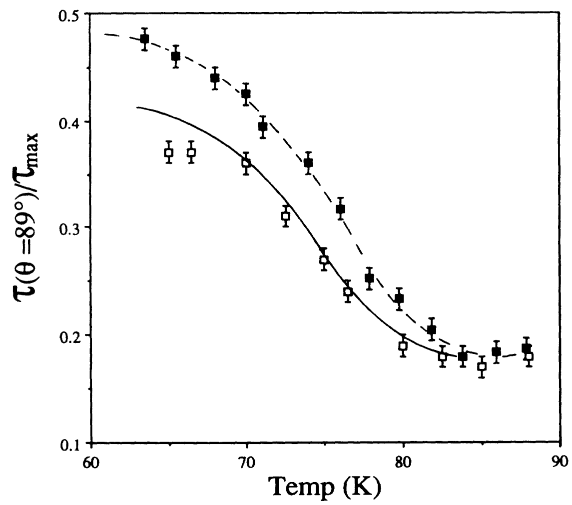

FIG. 3. The temperature dependence of the normalized torque, measured at an angle of 89° to the **c** axis, in a field of 2.3 T (solid squares) and 1.0 T (open squares). The dashed line through the 2.3-T data is to guide the eye. The general crossover argument discussed in the text provides a prediction of the 1-T data which is represented by the solid line.

gence of 2D behavior. We note that the 3D result, Eq. (1), was obtained with the London model, which considers only those energy contributions associated with supercurrents circulating outside the vortex core. In the 2D regime, the core dimension ($\sim\xi$) is less than the spacing of the Cu-O planes. If the field lies in the **a**-**b** plane, it is possible for the core to avoid passing through any plane material. On condensation-energy grounds, this field orientation will therefore correspond to a minimum in the core free-energy contribution. Whatever the specifics of the 2D model, angular displacements of the field out of the **a**-**b** plane in the 2D regime will require an *increased* torque. On the same grounds, the anomalous contribution will decrease in magnitude as the field is moved away from the **a**-**b** plane. Both these features are observed in our data.[16]

For angles below the peak of the torque characteristic in Fig. 1, the angular variation in a field of 1 T is completely reversible at all temperatures. However, at higher angles, a small irreversibility appears at about the same temperature as the anomalous torque (80 K) and grows in magnitude as the temperature is reduced. For example, at $T=75$ K the difference between angle-increasing and angle-decreasing data is 3% of the maximum torque. *This is significantly less than the deviation of either data set from the 3D theory* ($\sim 10\%$), and so the normal procedure of taking the mean of both data sets was used to obtain the equilibrium torque plotted in Fig. 2. We speculate that this small irreversibility is the result of an intrinsic pinning mechanism involving the trapping of vortex cores between the Cu-O planes. It has already been suggested[17-19] that such a phenomenon should be associated with a dimensional crossover. Nonetheless, we cannot dismiss the possibility that the irreversibility is due to some sort of conventional (anisotropic) temperature-dependent pinning. Although it would be remarkable if a 3% irreversibility somehow produced a 10% increase in the equilibrium torque, it might still be argued that, in principle, the irreversibility could be contributing significantly to the anomaly.

However, a further piece of evidence argues strongly against the possibility just mentioned and provides quantitative support for our crossover interpretation. In general, irreversibility decreases as the magnetic field is increased, so the anomaly will also *decrease* if it is linked to pinning. The general 2D torque argument mentioned previously predicts a completely different field dependence. We assume that the (2D) torque due to the core, τ_{2D}, and the (3D) vortex current torque, τ_{3D}, can be summed to give the observed torque. In the field range of interest here, $H_{c1} \ll H \ll H_{c2}$, the vortices are widely separated, and the energy contributed by an individual core will be essentially field independent. However, their number is directly proportional to the field, so τ_{2D} should increase linearly with field. The maximum (3D) torque at all temperatures is expected[5] and observed to increase more slowly with field. This is because it involves the product of the field and the transverse supercurrent magnetization, the latter decreasing logarithmically with field. On this basis, the (normalized) anomalous torque is predicted to *increase* with field.

The normalized torque, measured at $\theta=89°$ in a field of 1 T and also 2.3 T (the maximum field available to us) is plotted as a function of temperature in Fig. 3. At this angle, the hysterisis below 80 K *decreases* by a factor of ~ 2, when the field is increased from 1 to 2.3 T, whereas the anomaly increases significantly. Furthermore, using the 2.3-T data as input, the argument given above allows us to predict[20] numerically the torque at 1 T. This prediction is shown as the solid line in Fig. 3, and is in reasonable quantitative agreement with experiment. Finally, the results shown in Fig. 3 indicate that the anomaly develops in the general vicinity of the temperature at which a dimensional crossover has been anticipated.

Our best present estimates for the zero-temperature coherence length in high-T_c materials are provided by extrapolations from magnetization measurements[1,2] performed close to T_c. Observation of a dimensional crossover offers a direct spatial measure of the coherence length. In the work presented, high-resolution torque-magnetometry data provide evidence that a dimensional crossover occurs at approximately the anticipated temperature in $Y_1Ba_2Cu_3O_{7-\delta}$. Our work therefore provides direct evidence that the zero-temperature coherence length in this material is close to atomic dimensions.

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[14]The γ value found here is somewhat higher than that obtained previously (Ref. 3) from torque measurements on a grain-aligned sample ($\gamma \sim 5$). However, we have found that the anisotropy varies significantly between different crystals. There is a (rough) correlation between higher γ values and higher T_c's, but we have not yet established a definitive (torque) value for the intrinsic anisotropy of $Y_1Ba_2Cu_3O_{7-\delta}$. We remark that in the temperature range described by Eq. (1), $80 < T < 88$ K, the $\eta H_{c2\parallel}$ values systematically decrease, with a slope of -2.8 T/K close to T_c. Since $d(H_{c2\parallel})/dT$ is equal to -1.9 T/K at T_c (Ref. 1), this is consistent with the theoretical expectation (Ref. 4) of an η value of the order of unity.

[15]At $T=75$ K, we found $\eta H_{c2\parallel} = 370 \pm 80$ T. This is more than 10 times its value at 80 K, which is an unphysical result.

[16]The observed torque at $T=75$ falls slightly below the 3D result for angles less than 87°, in apparent conflict with our prediction that the anomalous contribution should always be positive. However, the theoretical curve in Fig. 2 was obtained by force fitting the entire data set, including the high-angle region where the 3D theory is clearly inapplicable. As mentioned in the text, an excellent fit to the 3D expression can still be obtained using all the data below the peak in the torque, giving the same anisotropy parameter as found above 80 K, and also physically reasonable values of $\eta H_{c2\parallel}$. This fit differs only very slightly from the force fit to all the data, but provides a better representation of the 3D result. With respect to this fit, the torque anomaly is positive everywhere.

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[20]To make this prediction, we assumed that τ_{3D} is given at all temperatures by its observed value above ~80 K, namely, $0.17\tau_{max}$.