RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 44, NUMBER 14 1 OCTOBER 1991-II

Flux-lattice melting, anisotropy, and the role of interlayer coupling in Bi-Sr-Ca-Cu-O single crystals

C. Duran, J. Yazyi, and F. de la Cruz
Centro Atomico Bariloche and Instituto Balseiro, 8400 S. C. de Bariloche, Rio Negro, Argentina

D. J. Bishop
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

D. B. Mitzi* and A. Kapitulnik
Stanford University, Stanford, California 94305

(Received 20 March 1991)

We have used the high-Q mechanical-oscillator technique to probe the vortex-lattice structure in high-quality Bi-Sr-Ca-Cu-O single crystals over a wide range of magnetic fields (200 Oe to 40 kOe), and relative orientations θ between the magnetic field and the crystalline $\hat{c}$ axis. In addition to the large softening and dissipation peak previously observed and interpreted as due to flux-lattice melting, another distinctly different peak at higher temperatures is seen. The temperatures where the dissipation peaks take place are solely defined by the parallel component of the field $B\cos\theta$, while the restoring force on the oscillator is due to both field components. We suggest that the two peaks are due to the softening of interplanar coupling at the low-temperature peak, and melting or depinning of the two-dimensional pancake vortices at the higher-temperature peak.

Despite the general acknowledgment that anisotropy plays a fundamental role in understanding the magnetic-flux response of the oxide superconductors, few experiments have explicitly studied the angular dependence of their superconducting properties. There are torque-magnetometry[1] measurements which have shown that the square-root ratio of effective masses is 55 in the Bi-Sr-Ca-Cu-O compound. Recent results on the low-field relaxation[2] in Bi-Sr-Ca-Cu-O have shown that the normalized relaxation rate exhibits two peaks in its temperature dependence, and based on this, the existence of two different pinning mechanisms was proposed. The results were consistent with a picture in which the magnetic moment is determined solely by the normal component of the induction field, $B\cos\theta$. At large angles this picture fails since a large amount of flux was found to be trapped in the direction perpendicular to the $\hat{c}$ axis. The low-temperature peak in the relaxation rate at approximately 20 K was attributed to the magnetic decoupling of two-dimensional vortices.

Using decoration techniques, Bolle *et al.*[3] have found a flux-lattice structure of flux chains in Bi-Sr-Ca-Cu-O single crystals when the magnetic field is applied at angles between 60 and 85 degrees with respect to the $\hat{c}$ axis. They have also found that the average density of vortices as a function of angle is determined only by the $B\cos\theta$ field component; no distortion of the lattice due to the other field component parallel to the crystal surface occurs.

All these results point toward the important role of the anisotropy in this system. The interpretation within a picture in which the flux structure is determined only by the $B\cos\theta$ component implies that the magnetic flux is quantized only in the $\hat{c}$ direction, while the field component parallel to the Cu-O planes penetrates the sample as if it were a magnetically transparent material.

The mechanical-oscillator technique[4-7] has been shown to be quite sensitive in the detection of transitions in the vortex state of superconductors. In this paper we report the results of mechanical-oscillator measurements in a wide range of fields and temperatures in high-quality Bi-Sr-Ca-Cu-O single crystals. Our measurements show dissipation peaks at two different temperatures for a given field and angle between the field and the crystallographic $\hat{c}$ axis. We have found that, although the temperatures at which the peaks take place are only determined by the $B\cos\theta$ field component, the restoring force on the oscillator is due to both field components. If the flux is quantized only in the $\hat{a}$ and $\hat{b}$ directions, the pinning force would be active only against the $B\cos\theta$ component. On the other hand, if Josephson vortices interconnect the pancake vortices, the pinning centers would react against the force exerted by both field components. This means that the magnetic flux is quantized in the $\hat{c}$ direction as well as in the $\hat{a}$ and $\hat{b}$ directions, giving support to the picture of two-dimensional Abrikosov pancakes in the Cu-O planes coupled by Josephson vortices through the interplane regions.

In our experiment, the sample to be studied is attached to a thin-silicon paddle, which is gold coated on both sides for capacitive drive and detection. The motion is driven self-resonantly by means of a feedback loop. Our measurements were carried out at frequencies of 1.5 and 4.8 kHz, corresponding, respectively, to the first cantilever and torsional modes of the oscillator. Typical Q's were above 10^5 at liquid-helium temperatures and zero-magnetic field, and our oscillators had frequency stabilities of better than 0.1 ppm. The sensitivity at low fields is essentially controlled by the size of the sample, and in the present experiment we were able to measure at fields as low as 200 Oe using large crystals ($6\times3\times0.03$ mm^3). The measurements with **H** applied exclusively parallel to the $\hat{c}$ axis were made using a superconducting magnet, while the angular dependence was studied by means of a rotating iron-core magnet placed outside of the cryostat. This configuration allowed us to change the relative orientation θ between the magnetic field and sample in a precise and reproducible way ($\pm 0.2°$).

The samples used were $Bi_{2.1}Sr_{1.9}Ca_{0.9}Cu_2O_{8+\delta}$ single crystals grown and characterized as reported elsewhere.[8] The crystals were platelets shaped with the short dimension along the crystallographic $\hat{c}$ axis. High-resolution superconducting-quantum-interference-device (SQUID) magnetization measurements for these crystals have shown sharp bulk-superconducting transitions with an onset of superconductivity at 86 K, and zero-field-cooled full shielding at low fields. Decoration experiments[9] on these particular samples have shown very well-ordered flux lattices indicating the absence of macroscopic inhomogeneities.

In Fig. 1 we show the results of a typical run for our experiment with **H** parallel to $\hat{c}$ at a constant applied field of $H = 5$ kOe as a function of temperature. At low temperatures the solid flux-line system is pinned to the superconducting crystal, and the tilting of the flux-line system with respect to the external magnetic field gives rise to an extra restoring force that increases the resonant frequency of the oscillator. When the temperature is increased, the response of the oscillator indicates a large softening of the flux structure. Associated with this softening there is a sharp dissipation peak, and at its maximum we define the melting temperature T_m. These measurements are in excellent agreement with results obtained on crystals of different quality and oxygen treatment.[7,10] The main result of this paper is that above the melting temperature the flux lines are still softly coupled to the sample, a fact that is made evident by a residual stiffness associated with the magnetic field. This residual stiffness disappears at the same temperature where a second, smaller, dissipation peak takes place. We define T_i as the maximum of this second peak.

The amplitude at T_i is found not to be proportional to the oscillator driving force. In fact, it is found that below T_i the amplitude is never proportional to the driving force, even for the smallest detectable signals, while it is linear with the driving force over the entire range of drives for $T > T_i$. This onset of intrinsically nonlinear behavior can be understood as a consequence of the onset of long-range coherence in the system and will be discussed in more detail later.

Figure 2 shows the dissipation of the oscillator as a function of temperature for four different angles, θ, of the applied field relative to the $\hat{c}$ axis. The strong anisotropy of both T_m and T_i is evident. Although the amplitudes of the peaks depend on θ, the characteristics of each peak are preserved when the field is rotated. The peak at T_m remains sharp (~ 3 K at full width at half maximum) in the whole range in which it is detectable, while the width of the peak at T_i is always ~ 10 K. The maximum amplitude of T_m takes place at $\theta \sim 0°$, decreasing rapidly for higher angles and becoming undetectable close to $\theta \sim 90°$. The amplitude of the peak at T_i remains finite in the whole range of angles, being a minimum at $\theta \sim 0°$ and a maximum at $\theta \sim 90°$. These results show that the two peaks arise from qualitatively different processes, and not from a single process with two different characteristic temperatures. The trivial explanation that these effects could arise from the existence of two different crystallographic phases can also be easily ruled out. Low-field SQUID magnetization curves on this sample show no evidence of two different critical temperatures with a precision of a few flux quanta, either in field-cooling or zero-field-cooling experiments. Moreover, if the two peaks were the consequence of regions with different crystallo-

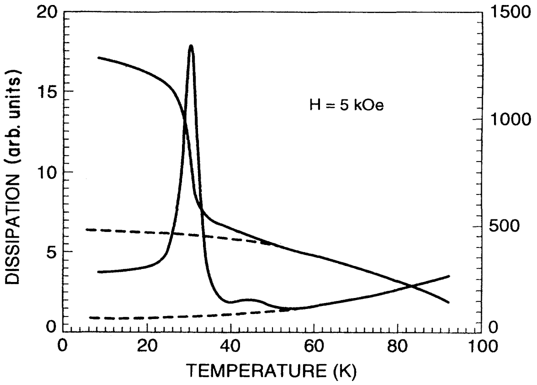

FIG. 1. Temperature dependence of the real and imaginary response of the mechanical oscillator with the Bi-Sr-Ca-Cu-O crystal attached. The dashed lines represent the behavior at zero field, i.e., the background due to the elastic properties of the silicon paddle itself.

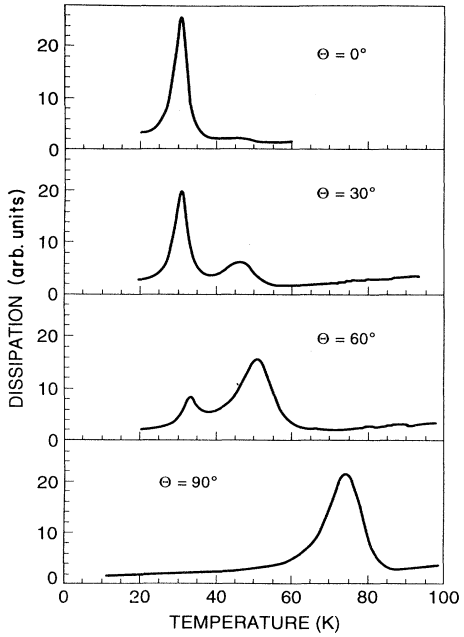

FIG. 2. Dissipation peaks at a fixed applied field of 6.5 kOe, for different relative orientations to the sample. θ is the angle defined by the field and the $\hat{c}$ axis of the single crystal. Notice the disappearance of the melting peak as **H** becomes parallel to the Cu-O planes.

graphic orientations, they should cross each other upon rotating the field from 0° to 90° with respect to the $\hat{c}$ axis, in contrast to our results.

The measurements shown in Fig. 2 were carried out with the oscillator in the cantilever mode, because for this mode of vibration the field distortion is the same independent of the field orientation. This fact was checked at low temperatures, where the magnitude of the frequency shift was observed to be independent of θ. For this reason, it is meaningful to consider the changes in the height of the dissipation peaks as characteristic of the physical processes that take place in the vortex structure, and not as arising from changes in the geometry of the experiment. On the other hand, when working in the torsional mode, although the angular dependence of T_m and T_i remains the same, the height of the peaks measured at different angles cannot be compared as in the case of the cantilever mode. The presence of these two distinct lines in the H-T phase diagram is a challenge for theoretical work. The theories[11] based on thermally activated depinning, in the present form, do not predict a phase diagram with two characteristic lines. Phase-transition models[12,13] at least, in principle, allow for such a possibility.

We have studied the angular dependence of T_m and T_i and have found that the two dissipation peaks follow a universal temperature dependence. The scaling parameter is the normal component of field $B_{\parallel} = B\cos\theta$. We find that this is the only relevant field component in determining the location with temperature of the dissipation peaks. In Fig. 3 we have plotted the parallel component of the field $B_{\parallel}$ where the dissipation peaks take place, as a function of the peak temperature for a wide range of total applied fields. In the inset the results for T_i in the low-field range are shown. It is clear that the scaling is quite good. From an experimental point of view, it is important to emphasize that the $B\cos\theta$ scaling law combined with the shape of the field dependence of T_m and T_i implies that the angular dependence of both features at a constant applied field becomes sharper around $\theta \sim 90°$ as the field is increased. This in turn means that the proper measurement of any property in that direction for fields higher than 10 kOe requires a very careful alignment of the sample.

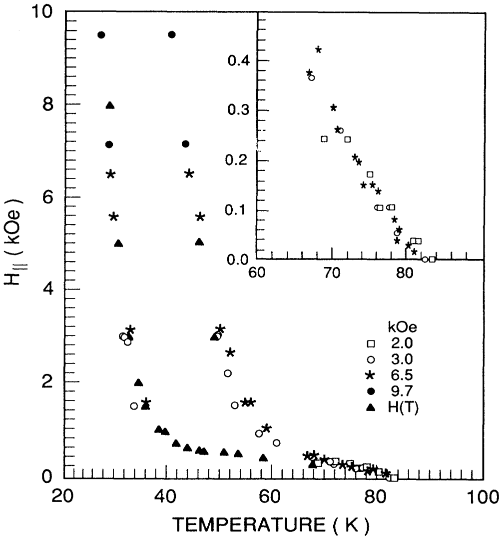

FIG. 3. Phase diagram for the field component $B_{\parallel}$. The solid symbols represent the melting temperature T_m and the open symbols the depinning T_i. The triangles indicate the measurements for varying fields applied parallel to the $\hat{c}$ axis, while the other symbols correspond to measurements performed with fixed values of the applied field at different orientations. The inset shows the low-field detail for the measurements of T_i.

The results of Fig. 3 show that the softening at T_m as well as the complete depinning at T_i are only determined by $B_{\parallel}$. This result is in agreement with previous experiments, leading to a picture where vortices are considered as two-dimensional pancakes nucleated in the Cu-O planes, with a vortex density determined by $B\cos\theta$. In this picture, the field component perpendicular to $\hat{c}$ is assumed to penetrate freely between the Cu-O planes. There is, however, an apparent contradiction between our results and this picture when considering that the relative changes in frequency as well as in the amplitude of the dissipation peak at T_i increase when θ increases. See, for example, Fig. 2.

Both experimental results, the universal behavior of T_m and T_i and the increase in the stiffness of the flux structure with $\cos\theta$ can be reconciled within the same picture, if one assumes that the active pinning centers are effective only on the Abrikosov two-dimensional vortices nucleated in the planes, while the field component between the Cu-O planes induces unpinned Josephson vortices interconnecting the pancakes. In this scenario, T_m represents the softening of the interconnection between the two-dimensional vortices while T_i determines the temperature where the Abrikosov vortices are fully depinned in the planes.

The restoring force which leads to an increase of the oscillator frequency is due to both types of vortices. When θ increases, the length of the Josephson vortices increases. When the oscillator moves, the pinned pancakes do not allow the free displacement of the Josephson vortices along the Cu-O planes, increasing their rigidity. Once the pinning of the two-dimensional vortices is overcome by thermal energy the Josephson vortices slide freely between the planes and the dissipation is only associated with vortex viscosity which should produce a linear-response regime above T_i as is observed.

Previous measurements[7] using the high-Q oscillator technique can be interpreted within the framework of the vortex-glass theory;[12] in that theory there is a true phase transition at the glass temperature T_g. As one approaches the transition from above, there is a diverging correlation length ξ which diverges as $(T_g - T)^{-\nu}$. There is also a critical slowing down with the time scale for critical fluctuations diverging as ξ^z. The oscillator, which is a finite-frequency probe, shows a change in response or a transition when the critically diverging time scale for the transition equals the oscillator resonant frequency. This will always be slightly above the true T_g. Above T_g the current scale for linear-response shrinks as the coherence length grows. The way to understand this is to realize that the measuring current disturbs the vortex configuration that we wish to probe. The larger the current, the shorter the distance over which we disturb any particular configuration. If one disturbs the vortex lattice over some length scale longer than the correlation length, then one should still be in the linear-response regime. The current scale for the crossover from linear response to nonlinear response is given by $J_0 \sim cT/(\Phi\xi^2)$ which goes to zero as one approaches T_g from above. Below the transition, ξ is infinite and there is no linear-response regime. Below T_g one expects that $V \sim \exp[-(J_t/J)^{\mu}]$. The oscillator, which is a very-low-amplitude probe, always detects linear response above T_g but never detects linear response below T_g. This is a consequence of the infinitely long-ranged correlation length below T_g. In contrast, a thermally activated flux-flow model would predict that at low temperatures one would always find linear response when the driving current is small in comparison to the temperature.

The fact that below T_i one always finds a nonlinear response at all driving amplitudes suggests that T_i and T_m are transitions into states with long-range correlations. We would then expect that these might represent true phase transitions[12] involving large numbers of correlated vortices. The lattice then "melts" in two steps with different temperatures for the interlayer and intralayer transitions.

In conclusion, mechanical-oscillator measurements on high-quality Bi-Sr-Ca-Cu-O single crystals have shown that there exist two distinct features upon warming. The angular dependence suggests that they represent features associated with the vortices between the planes and in the planes.

We would like to acknowledge support from M. E. de la Cruz, technical assistance from R. Fuentes and H. Tutzauer, and interesting discussions with J. R. Clem and D. Huse. The SQUID characterization of the samples by H. Safar and H. Pastoriza and a critical reading of the manuscript by J. Luzuriaga are gratefully acknowledged. This work was partially supported by CONICET of Argentina through PID 003900/88, and by TWAS through Research Grants. No. 87/30 and No. 87/63.

---

*Present address: IBM Thomas J. Watson Research Center, Yorktown Heights, NY 10598.

[1] D. E. Farrel, S. Bonham, J. Foster, Y. C. Chang, P. Z. Jiang, K. G. Vandervoort, D. J. Lam, and V. G. Kogan, Phys. Rev. Lett. **63**, 782 (1989).

[2] M. Tuominen, A. M. Goldman, Y. C. Chang, and P. Z. Jiang, Phys. Rev. B **42**, 8740 (1990).

[3] C. A. Bolle, P. L. Gammel, D. G. Grier, C. A. Murray, D. J. Bishop, D. B. Mitzi, and A. Kapitulnik, Phys. Rev. Lett. **66**, 112 (1991).

[4] R. N. Kleiman, G. K. Kaminsky, J. D. Reppy, R. Pindak, and D. J. Bishop, Rev. Sci. Instrum. **56**, 2088 (1985).

[5] R. N. Kleiman, P. L. Gammel, E. Bucher, and D. J. Bishop, Phys. Rev. Lett. **62**, 328 (1989).

[6] P. L. Gammel, A. F. Hebard, and D. J. Bishop, Phys. Rev. Lett. **60**, 144 (1988).

[7] P. L. Gammel, L. F. Schneemeyer, J. V. Waszczak, and D. J. Bishop, Phys. Rev. Lett. **61**, 1666 (1988).

[8] D. B. Mitzi, L. W. Lombardo, A. Kapitulnik, S. S. Laderman, and R. D. Jacowitz, Phys. Rev. B **41**, 6564 (1989).

[9] C. A. Murray, P. L. Gammel, D. J. Bishop, D. B. Mitzi, and A. Kapitulnik, Phys. Rev. Lett. **64**, 2312 (1990).

[10] C. Duran, J. Yazyi, F. de la Cruz, D. J. Bishop, D. B. Mitzi, and A. Kapitulnik (unpublished).

[11] P. H. Kes, J. Aarts, J. van den Berg, and C. J. van der Beek, Supercond. Sci. Technol. **1**, 242 (1989); A. Gupta, P. Esquinazi, H. F. Braun, and H. W. Neumuller, Phys. Rev. Lett. **63**, 1869 (1989).

[12] D. S. Fisher, M. P. A. Fisher, and D. A. Huse, Phys. Rev. B **43**, 130 (1991).

[13] D. R. Nelson and H. S. Seung, Phys. Rev. B **39**, 9153 (1989); A. Houghton, R. A. Pelcovits, and A. Sudbo, *ibid.* **40**, 6763 (1989).