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Giant torque magnetization anisotropy in $Tl_2Ba_2CaCu_2O_x$ thin films

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We report the first equilibrium torque measurements on a high-T_c superconducting film. The sample is a film of $Tl_2Ba_2CaCu_2O_x$ which exhibits a high degree of c-axis orientation, and the results indicate a giant superconducting effective-mass anisotropy of almost 10^4, and a recent determination from resistive transitions of the upper critical field on a similar film is consistent with this value. We also point out that such torque measurements form a basis for determining $H_{c2}(\theta)$.

Anisotropy in the superconducting properties of the high-T_c oxide superconductors is expected because of their highly anisotropic crystal structure, and a superconducting effective-mass anisotropy of ~ 25 has been reported in[1,2] $YBa_2Cu_3O_7$. In both $Bi_2Sr_2CaCu_2O_x$ and $Tl_2Ba_2CaCu_2O_x$ the double layers of CuO_2 are separated by a greater distance than in $YBa_2Cu_3O_7$, suggesting the possibility of a larger anisotropy. Measurements in $Bi_2Sr_2CaCu_2O_x$ have reported anisotropies from 1 (Ref. 3) to 2 (Refs. 4 and 5) orders of magnitude higher than $YBa_2Cu_3O_7$. For $Tl_2Ba_2CaCu_2O_x$, resistive transitions of the upper critical field H_{c2} in highly c-axis-oriented thin films[6] gave strong support to an even larger anisotropy. However, the actual magnitude was uncertain due to the presence of resistive losses and experimental difficulties in ascertaining the extremely high upper-critical-field slope of ~ 70 T/K. Torque magnetization measurements do not require large fields and are well suited to the low-pinning regime encountered in these materials. Therefore, we have made such measurements on $Tl_2Ba_2CaCu_2O_x$ films, and find a high effective-mass anisotropy of almost 10^4. These measurements emphasize the power of the torque technique[2] by extending it to thin films.

Sputtered films of $Tl_2Ba_2CaCu_2O_x$ were prepared[7] in a three-gun dc magnetron sputtering system which used a turbomolecular pump to provide a typical base pressure of $\sim 3\times10^{-8}$ Torr. The three guns were aimed at a common point above the sources, which provided compositional uniformity to $\pm 1\%$ over a 2-cm^2 substrate area. Targets of Tl, Cu, and a BaCa mixture are simultaneously sputtered in a 20-mTorr argon atmosphere with an oxygen partial pressure of 0.1 mTorr being introduced directly adjacent to the substrates. A load-lock mechanism permitted changing samples without breaking vacuum in the growth chamber: This feature reduced any contamination of the target surfaces and was crucial to maintaining reproducible conditions in successive runs. These films were deposited onto (100)-oriented, single-crystal ZrO_2-9%Y_2O_3 substrates, which were kept at ambient temperature during the deposition. Films of thickness 0.3 μm were annealed in a closed Au crucible, which was placed in a flowing-oxygen tube furnace at 870 °C for about 5–30 min. It was found necessary to include a small pellet of target material in the Au crucible in order to adequately reduce Tl volatilization. The film composition was confirmed by energy-dispersive x rays. X-ray diffraction indicates a very high degree of orientation with the c axis perpendicular to the substrate, but *in the plane* of our $Tl_2Ba_2CaCu_2O_x$ films the grains are not epitaxial to the substrate.

Detailed rocking-curve analysis revealed two slightly misaligned regions. In one region, the c axis is perpendicular to the substrate with the rocking curve showing a half-width at half maximum (HWHM) of $\sim 0.3°$. The c axis of the entire other region, which exhibits a slightly broader rocking curve (HWHM of $\sim 0.5°$), is tilted at an angle of $\sim 1.15°$ with respect to the first, but both c axes lay in a single plane that is perpendicular to the substrate. The most crucial data are obtained with the field approximately parallel to the CuO planes, so for the torque measurements, the sample was oriented inside a conventional rotatable electromagnet such that the magnetic field was simultaneously in these planes *for both regions.* As a result, the misalignment of these regions will only weakly affect the measured anisotropy.

The application of torque magnetometry to superconducting anisotropy measurements is described in detail in Refs. 2 and 4. The torque was measured as a function of the angle θ between the field and c axis, and in the present case, the substrate exhibited a θ-dependent contribution that was comparable to that of the film. However, since the substrate background was temperature independent (between 110 and 140 K) and could be accurately determined, we just subtracted it to obtain the torque $\tau(\theta)$ of the film. Typical values of $\tau(\theta)$, normalized to its maximum value τ_{max}, are shown in Fig. 1 for a $Tl_2Ba_2CaCu_2O_x$ film in a field of 10 kOe between 90 and 98 K. For the fields and temperatures reported in this paper, any irreversibility was below our instrumental resolution, demonstrating *also* the absence of significant flux pinning.

The transition temperature was estimated from the torque measurements shown in Fig. 2 for $\theta = 80°$ and $H = 10$ kOe. Extrapolating the linear dependence of τ on T between 90 and 98 K results in $T_c \sim 100$ K. Note that

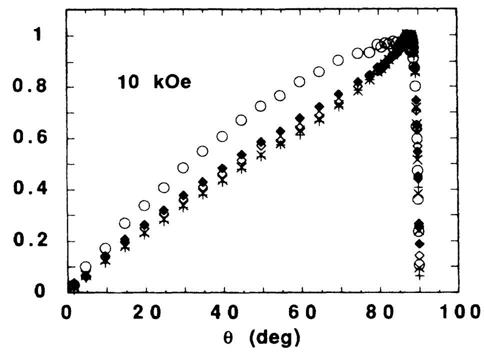

FIG. 1. The normalized torque measured at various temperatures near T_c in a 10-kOe field: ○, 98 K; ◆, 96 K; ◇, 94 K; ×, 92 K; and +, 90 K.

such measurements can form a basis for determining[8] $H_{c2}(\theta)$, and although the symmetry directions at 0° and 90° are unattainable because $\tau(\theta)$ vanishes, from a practical point of view reasonable extrapolations can be made. The data for this $Tl_2Ba_2CaCu_2O_x$ film show a tail which persists up to 103–108 K. This tail is likely due to both fluctuations and sample inhomogeneities (the Tl-Ba-Ca-Cu-O system has a number of superconducting phases). In a similar manner, measurements of conductivity fluctuations above T_c in equivalent films[9] showed excellent agreement with the two-dimensional theory over a temperature range from 240 down to 2 K above T_c. Thus ± 2 K may be close to the limit of sample homogeneity for these films.

The theoretical treatment[10] of the torque is based on the London equations, being valid for weak pinning and for H between, but not too close to, the lower and upper critical fields; i.e., $H_{c1} \ll H \ll H_{c2}$. Both of these conditions are satisfied for the measurements reported here (except perhaps at the highest T), and the torque is given by

$$\tau(\theta) = \frac{\phi_0 HV}{64\pi^2\lambda^2}\frac{\gamma^2-1}{\gamma^{2/3}}\frac{\sin 2\theta}{\epsilon(\theta)}\ln\frac{\gamma\eta H_{c2\parallel}}{H\epsilon(\theta)}, \tag{1}$$

where $\epsilon(\theta) = (\sin^2\theta + \gamma^2\cos^2\theta)^{1/2}$, ϕ_0 is the flux quantum, V is the sample volume, λ the average penetration depth, $H_{c2\parallel}$ is the upper critical field along the c axis, η is a constant expected to be of order unity,[10] and γ is the square root of the effective-mass anisotropy.

As T increases towards $T_c \sim 100$ K, Fig. 1 shows a progressive change in the character of $\tau(\theta)$ below $\theta \sim 80°$ which is opposite to the expectations of Eq. (1), using reasonable estimates of $H_{c2\parallel}(T)$, but rather reflects a similar tendency found in both[11] $Bi_2Sr_2CaCu_2O_x$ and $YBa_2Cu_3O_7$ single crystals. When T further increases above T_c, this trend continues and all three materials approach a simple $\sin(2\theta)$ variation, which is the behavior associated with a *linear*, anisotropic susceptibility. Because of the systematics, it is possible that the $\sin(2\theta)$ deviations result from intrinsic fluctuations, an effect which has not been incorporated into the theoretical treatment of the torque. In any case, to minimize this effect, we fit the data by Eq. (1) at the lowest temperature (the onset of pinning precluded measurements below 90 K) and the highest available field of 10 kOe. The results, found by minimizing the rms deviation between the experimental data and Eq. (1), are shown in Fig. 3 and indicate excellent agreement with theory with a 2% rms deviation. The anisotropy parameter γ is 94 and a value of 24 is determined for η if $H_{c2\parallel}$ is determined from an upper-critical-field slope of 1 T/K found in the resistive measurements.[6] The extremely high anisotropy found here implies that the effect of finite rocking-curve width cannot be ignored, and Eq. (1) was averaged over a Gaussian distribution (HWHM$\sim$0.3°) of c-axis orientations. Using a HWHM $\sim$0.5° resulted in a poorer fit and $\gamma \sim 130$.

All the data shown in Fig. 1, together with data at the same temperatures taken in a field of 6 kOe, were compared to the prediction of Eq. (1) using $\eta = 24$ and the linear upper-critical-field slope of 1 T/K for $H_{c2\parallel}$. These are displayed as difference plots in Fig. 4 and reinforce the tendency to approach the $\sin(2\theta)$ dependence as T approaches T_c. It is comforting to note, however, that for the angles very near 90°, which are most sensitive to the anisotropy, there are no essential differences in $\tau(\theta)$ seen

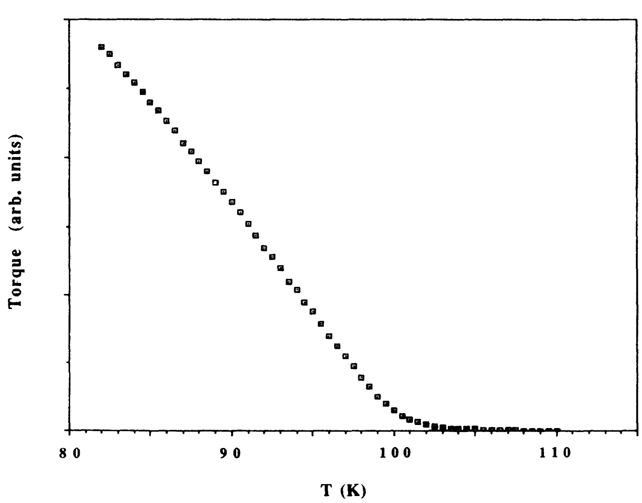

FIG. 2. The torque measured at $\theta = 80°$ in a field of 10 kOe as a function of T.

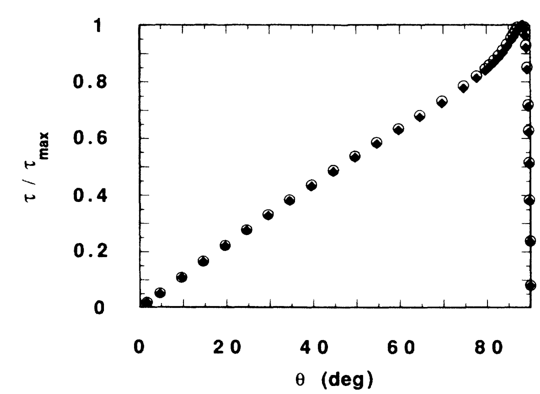

FIG. 3. The normalized torque measured at 90 K in a 10-kOe field at various angles with respect to the c axis of a $Tl_2Ba_2CaCu_2O_x$ film (◆). The predictions of Eq. (1) for $\gamma = 94$, $\eta H_{c2\parallel}/H = 240$, and a Gaussian distribution (HWHM = 0.3°) of c-axis orientations are shown as ○.

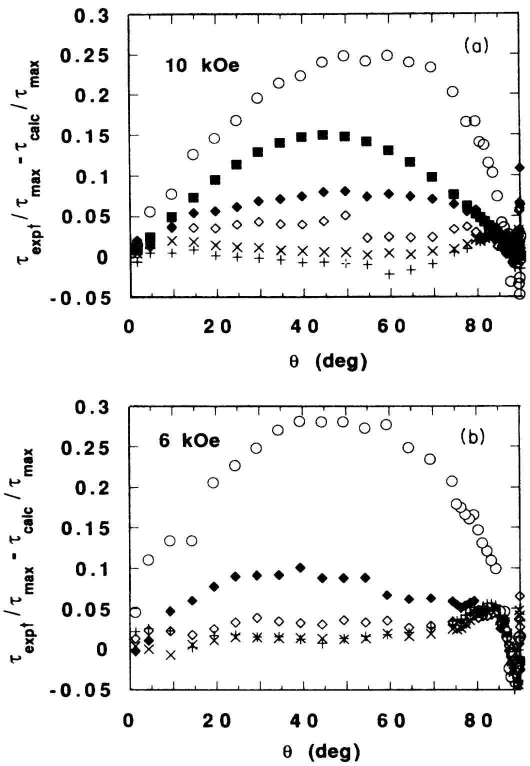

FIG. 4. The differences between the experimental and calculated normalized torques: (a) 10-kOe field: ○, 98 K; ◆, 96 K; ◇, 94 K; ×, 92 K; +, 90 K. Also shown as ■ is the function $0.15\sin(2\theta)$. (b) 6-kOe field: ○, 98 K; ◆, 96 K; ◇, 94 K; ×, 92 K; and +, 90 K.

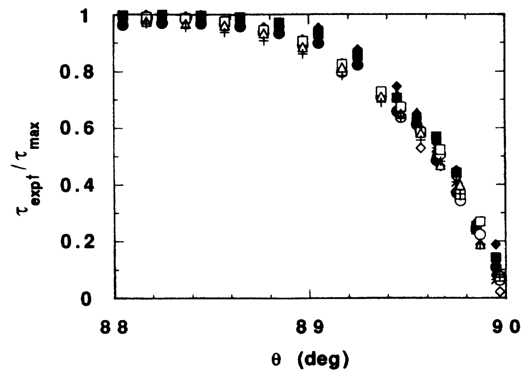

FIG. 5. The normalized torque for θ close to 90° for various temperatures near T_c in a 10-kOe field: ●, 98 K; ◆, 96 K; ■, 94 K; ▲, 92 K; and ✱, 90 K. In a 6-kOe field: ○, 98 K; ◇, 96 K; □, 94 K; △, 92 K; and +, 90 K.

in Fig. 5 for T between 90 and 98 K and at both field values. The data exhibit an average value of the angular half width, $\theta_h = 0.35 \pm 0.05°$, where θ_h is the angle measured from 90° for which $\tau(\theta)/\tau_{max}$ is $\frac{1}{2}$. The lack of T and very small H dependence is expected from Eq. (1).

In summary, we have performed the first equilibrium torque measurements on a high-T_c superconducting film and find for $Tl_2Ba_2CaCu_2O_x$ an anisotropy parameter of ~ 94, in reasonable agreement with estimates from resistive transitions on similar films.[6] This implies an effective-mass anisotropy of almost 10^4. In addition, the power of the torque technique is further emphasized by extending it for the first time to thin films. Finally, we point out that torque measurements can form a basis for determining $H_{c2}(\theta)$.

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