Magnetic anisotropy of high- T_c superconductors

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The magnetic anisotropies of Bi₂Sr₂CaCu₂O_y and YBa₂Cu₃O_{7-x} single crystals have been investigated by simultaneously measuring the longitudinal (M_L) and transverse (M_T) components of the equilibrium magnetization of crystals oriented at arbitrary angles with respect to the applied field direction. The variations of M_L and M_T as a function of orientation, field, and temperature have been measured. In the regime where the simple three-dimensional anisotropic London theory is valid, it is shown that the measurement of M_T/M_L with an applied field in the range $H_{c1} \ll H \ll H_{c2}$ yields the anisotropic mass ratio (m_3/m_1) directly. The possible breakdown of the simple theory is discussed.

I. INTRODUCTION

Many recent investigations have probed the anisotropic properties of the high-temperature copper oxide superconductors. Measurements of the upper critical field H_{c2} parallel and perpendicular to the basal plane indicate large anisotropy in the value of the Ginzburg-Landau (GL) coherence length in those directions.¹ A transverse magnetization in the mixed state has been observed.² Measurements of other properties including the lower critical field, H_{c1} ,³ and the resistivity⁴ also show anisotropy. Most of these anistropic properties are very likely a consequence of the crystalline anisotropy of these compounds. Interestingly, measurements of fluctuation conductivity^{4,5} demonstrate the existence of two-dimensional (2D) fluctuations which are associated with weakly coupled 2D superconducting layers. Indeed, the high- T_c copper oxides are layered perovskites, and the superconductivity has been speculated to be layerlike in the sense that it nucleates in the copper oxide planes which are in turn coupled by the Josephson effect.⁶ If such a description were accurate, then earlier work on layered superconductors could be applied to the high- T_c systems.

A different source of anisotropy may also have to be considered. If the Cooper pairs of high- T_c superconductors involve states of nonzero orbital angular momentum, then there can be anisotropy present which is not directly related to the crystalline symmetry.⁷ However, such unconventional pairing has not been demonstrated in the high- T_c systems, so that only models which assume that this type of anisotropy is absent will be considered here, i.e., those in which s-wave pairing is present exclusively.

The crossover between the anisotropic threedimensional (3D) regime and the quasi-2D regime of weakly coupled layers may be characterized by the dimensionless parameter $\epsilon = 2[\xi_z(T)/s]^2$. Here $\xi_z(T)$ is the GL coherence length perpendicular to the layers and s is the interlayer spacing.⁸⁻¹⁰ In the regime $\epsilon \gg 1$, the layers are strongly coupled, and the system is essentially an anisotropic 3D superconductor. Ginzburg-Landau or London theories may be generalized to describe such systems by replacing the effective mass of the isotropic case with an effective mass tensor.^{11,12} In the opposite limit, $\epsilon < 1$, the layers are weakly coupled, and the system can exhibit 2D character. In this regime the system is described by differential-difference equations instead of the anisotropic GL differential equations, resulting in rather different behavior.¹³ The order parameter along z (the direction normal to the layers) is very nonuniform in this case. It is important to note that the parameter ϵ is only a measure of the relative coupling strength, i.e., the strength of the Josephson energy that couples layers relative to the condensation energy in the layer. If the thermal energy kT were comparable to the magnitude of the Josephson coupling energy, then different physical properties would enter the problem. The layers could become phase decoupled and 2D fluctuations would become evident. Indeed, there is evidence for a 2D topological phase transition, or Kosterlitz-Thouless transition, in the high- T_c superconductors.¹⁴ Keeping this in mind, one must remain well outside of the fluctuation regime to apply the arguments involving ϵ .¹³

It should be noted that a discussion concerning the strong or weak coupling of layers has also been given for the case of granular superconductors.⁹ A layered superconductor is in some sense just a one-dimensional realization of a more general granular system. In a limit in which the variation of the local internal magnetic field is small over distances of the order of the layer thickness (grain size), the granular theories give additional insight into many concepts in both the strong and weak coupling cases.

Extensive experimental and theoretical studies have been carried out on layered superconducting systems, and as mentioned above, many earlier results should be relevant to the new oxide superconductors. Previous experimental work involved studies on Nb/Ge, Nb/Cu, and V/Ag superlattices in which superconducting layers were coupled by either the Josephson effect or the proximity effect.¹⁵ The coupling could be controlled by varying the thickness of the nonsuperconducting material resulting in different values of ϵ . In the high- T_c superconductors the coupling between layers cannot be continuously tuned this way because the crystal structure is fixed. However, the different classes of oxides are likely to have dissimilar coupling strengths due to the differing distances between their copper oxide layers. Also, as in the case of the superlattices, another way to vary ϵ is by changing the temperature. This can be achieved as a consequence of the divergence of the GL coherence length, $\xi_z(T)$, as T approaches T_c . In the high- T_c systems, the value of $\xi_z(0)$ is smaller than the unit-cell size in the c direction. Thus a crossover from quasi-2D to 3D may be observable as the temperature is increased towards T_c . This type of crossover has been observed in multilayers of metallic superconductors where measurements of the angular and temperature dependence of the upper critical field $H_{c2}(\theta, T)$ were used to determine the regime the system was in. There is also evidence from similar measurements that the $Bi_2Sr_2CaCu_2O_{\nu}$ system may be in the $\epsilon < 1$ regime for most temperatures while the $YBa_2Cu_3O_{7-x}$ compounds are in the opposite limit.¹⁶ Here these issues will be investigated further by studying the anisotropy through measurements of the angle, temperature, and field dependent magnetization, $\mathbf{M}(\theta, T, H)$.

II. THEORETICAL BACKGROUND

The behavior of a strongly coupled system will be considered first. The effective mass formulations of the GL (Ref. 11) and London (Ref. 12) theories can be used to calculate thermodynamic parameters such as H_{c1} , H_{c2} , and the vortex line energy as angular dependent quantities. Also for the 3D anisotropic superconductor, both theories predict the existence of a transverse magnetization (perpendicular to B) in addition to the usual longitudinal magnetization (along B) in the mixed state. This is true for the field applied in any direction except along the principle axes. It is also in contrast with the case of an isotropic superconductor in which a magnetization is found only along the direction of the field, neglecting possible shape effects. The physical origin of the transverse magnetization of an anisotropic superconductor can be seen in the following argument: a consequence of the anisotropic effective mass tensor is that currents will have both "easy" and "hard" directions. The "easy" direction is that in which the effective mass is a minimum. Consequently, the kinetic part of GL free energy is a minimum when the loops of current flow in the "easy" direction. Hence, to satisfy energy considerations, the currents associated with a vortex will not in general flow in a plane which is orthogonal to the vortex direction. In the case of high- T_c systems, the currents will tend to flow in the CuO planes. For a vortex oriented at angle θ relative to z, the generalized GL and London theories^{11,12} predict that the currents will flow in a plane which is inclined from the basal plane by an angle α such that

$$\tan\alpha = \frac{m_1}{m_3} \tan\theta , \qquad (1)$$

where m_1 and m_3 are the in- and out-of-plane principle values of the effective mass tensor, respectively. Therefore, if a field is applied at some angle which is not either 0° or 90°, the currents will flow in a plane which is close to the basal plane and the magnetic moments associated with these canted current loops will produce the transverse magnetization. For a system with large anisotropy the tendency for currents to stick close to the basal plane will increase and a larger relative transverse magnetization will be evident.

Kogan has used 3D anisotropic London theory to predict the magnitude of both the longitudinal and transverse components of magnetization for a uniaxial superconductor $(m_1 = m_2 \neq m_3)$:¹⁷

$$M_{L}(\theta) = -\frac{\phi_{0}}{32\pi^{2}\lambda(T)^{2}} \ln \frac{(H_{c}\beta)_{\theta}}{H} \times \sqrt{m_{1}\sin^{2}\theta + m_{3}\cos^{2}\theta}, \qquad (2)$$

$$M_{T}(\theta) = \frac{\phi_{0}}{32\pi^{2}\lambda(T)^{2}} \ln \frac{(H_{c2}\beta)_{\theta}}{H} \times \frac{m_{3} - m_{1}}{\sqrt{m_{1}\sin^{2}\theta + m_{3}\cos^{2}\theta}} \sin\theta\cos\theta .$$
(3)

Here again, θ is the angle between the vortex direction, i.e., the direction of \mathbf{B} , and the direction of the c axis. The geometric average penetration depth is $\lambda(T)$, ϕ_0 is the superconducting flux quantum $(2 \times 10^{-7} \text{ G cm}^2)$, and $H_{c2}\beta$ is the angular dependent upper critical field times a factor which is the result of an integral cutoff in the theory. It should be noted that in an experiment the angle that is measured is not θ (the angle between **B** and the c axis) but rather θ_a (the angle between the applied-field direction, \mathbf{H}_a , and the c axis). The difference between θ and θ_a is of order M/H, which for large fields $(H \gg H_{c1})$ is small and will be ignored in the treatment of the experimental data. However, for the case of large anisotropy, it is possible that the difference between θ and θ_a may become nonnegligible. It is in principle possible to calculate θ if one knows θ_a , the magnitudes of both magnetization components, and the extent of the demagnetization effects. Work addressing this issue is still in progress.

Torque magnetometry studies of both YBa₂Cu₃O_{7-x} and Bi₂Sr₂CaCu₂O_y samples have been used to determine the transverse magnetization and extract an anisotropic mass ratio through the use of Eq. (3).² The data is very clean but the fit depends not only on m_3/m_1 , but also on $H_{c2}\beta/H$ which is not determined independently. In addition, investigation of the temperature dependence of m_3/m_1 using torque magnetometry requires a detailed knowledge of the temperature dependence of λ and $H_{c2}\beta$. A very important feature of Eqs. (2) and (3) is that simultaneous measurements of M_L and M_T provide a way to determine m_3/m_1 directly without the use of any additional fitting parameters. The ratio of transverse to longitudinal magnetization

$$\frac{M_T}{M_L} = \left(\frac{m_3}{m_1} - 1\right) \frac{\sin\theta\cos\theta}{\sin^2\theta + (m_3/m_1)\cos^2\theta} \tag{4}$$

is a function of only m_3/m_1 and θ . The quantities with either unknown or undetermined temperature dependences such as λ and $H_{c2}\beta$ have cancelled out. The above relation can also be shown to be valid in the regime $H < H_{c2}$ by using GL theory.¹¹ Therefore, when the 3D anisotropic effective mass formulation is applicable, one may measure M_T/M_L as a function of θ and T to determine m_3/m_1 directly, as well as study its temperature dependence.

For the quasi-2D system of weakly coupled superconducting planes, qualitatively different superconducting properties are expected. For example, $H_{c2}(\theta)$ exhibits a cusp at $\theta = 90^{\circ}$ (field parallel to the layers) (Ref. 15) while for the 3D anisotropic system it is continuous. The cusp reflects the 2D character in the sense that H_{c2} is controlled by the thickness of the superconducting layer and not by the effective medium as a whole. At angles some distance away from $\theta = 90^{\circ}$ the behavior of the upper critical field for the quasi-2D system is the same as that of a 3D anisotropic system. Therefore, it is apparent that the search for quasi-2D behavior should be conducted at angles which are near $\theta = 90^{\circ}$.

As mentioned before, quasi-2D behavior will only occur in a certain temperature regime. The reduced temperature demarking the crossover point, $t^* = T^*/T_c$, is determined by the condition $\xi_z(t^*) = s/\sqrt{2}$.¹³ For $t > t^*$ the in-plane upper critical field is governed by the orbital-pair breaking effect just as in ordinary 3D superconductors while for $t < t^*$ this effect vanishes. Consequently, the slope of $H_{c2}(T)$ for a field applied in-plane increases drastically as t passes through t^* from above. The disappearance of the orbital-pair breaking effect can be understood by realizing that for $t < t^*$ the coherence length, which is the diameter of the normal vortex core, is less than the spacing between layers, and therefore the core can reside between layers without disturbing the order parameter in the layers very much. It should be recognized that a coupled layered system will always pass through a 3D regime near T_c because of the divergent coherence length. In fact, upward curvature in $H_{c2}(T)$ observed in many systems such as superlattices, granular superconductors, and high- T_c cuprates may have this crossover as its cause.

In the case of Bi₂Sr₂CaCu₂O_y, the distance between the CuO planes is ≈ 15 Å (Ref. 18) and the extrapolated zero-temperature coherence length in the *c*-axis direction is ≈ 1 Å.¹⁶ From the previous argument, the system should be in the quasi-2D limit for t < 0.99(T < 84 K)—in other words, for nearly all temperatures. In the case of YBa₂Cu₃O_{7-x}, the spacing is ≈ 12 Å (Ref. 19) and $\xi_z(0)$ is ≈ 3 Å,¹ so it too should show 2D character but for t < 0.88 (T < 79 K).

The previous discussion compares $H_{c2}(T,\theta)$ for strong

and weak interlayer coupling and indicates where one might expect to observe quasi-2D behavior. However, the issue remains as to what changes might be observed in the magnetization of a quasi-2D system compared to that of a bulk anisotropic 3D system. In the quasi-2D limit, "hard" and "easy" directions can still be defined, and there still must exist a transverse magnetization. However, for this case the system has pronounced Josephson character in the z direction. A vortex lying between the layers is essentially a Josephson vortex rather than an Abrikosov vortex. The former has no real normal core, whereas the latter has a normal core and an associated core energy. For a vortex oriented at an angle to the layer, the core will have a tendency to lie in between the layers as much as possible since this will save condensation energy in the layer. Therefore, the assumption that the vortex line is straight (as in the case of the effective mass theory) may break down. However, for $\kappa \gg 1$ materials (such as the high- T_c materials), the condensation energy lost by the creation of a vortex is a small portion of the overall balance compared to the field and kinetic parts.²⁰ [κ is the GL parameter, which is defined as $\kappa = \lambda(T) / \xi(T)$.] A theory constructed to determine the structure of a vortex oriented at an arbitrary angle would do so by minimizing all contributions to the energy. It is possible that when this is done the vortex line may have kinks on a semimicroscopic scale.²¹ The actual form for the components of magnetization in this case may differ from that predicted by the effective mass London theory. However, for large κ materials, it would seem that the form would be similar since the core correction is usually negligible compared to the overall line energy.

III. EXPERIMENTAL

Single crystals of $YBa_2Cu_3O_{7-x}$ and $Bi_2Sr_2CaCu_2O_y$ with zero-field transition temperatures of 90 and 85 K respectively, were prepared by a partial melting technique. Measurements were made using a commercial SQUID susceptometer²² equipped with a pair of orthogonal pickup coils—one coaxial with the superconducing solenoid, and the other at right angles to the solenoid axis. A sample holder, which permitted rotation of the sample about an axis at right angles to the field direction in addition to rotation about the axis of the field, was built specifically for these measurements.²³ The rotation angle was measured to be accurate to $\pm 0.1^\circ$.

The transverse and longitudinal magnetization components were then investigated as a function of temperature, angle, and magnetic field in the regime of reversibility. The method by which the regime of reversibility was determined depended on the type of measurement being made. First, when magnetization was measured as a function of temperature at fixed field and angle, the onset of reversibility was found by noting the temperature at which the field-cooled (FC) and zero-field-cooled $(ZFC)^{24}$ data became indistinguishable. Measurements were then carried out at temperatures above this point so as to assure reversibility. For measurements as a function of angle at fixed temperature and field, the angle was swept in both directions. In this way one can be certain that the data is independent of the rotation direction. At fixed temperature and angle, both components of magnetization show the $\ln H$ dependence predicted by Eqs. (2) and (3). Data can be taken for both increasing and decreasing fields to demonstrate reversibility in this case. Clearly, equilibrium theory can only be applied to data obtained in the reversible region.

One type of run consisted of cooling the sample in a field to the desired temperature and then measuring M_I and M_T as a function of θ . Figure 1 shows typical data for a $Bi_2Sr_2CaCu_2O_{\nu}$ crystal in a field of 1.0 T. The crystal dimensions were $3.4 \times 1.1 \times 0.28$ mm.³ In agreement with theory, M_L is a symmetric function of angle centered at $\theta = 90^{\circ}$ while M_T is antisymmetric. Also, the transverse magnetization goes to zero when the field is aligned with one of the principle crystal directions. Comparing the magnitudes of the two components, it can be clearly seen that the transverse magnetization can be very large for a highly anisotropic system such as $Bi_2Sr_2CaCu_2O_{\nu}$. In fact, for most of the angular range, M_T is quite a bit larger than M_L . This is a clear indication of the strong tendency for vortex currents to flow close to the basal plane.

Figure 2 shows the magnetization ratio, M_T/M_L , as a function of θ using the 70-K data of Fig. 1. The peak of the ratio occurs at an angle which is very close to 90°.



FIG. 1. Transverse and longitudinal magnetization as a function of θ for a Bi₂Sr₂CaCu₂O_y single crystal cooled in a 1.0 T field. Shown here are data for 60, 65, 70, 75, and 80 K, all in the reversible regime. Longitudinal and transverse moments are measured simultaneously with separate pickup coils. The background signal of the sample holder was carefully measured and subtracted to give the data shown here.



FIG. 2. The ratio of transverse to longitudinal magnetization plotted vs θ , using the 70-K data of the Bi₂Sr₂CaCu₂O_y crystal from Fig. 1. The solid curve represents the best fit of Eq. (4) to the data, giving $m_3/m_1 = 280\pm20$. Also shown is the magnetization ratio for a YBa₂Cu₃O_{7-x} crystal at 70 K and 5.0 T. The dashed line shows the best fit to Eq. (4), giving $m_3/m_1 = 30\pm5$.

Using 3D anisotropic London theory there are several ways to extract the mass ratio directly. Most simply, Eq. (4) can be solved for m_3/m_1 after having substituted the observed value of M_T/M_L at a known θ . Another way to proceed is to note that the magnetization ratio reaches a maximum value of $(m_3/m_1)^{1/2} (m_3 - m_1)/2m_3$ at an angle θ_m which is given by $\tan^2 \theta_m = m_3 / m_1$. This can also be used to compute the ratio. In practice, it is better to fit Eq. (4) to all of the $M_T/M_L(\theta)$ data, varying m_3/m_1 to obtain the best fit. The value obtained from the data of Fig. 2 is $m_3/m_1 = 280 \pm 20$. This is much lower than the values reported for $Bi_2Sr_2CaCu_2O_{\nu}$ by Farrell² and of Palstra²⁵ who obtained ratios of ≈ 3000 , but comparable to the value of ≈ 225 reported by Kang.¹ Lower criticalfield anisotropy experiments by Krusin-Elbaum³ give values of ≈ 20 . It must be noted that some experiments are plagued by systematic uncertainties which may cloud the true value of the mass ratio. Some of the values of the effective mass ratio reported may not represent a determination involving true thermodynamic values of the needed superconducting parameters. For example, the pronounced flux flow resistance in these compounds can hinder the determination of H_{c2} by transport methods. In addition, because of the strong angular dependence of many of the thermodynamic quantities, microstructural misalignments within a sample which is assumed to be a single crystal (or perfectly oriented), will affect the values determined by experiments. The wide range of values reported in the literature underline the importance eliminating all spurious effects.

Figure 2 also shows the magnetization data for a YBa₂ Cu_3O_{7-x} crystal. The mass ratio in this case is found to be 30±5. This is consistent with the measurements of other workers who have obtained values in the range from 25 to 90.²⁶ The YBa₂Cu₃O_{7-x} crystals used in this

study had a narrow reversible magnetization regime which severly limits the range of applicability of the analysis. This can be attributed to stronger pinning in the samples of YBa₂Cu₃O_{7-x} samples relative to that in Bi₂Sr₂CaCu₂O_y samples.

A close comparison of the fit of Eq. (4) to the data of Fig. 2 for $Bi_2Sr_2CaCu_2O_{\nu}$ is of great importance. Equation (4) does account for the general trend of the data, but the fit is not within the experimental error of the data points. From the considerations of Sec. II, the sample should be in a regime where it shows 2D character. To our knowledge, explicit calculations which predict the magnitude of the longitudinal and transverse magnetization components for a system in the weakly coupled limit have not been made, so we cannot directly determine if such theory would fit the data better than the anisotropic London model. It is essential that such calculations be carried out. It is possible that a quasi-2D model could exhibit an angular dependence of M_T/M_L similar to the 3D anisotropic case, even though other parameters such as $H_{c2}(T)$ are vastly different for the two cases.

A useful relation can be derived from Eq. (2) which allows determination of the anisotropic mass ratio by measuring the longitudinal magnetization alone. With a little algebra it can be shown that

$$\frac{dM_L(0^\circ)/d\ln(H)}{dM_L(90^\circ)/d\ln(H)} = \left(\frac{m_3}{m_1}\right)^{1/2}.$$
 (5)

From this equation the anisotropic mass ratio can be determined by measuring M_L as a function of field (in the regime where $H_{c1} \ll H \ll H_{c2}$) with the field both parallel and perpendicular to the *c* axis. Since measurement of the transverse moment is not required, this technique



FIG. 3. Longitudinal magnetization as a function of log field for $\theta = 0^{\circ} \pm 0.3^{\circ}$. (\mathbf{H}_a parallel to the *c* axis) and for $\theta = 90^{\circ} \pm 0.3^{\circ}$. (\mathbf{H}_a perpendicular to the *c* axis) for the Bi₂Sr₂CaCu₂O_y crystal. Using this data in Eq. (5) gives $m_3/m_1 = 80 \pm 10$. The sample was oriented at $90^{\circ} \pm 0.3^{\circ}$ by measuring M_L and M_T as a function of θ , and then positioning the sample at the position where M_L was a minimum and M_T crossed through zero.

would be useful when only M_L can be measured. However, there is an important caveat. For the case of a system with large anisotropy, precise alignment is absolutely necessary for the measurement at $\theta = 90^{\circ}$. If there is even a slight misalignment from the field in plane, Eq. (2) shows that the term containing m_3 will begin to contribute substantially to $dM_L/d\ln H$. Figure 3 shows the dependence of M_L on H for both the 0° and 90° case using the same $Bi_2Sr_2CaCu_2O_y$ crystal. From these plots $dM_L/d\ln(H)$ can be determined easily. Applying Eq. (5) gives an anisotropic mass ratio of 80 ± 10 . This value is inconsistent with the value determined from Fig. 2. This inconsistency may again be an indication that the 3D theory is not entirely applicable in this case. For a system in the quasi-2D limit, the character of the vortices for the two orientations would be in two extremely different limits. For the field applied along the c axis, the vortices would be Abrikosov-like, while for the field applied in plane, they would be Josephson-like.

Figure 4 shows field-cooled M_T/M_L data as a function of temperature on Bi₂Sr₂CaCu₂O_y for H=1.0 T and at various orientations. An important feature to note is that within experimental resolution there is effectively no temperature dependence of M_T/M_L over the region of reversibility. This implies a temperature-independent anisotropic mass ratio. Theories developed for multilayered metallic systems state that the anisotropy ratio should be a temperature dependent quantity when $\epsilon < 1.^{10}$ The behavior of multilayered systems in this regime indeed indicated that this was true at least in the case of measurements of $H_{c2}(T)$. It is not known whether the magnetization should also reflect this same temperature dependence. The 3D effective-mass theory does not imply any



FIG. 4. The magnetization ratio plotted as a function of temperature for various fixed angles for the $Bi_2Sr_2CaCu_2O_y$ crystal. In each instance, the crystal was field cooled in a 1.0-T field. Note that the ratio is independent of temperature in the reversible region. The arrows denote the position of the irreversibility temperature for those select angles. The irreversibility temperature was determined by the point at which FC and ZFC data first become indistinguishable.

temperature dependence of the effective-mass ratio.

The temperature dependence of M_T/M_L was determined a second way. When M_T/M_L is measured as a function of θ at fixed temperature, the data is reproducible within experimental error when the measurements are carried out at different temperatures as long as the data is taken in the reversible regime. The peak in M_T/M_L at low temperatures and high angles in Fig. 4 is in the irreversible regime. The difficulties associated with this will be discussed later.

Figure 5 shows the field dependence of M_T/M_L at a few angles for $Bi_2Sr_2CaCu_2O_{\nu}$. In general, the magnetization ratio is independent of field, which is to be expected from Eq. (4) for field strengths $H_{c1} \ll H \ll H_{c2}$. The relationship between the applied field H_a and the internal field H is given as $H_i = H_{ai} - 4\pi N_{ik}M_k$ (sum over repeated indices).²⁷ The effect of the specific geometry is accounted for by N_{ik} , the demagnetization tensor. A single crystal may be approximated as an ellipsoid of rotation for the present purposes. In large fields, shape effects are negligible since the demagnetization term is much less than the applied field term. This is because the equilibrium magnetization M has a magnitude considerably less than $H_{c1}/4\pi$, and for large fields $H_a \gg H_{c1}$. In other words, for $H_{c1} \ll H \ll H_{c2}$, taking the internal field to be equal to the applied field is a good approximation. However, at much lower fields, a sample with a demagnetization tensor that has a nonzero principle value along its rotation axis will produce a transverse magnetization due to the sample shape when the field is applied in a direction other than 0° or 90° . If the rotation axis is collinear with the c axis, this transverse magnetization will have a $sin(2\theta)$ dependence which is in contrast with that given by Eq. (3).

It is useful to consider further the behavior of the system in the irreversible region. Irreversibility is an important and technologically relevant issue, since large



FIG. 5. The magnetization ratio vs field for select temperatures at $\theta = 79.0^{\circ}$ and $\theta = 40.0^{\circ}$ for the Bi₂Sr₂CaCu₂O_y crystal. Note that M_T/M_L is essentially independent of field, except for low fields where shape effects become appreciable.

current applications are contingent on the ability of the superconductor to pin flux. So far there is no general theory describing the "critical state" of an anisotropic superconductor. By "critical state" we mean the response of the flux density distribution and the resulting magnetization to a changing field, as described, for instance, by Bean for isotropic superconductors in the presence of pinning.²⁸ For an anisotropic superconductor in an applied field oriented not along a principle crystal direction, it is also necessary to consider the direction of the magnetization vector in analyzing the "critical state." Whereas, in equilibrium the vortex will assume an orientation determined by purely thermodynamic considerations, it is likely that in the irreversible region pinning forces play a role in determining the direction of the flux. The direction of the vortices could be very different than for the equilibrium case, as well as dependent on past thermomagnetic history. The direction of the magnetization produced by the circulating currents induced by the presence of flux density gradients may also depend on anisotropy in a nontrivial way. Recent calculations predict the existence of a new vortex interaction force which arises only in the presence of anisotropy.²⁹ Earlier experiments demonstrate the tendency of the $YBa_2Cu_3O_{7-x}$ compound to pin vortices in the c-axis direction.³⁰ We have investigated some aspects of the problem by studying M_L and M_T for the FC and ZFC cases as a function of temperature at fixed angle.

Figure 6 shows M_T/M_L versus temperature for $Bi_2Sr_2CaCu_2O_y$ at $\theta = 79^\circ$ in a 1.0 T field for the FC and ZFC cases. As one lowers the temperature, the two curves are indistinguishable until one reaches a temperature $T_{irr} \approx 38$ K. Below this point the system is out of equilibrium, as the value of magnetization becomes a function of the sample's thermomagnetic history. For



FIG. 6. M_T/M_L vs temperature for $\theta = 79.0^\circ$ and H = 1.0 T for the Bi₂Sr₂ CaCu₂O_y crystal. The circles represent ZFC data and the X's represent FC data. T_{irr} is the temperature above which the data is independent of thermomagnetic history. The peak in M_T/M_L at low temperatures is attributed to be a consequence of the anisotropic "critical state."

temperatures less than $T_{\rm irr}$, the magnetization ratio has a very strong temperature dependence for both FC and ZFC curves. At very low temperatures M_T/M_L is small, for increasing temperatures it rises to a value which is even larger than for equilibrium, and for yet higher temperature the value decreases to the reversible one. It is peculiar that both the FC and ZFC data show this peak in M_T/M_L . In addition, both magnetizations are time dependent below $T_{\rm irr}$. This is another indication that the system is out of equilibrium. If we examine Fig. 4, we see that the peak in M_T/M_L disappears at low values of θ . Because of these observations, it seems likely that this peak is a consequence of an anisotropic critical state, although no detailed explanation is available.

IV. DISCUSSION

Experiments were done which measure the longitudinal and transverse components of magnetization simultaneously for fields in the regime $H_{c1} \ll H \ll H_{c2}$. The angular dependence of the magnetization ratio was compared to the predictions of 3D anisotropic theory. A general correlation of experiment and theory was found, however there was not a perfect fit. The possibility of the system being in the weakly coupled limit was discussed. It should be noted that there is no theory which accounts for magnetization in this regime explicitly.

The fact that vortex currents are flowing in the CuO planes even at high angles is an important consideration in other types of experiments as well. For measurements in which the applied field is assumed to be in the basal plane by rough positioning in that direction, a slight tilt will result in enormous in-plane currents. If the in-plane

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currents are so large that they are approaching the GL critical current, the order parameter in the layer will be reduced. This would affect not only the condensation energy in the layer but also the interlayer Josephson coupling. (See, for example, the Lawrence-Doniach model—Ref. 8.) The relative coupling could then actually be a function of angle when the system is oriented in a large field. Also, in the quasi-2D limit, the possibility of having vortices that have kinks as they pass through the plane may affect not only magnetization measurements, but others as well (e.g., in-plane transport measurements).

Given the apparent contradictions in the recent literature relating to the coupling regime which actually describes the high- T_c superconductors, i.e., quasi-2D or anisotropic 3D, it is clear that further experimental and theoretical work on this problem is required.

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Note added in proof. Recently Farrell³¹ presented torque measurements on YBa₂Cu₃O_{7-x} that suggest a dimensional crossover at $T \approx 80$ K, which is close to the temperature predicted for crossover in the present text.

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