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Anomalous behavior of the angular-dependent magnetic relaxation in single-crystal Bi₂Sr₂CaCu₂O_y

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Relaxation of the longitudinal and transverse components of the remanent magnetic moment of single-crystal $Bi_2Sr_2CaCu_2O_y$ has been investigated as a function of angle. When the angle between the applied field and the *c* axis is not too large, the remanent moment quickly aligns with the *c* axis. At extremely high angles the alignment is slow and at low temperatures the transverse magnetization even increases with time. The temperature dependence of the normalized relaxation rate exhibits two peaks, suggesting the existence of two separate pinning mechanisms.

Recently there has been considerable interest in the structure and behavior of vortices in strongly layered superconductors such as the extremely anisotropic hightemperature superconductor $Bi_2Sr_2CaCu_2O_x$.¹⁻⁵ The equilibrium or quasiequilibrium phase diagram of such a system in a field is of interest because of the possibility of vortex-lattice melting⁶ or a vortex-glass transition.⁷ A quantitative understanding of the motion of vortices in this context will involve ideas relating to pinning,⁸ the distribution of activation energies in a flux creep picture,⁹ the effects of anisotropy,¹⁰ and the quasi-two-dimensional (quasi-2D) layered nature of the material.^{2,3} The structure of a vortex directed at an angle to the c axis though may also be more complicated than an Abrikosov vortex in an isotropic system, as it may have both Abrikosov and Josephson character. Such vortices have been characterized as having long lengths parallel to the layers with the segments passing through them described as kinks.⁵ It has also been suggested that "pancake" vortices may be found in the angular geometry.¹¹ Current loops in successive planes would be uncorrelated for these tilted entities.

Here we present the results of magnetization-relaxation experiments on single crystals of the high- T_c superconductor Bi₂Sr₂CaCu₂O_y which highlight the complex behavior of the vortices. Specifically, we have measured as a function of various crystal orientations, the time dependence of the longitudinal (parallel to the applied direction) and transverse (perpendicular to the field) components of the remanent magnetization after an applied field has been removed. Qualitatively unusual and unexpected features of the data include: (i) the observation of two peaks rather than one, in the temperature dependence of the normalized relaxation rate, when the angle between the field and the c axis is nonzero, and (ii) the observation of an *increase* in the c-axis magnetization with time at low temperatures and high angles.

 $Bi_2Sr_2CaCu_2O_y$ single crystals were grown by a flux method and were found to be of high quality by x-raydiffraction rocking-curve analysis. In the process of a measurement, a crystal was mounted on a quartz sample holder at the desired orientation angle. The latter was determined by reflecting a laser beam off the shiny crystal surface and measuring the angle between the incoming and outgoing beams. The accuracy of the angle between the applied field and the crystal c axis, θ_a , was $\pm 0.5^\circ$. Magnetization measurements were made using a Quantum Design susceptometer which was equipped with two superconducting quantum interference devices (SQUID's), one for measurement of the longitudinal component of the magnetic moment and the other for the transverse component.

Relaxation measurements were performed as a function of crystal orientation and temperature. In each case, the crystal was cooled in a 500-G field, through T_c , down to the temperature of interest. Once temperature stability was achieved, the field was turned off, and simultaneous measurements of both the longitudinal and transverse moments were made as a function of time. This procedure was repeated for all the other temperatures and other crystal orientations.

Both components of magnetization were found to change slowly in time and to have ln(t) dependences. The $M_i(t,T)$ data, where i=l,t for longitudinal and transverse, respectively, t is time, and T temperature, can be represented in the form of the normalized logarithmic rate of decay,

$$S_{ni} = \frac{1}{M_i} \frac{dM_i}{d(\ln t)} \,. \tag{1}$$

Presenting the approach to equilibrium in such a form can be justified both intuitively and by using the simplest model of flux creep. Figure 1 shows S_{nl} and S_{nl} as a function of temperature for various orientations. Examining the temperature dependence in Fig. 1, one sees the emergence of two peaks in S_{nl} . Their relative magnitudes change systematically as a function of the orientation, in the sense that the peak at lower temperatures decreases in magnitude and that at higher temperatures increases in magnitude as θ_a increases. One sees a similar behavior for S_{nl} .

In Fig. 2 the data at each orientation for longitudinal and transverse relaxation are plotted superposed. In this way their functional dependences can be compared. At low angles data for the two directions follow each other.



FIG. 1. Normalized logarithmic relaxation rates for the (a) longitudinal and (b) transverse components of magnetization for different orientations of the crystal in the field, where θ_a is the angle between the *c* axis and the direction of the field which the sample was cooled in. The normalized rate was calculated at $t_{exp} = 10$ min. This particular crystal was $3.4 \times 1.1 \times 0.28$ mm¹³ in size with its *c* axis along the smallest dimension.

However, at higher angles the two components exhibit quite different temperature dependences with the difference most pronounced when the crystal is oriented at 89.5° (field directed nearly in plane; again see Fig. 2). This implies that ϕ , the angular orientation of the magnetization vector relative to the *c* axis as well as M_0 , its magnitude, must be changing, and there are two different types of relaxation. Where the difference between the normalized rates or decay, S_{nl} and S_{nt} versus temperature is most pronounced at $\theta_a = 89.5^\circ$, at low temperatures S_{nt} is actually positive. That is the transverse moment actually *increases* with time rather than decreases. Its time dependence is logarithmic over the time scale of the experiment (2 h), as before. This behavior was *not* expected, as the approach to equilibrium usually involves ejection of vortices resulting in decay of the remanent moment.

Flux creep theories for crystals oriented at arbitrary angles to an applied field have not been developed in either the 3D anisotropic crystal or a strongly layered (quasi-2D) models. These models would have rather different vortex cores. In the quasi-2D layered model, the order parameter is modulated in space along the direction normal to the layers (c axis).¹ In the mixed state, if the field is directed at an angle to the planes, vortex cores will reside in the region between planes, except when they cross the planes.⁵ The vortex core line will thus have bends in it. In the anisotropic 3D model, vortices run through the system at an angle, losing condensation energy in their cores. For vortices oriented along the c axis, there would be little difference between the two models. In the quasi-2D model the current sustaining the vortices would be confined to the layers. For Abrikosov-like vortices, the current loops are like pancakes stacked on top of one another with a space in between.¹¹ In either picture, if the applied field is turned off, the system will try to reach equilibrium, a state in which there are no vortices in the sample. If the vortices are directed at an angle, they might rotate while leaving the sample (or rotate and then leave, depending on the relative energy barriers), rather than just leave the sample at some angle.

As was mentioned earlier, the temperature dependence of the normalized relaxation rate as shown in Fig. 1, exhibits two distinct peaks for the different orientations. Qualitatively this suggests two different types of pinning with different scales of activation energy. One type may be intrinsic pinning associated with motion of vortices across the planes.^{2,3} In previous analyses of relaxation data,⁹ peaks in the normalized rate of decay were explained by assuming that the system could be described by a distribution of pinning sites of different activation energies, $m(U_0)$ (where U_0 is an activation energy), instead of just one energy, U_0 . The appearance of two peaks would then indicate the contribution of two significantly different characteristic energies [peaks in $m(U_0)$], a low energy one which is overcome at about 20 K, and a higher energy one which is not overcome until about 60 K. We observe a decrease in the magnitude of the low temperature peak and an increase in the magnitude of the high temperature peak as θ_a is increased, and not a continuous shift of a single peak from low to high temperatures.

Just after the field is turned off, one can determine the total remanent moment vector by measuring its longitudinal and transverse components and adding these as orthogonal vector components. Figure 3 shows such data obtained after field cooling to 5 K in 500 G. With the exception of large angles, one finds that the direction of the remanent moment to be along the *c*-axis direction as soon as the measurement is made. That is, the magnetization vector can be described as rotating to the *c*-axis direction once the field is turned off. Also, the magnitude of the remanent moment scales closely with the component of applied field along the *c* axis. At $\theta_a = 89.5^\circ$ the system traps more flux than expected by this simple argument. This is similar to what has been reported by Felner *et al.*¹²

The path to equilibrium when the vortices are at an angle relative to the c axis appears not to involve the vortices

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FIG. 2. Logarithmic relaxation rates as in Fig. 1, but plotted for each orientation separately. For $\theta_a = 89.5^{\circ}$, the transverse relaxation rate is positive indicating an increase in the transverse moment with time (also logarithmic). The complex behavior at high angles implies a rotation of the remanent moment vector, while at low angles the direction of this vector remains fixed.

staying tilted and leaving the sample via the standard creep process. A possible scenario in the simplified picture of a quasi-2D layered superconductor with only magnetic coupling between neighboring layers might be the following: vortices oriented at high angle might be described as stacks of current loops displaced greatly from one another. When the applied field requiring them to stay at this angle



FIG. 3. Magnitudes of the longitudinal and transverse components of the remanent moment plotted for various fixed orientations. In each case the sample was cooled in the 500-G field down to 5 K where the field was then turned off. Given are values of the longitudinal moment, the transverse moment, and the magnitude of the remanent moment vector. The solid lines represent the expected values of these moments if the magnitude of remanent moment were determined by the component of applied field in the c-axis direction, as given by $|M| = M_1 \cos \theta_a$. For such a case $M_L = M_1 \cos^2 \theta_a$ and $M_T = M_1 \cos \theta_a \sin \theta_a$. The data indicates that the remanent moment is aligned with the c axis with the exception of $\theta_a = 89.5^\circ$.

is removed, there would be a force to restore them to a straight stack. In the process of a vortex attempting to rotate to align with the *c*-axis, it might encounter pinning sites while still at some high angle. The driving force for rotation might not be sufficient to overcome the pinning force, and thermal activation would be the only way for rotation to occur. When the temperature is raised, more thermal energy is available to allow rotation, resulting in an increased rate. In Fig. 1, for $\theta_a = 89.5^\circ$, the behavior of the transverse moment is consistent with the following picture: at temperatures slightly greater than 5 K, the increase in thermal energy appears to favor an increase in the rate of rotation of flux lines towards the c axis more than an increase in the rate of escape of trapped flux. As a consequence, the magnetization along the c axis grows in magnitude with time. In the vicinity of 20 K, these two competing processes cancel each other, and the transverse magnetization remains constant. As the temperature is increased above 20 K, the net change is dominated by the flux leaving the sample and not the rotation. Hence now the magnitude of the magnetization diminishes with time.

It is important to note that the transverse relaxation rate for $\theta_a = 89.5^{\circ}$ continues to increase in magnitude to a very high value with increasing temperature after the change in sign at about 20 K. This means that at higher temperatures either the characteristic energy of the pinning corresponds to a high-energy value, or a very different mechanism for flux motion is at work. In the quasi-2D layered picture, the vortices in question would be pancake vortices moving in the *a-b* plane. It is our speculation that the change in sign of the decay rate may correspond to a transition, or crossover from a vortex phase in which the pancake vortices in different layers are correlated, to one in which they are uncorrelated. (This is similar to the comparison made between independent vortex-antivortex pancake pairs at small length scale and multilayer vortex rings at large length scales in the consideration of thermally produced topological defects in a quasi-2D coupled layered system.¹³) Chakravarty, Ivlev, and Ovchinnikov⁴ have shown that the interaction energy ε_{int} between current loops of the same vortex on adjacent planes is of the form

$$\varepsilon_{\rm int} = \frac{\lambda_c}{\lambda_{ab} (4\pi\lambda_{ab})^2} \frac{\Phi_0^2 d^2}{2} \cos\theta \,, \tag{2}$$

where d is the interplanar spacing, λ_c and λ_{ab} are the penetration depths in the c direction and in the a-b plane, and θ is the angle of the vortex. The coupling between the loops becomes weaker as the angle is increased. Taking an angle of 89.5° and the parameters for $Bi_2Sr_2CaCu_2O_{\nu}$, one obtains a value of ε_{int}/k_B of 59 K. This is to be compared with 20 K at which the relaxation rate changes sign, and which we speculate to correspond to the temperature at which interplanar correlation is lost. This level of agreement should be considered to be promising given the relative imprecision of some of the parameters, and the actual lack of a detailed theory which includes the effects of fluctuations and the many vortex character of the interaction. In essence, at low temperatures the vortex rotates because it is strongly correlated (3D) and it has a lower energy to align with the c axis. At higher temperatures the pancake vortices in different layers can become uncorrelated and leave the sample independently.

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To summarize, we have observed complex relaxation processes in the escape of vortices from a single crystal of the highly anisotropic and strongly layered high-temperature superconductor, $Bi_2Sr_2CaCu_2O_y$. The most unusual behavior is prominent when the angle between the *c* axis and the field direction is large. It is not obvious that the observed phenomena are related to the vortex lattice melting or the glass transition which has been the subject of considerable research. Whether these observations are at all related to concepts such as "kinks"⁵ or "pancake"¹¹ vortices is not known. As the various available models make no specific predictions relating to these phenomena, it is clear that further theoretical work is needed in order to obtain a quantitative description of the vortex structure and dynamics in this regime.

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