Experimental Evidence for a Transverse Magnetization of the Abrikosov Lattice in Anisotropic Superconductors

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The torque on a superconductor in a magnetic field H has been thought to be dominated by trapped flux or sample shape effects, but it has recently been suggested that an anisotropic type-II material should experience an *intrinsic* torque for $H_{c1} \ll H < H_{c2}$. The predicted phenomenon results from a transverse magnetization of the Abrikosov lattice. Measurements are presented on copper-oxide superconductors which delineate the experimental regime in which extrinsic effects are negligible and confirm the existence of the predicted intrinsic torque.

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On the basis of London electrodynamics, one of us has recently suggested[1] that an intrinsic torque should be observed for an anisotropic type-II superconducting material in a field H of intermediate strength, i.e., for $H_{c1} \ll H < H_{c2}$. Its physical origin lies in a modification to the Abrikosov vortex structure that occurs in anisotropic materials. In such materials the normal to the plane of the current loops around the vortex core has a tendency to lie in a specific crystallographic direction.[2] For example, in the copper-oxide superconductors this is along the $\hat{\mathbf{c}}$ axis; i.e., the loops themselves prefer to lie in the "easy" Cu-O plane. In general, the magnetic moment of each vortex and the macroscopic magnetization then acquire a nonzero component normal to the magnetic induction.[3] The anisotropy torque is just the vector product of this transverse magnetization and the field. With the geometry shown in the inset to Fig. 1(a), the result[1,4] for the angular dependence of the torque is

$$T(\theta) = CH \ln\left(\frac{\gamma\eta H_{c2\parallel}}{H(\sin^2\theta + \gamma^2\cos^2\theta)^{1/2}}\right)\frac{\sin 2\theta}{(\sin^2\theta+\gamma^2\cos^2\theta)^{1/2}}. \quad (1)$$

In this expression, η is a constant of order unity and $\gamma = (m_3/m_1)^{1/2}$, where m_3 and m_1 are the components of the effective mass tensor along and perpendicular to $\hat{\mathbf{c}}$, respectively. The upper critical fields in these same two directions are $H_{c2\parallel}$ and $H_{c2\perp}$, and $\gamma = H_{c2\perp}/H_{c2\parallel}$. The parameter C is proportional to $(m_3 - m_1)/\lambda^2(T)$ and depends on temperature via the (average) penetration depth, λ. For $\gamma = 1$, $m_3 = m_1$, and so the torque vanishes. For a weak anisotropy, $\gamma \sim 1$, the angular dependence reduces to $\sin 2\theta$ and the torque is a maximum at $\theta = 45°$. For $\gamma > 1$, there is still a single maximum, but it now occurs at an angle $\theta_m > 45°$. The predicted torque is an intrinsic feature of uniaxial superconductors in a magnetic field and has no connection with the normally dominant shape effects or flux pinning.

Although the theory discussed above applies to any anisotropic type-II superconductor, there was a strong motivation to choose copper-oxide materials for an experimental test. Despite the attention these have received, there is a clear need for a reliable probe of their anisotropy in the superconducting state. Conventional measurements of the two physical quantities of principal interest (the energy gap and critical field) have proved to be difficult to interpret. The values reported for the gap scatter vary widely.[5] Most of the upper-critical-field values reported so far are based on resistive measurements. These also give widely scattered results and it has recently even been argued[6] that the method does not actually measure H_{c2}. (Critical-current anisotropies do exhibit some consistency between measurements[7,8] but, as in all known type-II materials, the critical current is a defect-controlled property which does not directly reflect fundamental anisotropies.) As well as confirming the existence of a transverse magnetization, this Letter also demonstrates that torque magnetometry can provide a

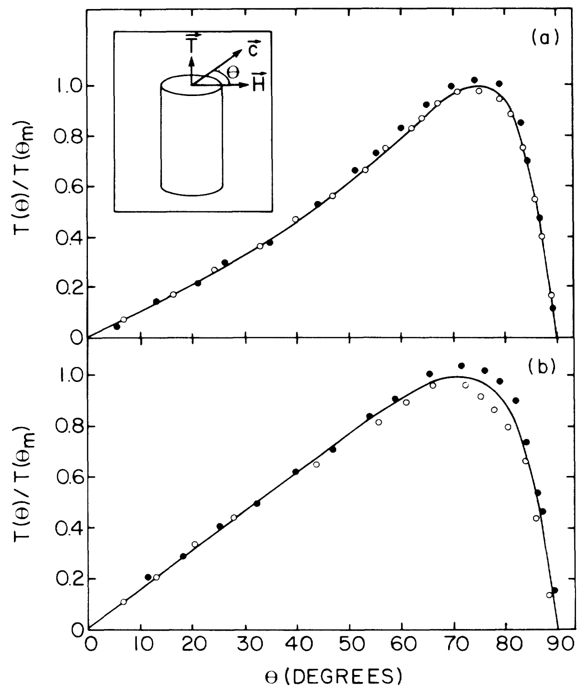

FIG. 1. Normalized torque as a function of angle (defined in the inset) for sample I at $T=84.5$ K with (a) $H=6$ T and (b) $H=1$ T. Open circles represent data taken with the angle increasing, and closed circles with angle decreasing. *Both* (theoretical) lines were obtained from Eq. (1) with the parameters $\gamma=5.1$ and $\eta H_{c2\parallel}=15$ T.

quantitative measure of anisotropy. Data are presented on two members of the copper-oxide family, $Y_1Ba_2Cu_3O_{7-\delta}$ and $Tl_2Ba_2Ca_2Cu_3O_{10}$, denoted hereafter by 1:2:3 and 2:2:2:3, respectively.

Grain-aligned samples were used in our work and prepared in the manner described previously.[8] Sample characteristics are noted in Table I. The resistive T_c values were obtained on the material in sintered form prior to grinding it into small grains. These grains were mixed in epoxy which was then cured in a 9.4-T magnetic field at room temperature. This procedure produces uniaxial grain alignment with the $\hat{c}$ axes lying along the field direction. The angular widths of the x-ray rocking curves recorded in the table indicate excellent alignment. (There is no alignment in a perpendicular direction, so any *a-b* anisotropy in the 1:2:3 samples will be averaged out.) Scanning-electron-microscope examination showed that the grains had a large spread in size, from about 2 to 20 μm. A small percentage had flat faces aligned perpendicular to the $\hat{c}$ direction, but the majority were irregular in shape. All samples were single phase, as judged by x-ray diffraction data, though the transition temperature of the 2:2:2:3 material does suggest the presence of some 2:2:1:2 intergrowths.[9] Sample shape was cylindrical and the magnetic field was applied perpendicular to the cylinder axis. Torques were measured in a magnetometer designed and built by one of us.[10] The sensitivity is ~1 dyn cm, the temperature stability is ~0.01 K, and magnetic fields up to 6 T can be applied to the sample with a rotatable superconducting Helmholtz pair.

Figure 1(a) displays normalized torque data obtained on sample I in a field of 6 T and at a temperature of 84.5 K. Open circles represent data taken with the angle increasing, and closed circles with angle decreasing. Note the negligible hysteresis except for a very small region near the peak. The full curve is the normalized torque calculated directly from Eq. (1) using the parameters $\gamma=5.1$ and $\eta H_{c2\parallel}=15$ T. The fitting uncertainty is estimated to be ± 0.3 for γ and ± 3 T for $\eta H_{c2\parallel}$. [Literature values for γ [11-13] scatter from about 3 to 9. Conventional determinations[7] of $H_{c2\parallel}$ (84.5 K), corrected for T_c differences, have an even larger scatter, between roughly 1 and 8 T. There is no reliable theoretical estimate for η at present, but it is thought[1] to be a constant of order unity.] Figure 1(b) presents torque data obtained on the same sample, at the same temperature, but in a field of 1 T. Again, they are essentially reversible over most of the range. Equation (1) now has no free parameters and the 1-T data have a markedly different angular character from those obtained at 6 T. Nonetheless, Eq. (1) with

TABLE I. Properties of the epoxy-encapsulated grain-aligned samples. The volume refers to that of the superconductor and the packing fraction is the percentage of the sample (by volume) occupied by the superconductor. The FWHM is the full angular width at half maximum of x-ray rocking curves, obtained using the (006) and (0014) reflections for the 1:2:3 and 2:2:2:3 samples, respectively. $T(\theta_m)$ values are for the 6-T data shown in Figs. 1(a), 2(b), and 3.

Sample	Material	$T_c(R=0)$ (K)	Volume (10^{-3} cm^3)	Packing fraction (%)	FWHM	$T(\theta_m)$ (dyn cm)
I	1:2:3	91.0	18.8	29	2.4	210
II	1:2:3	89.9	16.4	15	2.3	470
III	2:2:2:3	117	7.0	8	2.3	430

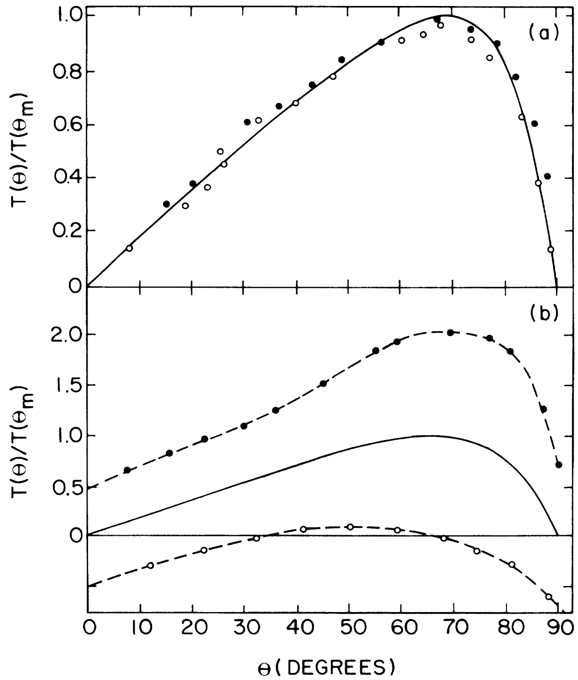

FIG. 2. Normalized torque observed for sample II with $H=6$ T and (a) $T=70$ K and (b) $T=40$ K. Open circles represent data points obtained with the angle increasing, and closed circles with angle decreasing. The full lines are theoretical fits by Eq. (1), with $\gamma=4.0$ in each case and with $\eta H_{c2\parallel}=75$ T and 140 T for (a) and (b), respectively. The dashed lines through the data in (b) are to guide the eye.

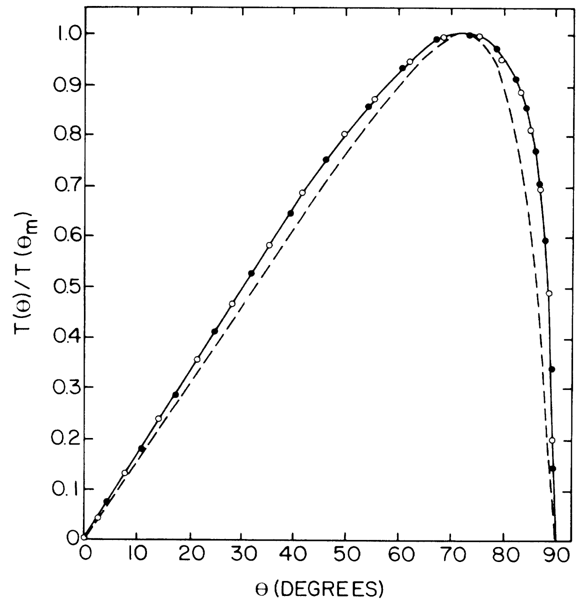

FIG. 3. Normalized torque observed for sample III at $T=80.3$ K in a field of 6 T. Open circles represent data points obtained with the angle increasing, and closed circles with angle decreasing. The full line is drawn through the points as a guide to the eye, while the dashed line is the prediction of Eq. (1) with $\gamma=5.1$ and $\eta H_{c2\parallel}=70$ T.

$H=1$ T gives the full line which provides an excellent fit.

Sample II was made from a different batch of 1:2:3 powder with a slightly lower T_c. Figure 2(a) shows the torque data at a temperature of 70 K and $H=6$ T, and the theoretical fit, which gives a somewhat smaller anisotropy parameter, $\gamma=4.0$. With γ and $\eta H_{c2\parallel}$ fixed, the only other unknown parameter that enters the full theoretical expression[1] for $T(\theta_m)$ is the average penetration depth. In this way we obtained estimates for $\lambda(0)$ in the range of a few tenths of a micron for both samples, in accord with most literature estimates. Thus, the angular dependence and magnitude of the observed torque argue strongly for its intrinsic origin.

We have carefully checked the contribution of both flux pinning and shape effects. The flux pinning contribution (hysteresis) was observed to increase sharply as the temperature was reduced. For conventional hysteretic magnetization measurements, the equilibrium magnetization can still be estimated as the mean of field-increasing and field-decreasing data.[14] The transverse magnetization displays analogous behavior. For example, Fig. 2(b) shows the angular dependence of the torque for sample II at $H=6$ T and at a temperature of 40 K. The flux-pinning and intrinsic contributions are now comparable but the mean torque is still given by Eq. (1) using the same value of γ (=4.0) as found for this sample at higher temperatures. In fact, the mean torque lies too close to its theoretical fit to be shown as a separate line in Fig. 2(b). For the 2:2:2:3 sample (III), torque data have been obtained down to 80 K *that are completely reversible,* to within our measurement uncertainty. In this case (Fig. 3), the data show some small systematic deviations from Eq. (1). Nonetheless, these 2:2:2:3 results clearly establish that a completely pinning-free torque exists with an angular dependence similar to that observed in the 1:2:3 samples. We conclude that a useful temperature window exists for both materials where flux-pinning effects are small and that results can be obtained outside the window by use of the conventional experimental *Ansatz* for dealing with pinning.

A conventional shape effect cannot exist in the sample geometry shown in Fig. 1 and the shape effect due to cylinder-axis misalignment is negligible.[15] Nonetheless, there is the possibility of a more subtle effect because the grains in our samples are relatively well separated[16] and may have some shape asymmetry linked to their crystallographic orientation. The existing theoretical treatment[17] of shape effects in the superconducting mixed state is only valid close to H_{c2}. We have therefore developed[15] a treatment, again based on London electrodynamics, which avoids this restriction. In the limiting case of a single plate of vanishing thickness with the field applied at some angle θ to the normal to the plate, this gives for T_s, the shape effect torque in dyn cm,

$$T_s \simeq 6\times 10^4 \bar{H}_{c1}^2 V \left[\ln \left(\frac{\eta \bar{H}_{c2}}{H} \right) \right]^2 \sin 2\theta , \qquad (2)$$

where $\bar{H}_{c2}$ and $\bar{H}_{c1}$ are the critical fields in tesla, assumed isotropic for the purposes of this estimate, and V is the plate volume in cm^3. For a sample consisting of well separated parallel platelets, an upper bound for the shape torque can be obtained from Eq. (2) with V identified as the total platelet volume. For $\bar{H}_{c2}$ we took the mean of $H_{c2\perp}$ and $H_{c2\parallel}$, i.e., $\eta\bar{H}_{c2}=(\gamma+1)\eta H_{c2\parallel}/2$, and used γ and $H_{c2\parallel}$ values obtained in this work. Similarly, for $\bar{H}_{c1}$ we took the mean of single-crystal data[13] for $H_{c1\perp}$ and $H_{c1\parallel}$. (Our estimates are therefore restricted at present to the 1:2:3 material, but 2:2:2:3 numbers should be comparable.) For example, in the case of sample I at $T=84.5$ K, $\eta\bar{H}_{c2}\sim 45$ T and $\bar{H}_{c1}\sim 4\times 10^{-3}$ T. For $H=6$ T we find $T_s\sim 0.1$ dyn cm, compared with the observed $T(\theta_m)=210$ dyn cm. The largest T_s associated with data reported here is for sample II at $T=40$ K. In this case, $T_s\sim 15$ dyn cm, compared with the observed $T(\theta_m)=470$ dyn cm. Although our scanning-electron-microscopy observations clearly ruled out the extreme platelet geometry, samples with such a geometry may be used in future single-crystal experiments. The above estimates show that the shape torque will still generally be negligible.

We have also looked for a shape effect experimentally[15] using a sample in the form of a flat rectangular plate containing 13×10^{-3} cm^3 of *unaligned* 1:2:3 powder. For such a sample *only* a conventional macroscopic shape effect and pinning torque can be present, since both the intrinsic and grain shape torques will average out. Small torques of a few dyn cm were observed at a temperature of 84 K and $H=1$ and 6 T, but they reversed sign on reversing the sense of angular change and therefore were clearly identified with pinning: Finally, we point out that the physical reason that the shape effect is negligible in our measurements is that the magnetization is much smaller than the applied field. In a type-II superconductor such as niobium, with $H_{c2}\sim 2H_{c1}$, this situation can only be realized very close to H_{c2}. For the extreme type-II materials used here, it is realized in a broad range of field, namely, $H_{c1} \ll H < H_{c2}$.

There are good reasons for hoping that torque magnetometry will be relatively free of the ambiguities that have plagued conventional measures of anisotropy in high-T_c superconductors. The transverse magnetization is the only physical property in the superconducting state whose actual existence is a direct result of anisotropy. We have shown that flux pinning and shape effects are negligible over a significant experimental range and that the agreement with theory is good. In our judgment, torque magnetometry promises to provide a clean measure of anisotropy in high-T_c superconductors. However, the chief significance of our work is that it provides evidence for a transverse magnetization, confirming that the mixed state of anisotropic (including high-T_c) superconductors is substantially different from that in the isotropic case. This physical difference is likely to be of importance for a number of areas, e.g., vortex-vortex interaction, flux lattice structures, and flux dynamics.

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