Evidence from Mechanical Measurements for Flux-Lattice Melting in Single-Crystal $YBa_2Cu_3O_7$ and $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_8$

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(Received 8 July 1988)

We have studied flux-lattice melting in single crystals of $YBa_2Cu_3O_7$ and $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_8$ using a high-Q mechanical oscillator. In $YBa_2Cu_3O_7$ with an applied magnetic field $\mathbf{H}\perp\hat{\mathbf{c}}$, flux-lattice melting occurs at H_{c2} as in a conventional three-dimensional superconductor. However, for $\mathbf{H}\parallel\hat{\mathbf{c}}$ flux-lattice melting occurs 3.2 K *below* H_{c2}. We believe that this is evidence for a transition into a vortex-liquid state similar to that seen in two-dimensional superconducting films. For $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_8$ the effect is even more pronounced; with a bulk superconducting transition of 75 K, the flux-lattice melting in this material occurs in both orientations near 30 K.

PACS numbers: 74.60.Ge

The potential for applications of the high-T_c superconductors has produced an unprecedented amount of effort towards the understanding of their physical properties. Many of these applications will require high critical currents which will, in turn, make detailed knowledge of the microscopics of the superconducting flux lattice of importance. To obtain high critical currents, the formation of a flux lattice is crucial because the finite shear modulus of the lattice allows a relatively small number of pinning centers to pin all of the flux lines. A number of authors[1-4] have pointed out that there exist good reasons why the flux lattices in these materials might be unconventional. In this paper we report on mechanical measurements of flux-lattice melting in single crystals of $YBa_2Cu_3O_7$ (YBCO) and $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_8$ (BSCCO). We find that in these materials there is indeed unconventional behavior in the flux lattice. We find a general tendency for the flux lattice to melt well below H_{c2} into a vortex-liquid state similar to that seen in two dimensions.[5] We suggest that a Lindeman criteria for melting might qualitatively explain our results.

In conventional, three-dimensional superconductors the flux lattice is most often a triangular lattice with long-range positional order. In a finite magnetic field approaching $T_c(H)$, the flux lattice melts at T_c because the coherence length $\xi(T)$ diverges and the vortices cease to be well defined objects. Generally, in three dimensions, the lattice is stable and uninteresting over the entire superconducting phase diagram.

However, in two dimensions things can become more interesting.[6] The increased importance of thermal fluctuations in two dimensions produces a flux lattice without long-range positional order which can melt well *below* $T_c(H)$ into a vortex-liquid state. The suppression of the melting temperature T_M from the superconducting transition temperature depends strongly on the normal-state resistance of the film.[6] In the dirty limit, $T_c/T_M = 1+3.8R_\square/A_1R_c$ with $R_c=4.12$ kΩ and $A_1=0.5$. Thus, the observation of a vortex-liquid state strongly suggests that the flux lattice is two dimensional in character.

There are also other possibilities for nonconventional behavior in the flux lattices in these materials. Nelson[7] has pointed out that at the high transition temperatures and for the small coherence lengths in these systems in the Lindeman criterion for melting can become relevant. In addition, even more complicated states can exist such as the vortex entanglement state suggested by Nelson.[2] Finally, if the superconducting state is anisotropic, symmetries of the flux lattice other than triangular can exist.[8,9] However, decoration experiments using the high-resolution Bitter pattern technique appear to rule this out.[3]

In this paper we report mechanical measurements of flux-lattice melting in YBCO and BSCCO and show that their phase diagrams are indeed nonconventional. The experimental setup used is shown in Fig. 1. The basis for the experiment is the high-Q silicon oscillator technique

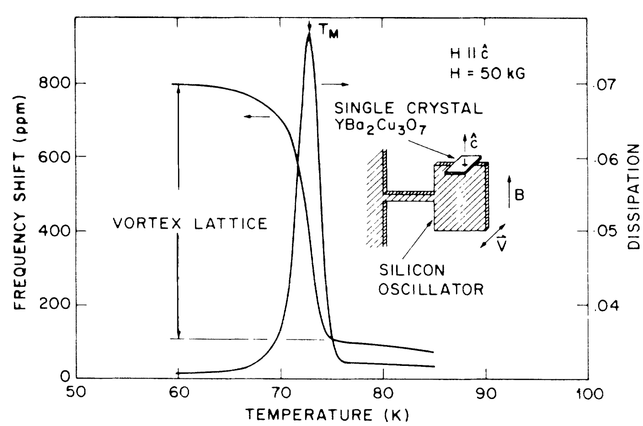

FIG. 1. The frequency and dissipation of the silicon oscillator as a function of temperature. The vortex mobility transition occurs at the peak in the attenuation, T_M. Below T_M, the vortex lattice is pinned and contributes to the elastic response. Inset: the geometry of the experiment. The oscillator was rotated in the plane of this drawing for other field orientations.

developed by Kleiman *et al.*[10] The oscillators were fabricated from p-type (110) silicon wafers 0.010 in. thick and patterned by means of photolithography and anisotropic etching. The silicon was coated with 100 nm of gold on both sides, which was used as part of a capacitive drive and detection scheme. The simplest beam-bending mode of the oscillator was used as indicated by the **V** in the drawing. The motion was driven self resonantly with use of a phase-locked loop. The structure had a Q of 10^5, a frequency of 2 kHz, and a frequency stability of 10^{-7} at 100 K. To avoid nonlinear effects, the amplitude at the end of the oscillator was kept well below 100 nm. This low amplitude represents an important difference between the oscillator technique and other similar measurements, particularly flux flow. In flux flow, there is no measurable signal until the flux lines have moved a macroscopic distance which then destroys the flux lattice that one wishes to observe. We typically dissipate 10^{-18} W in our measurement and can see effects of motionally induced melting at 10^{-16} W.

The superconducting crystal was epoxied to the silicon as shown. The single-crystal samples were grown as described elsewhere.[11] The YBCO crystals have a T_c of 87 K as measured by the Meissner effect, with a width of 3 K. The crystals contained many twins, usually in the form of long domains. The crystals were square flat platelets ~ 1 mm across and 0.1 mm thick, with the $\hat{c}$ axis perpendicular to the plane of the plates. While the upper critical filed, H_{c2}, was not measured on the crystals used, they are from batches from which several crystals were measured and found to give results similar to those reported by Worthington, Gallagher, and Dinger.[12]

The result of a typical measurement at fixed field as a function of temperature is shown in Fig. 1. Both the real and imaginary parts of the oscillator response are shown. Below the melting temperature, the bulk modulus of the vortex lattice contributes an additional stiffness to the oscillator. As the lattice melts, the response softens. This is accompanied by a peak in the dissipation. We define T_M, the melting temperature, as the location of the dissipation peak.

This identification can be justified as follows: For $T \ll T_M$ there is a well defined flux lattice which will remain pinned even in the presence of weak disorder. This will make the relaxation rate for compression of the flux lattice, τ, very long so that $\omega\tau \gg 1$ and the vortices will move with the underlying crystal. Thus, an elastic modulus appropriate to longitudinal sound in the vortex lattice will be added to the elastic response of the oscillator. For $T \gg T_M$, thermal fluctuations will dominate the motion of the vortices. In this limit the vortex liquid will rapidly relax ($\omega\tau \ll 1$) and the vortices will not contribute to the oscillator response. Near the melting temperature, T_M, the relaxation rate will be strongly temperature dependent. A peak in the dissipation will occur at $\omega\tau \sim 1$ which is our experimentally defined melting temperature. This identification is strengthened by comparison with similar measurements on a wide variety of other superconducting systems in which flux-lattice melting occurs at $T_c(H)$ with features identical to those shown here. This feature is *not* what is seen near flux-flow peaks[5] or depinning transitions.

Shown in Fig. 2 are these melting temperatures for YBCO as a function of magnetic field for $\mathbf{H}\perp\hat{c}$ and $\mathbf{H}\parallel\hat{c}$. For $\mathbf{H}\perp\hat{c}$, melting occurs at H_{c2} as in conventional three-dimensional superconductors. The critical-field slope defined by T_M agrees well with that measured at AT&T-Bell Laboratories and elsewhere,[12] and T_M extrapolates to $T_c(H=0)$. However, for $\mathbf{H}\parallel\hat{c}$ the flux-lattice melting line is suppressed *below* H_{c2} by ~ 3.6 K and it does *not* extrapolate to $T_c(H=0)$. In Fig. 2, H_{c2} for $\mathbf{H}\parallel\hat{c}$ is shown by the dashed line. We conclude that for this orientation the flux lattice melts into a vortex liquid before superconductivity is destroyed. This result is very similar to what is seen in two dimensions where flux-lattice melting can occur well below $T_c(H)$.[5] We suggest that the two-dimensional character of the system might be important for this behavior of the flux lattice. In this system the coherence length[12] is roughly equal to the spacing between the two-dimensional copper-oxygen planes and we suggest that the system could be in a crossover regime from a 3D system to a stack of decoupled 2D systems. Our experimental observations are consistent with our earlier flux-lattice decoration experiments[3] where at low temperatures and for $\mathbf{H}\perp\hat{c}$ we find only short-range (~ 1.5 lattice constants) positional order of the flux lines and at 77 K the images are consistent with a vortex-liquid state (i.e., no distinct, stable patterns of flux lines). We believe that the mechanical

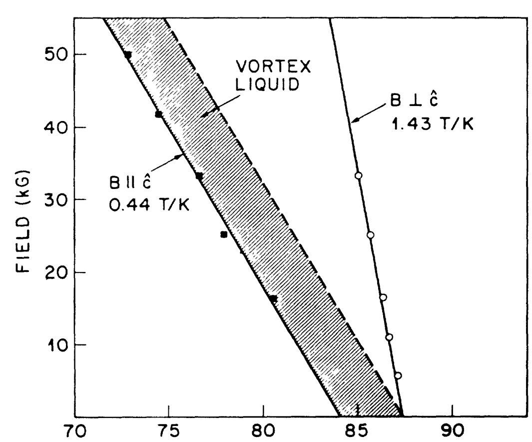

FIG. 2. The phase diagram for flux-lattice melting in YBCO. For $\mathbf{B}\perp\hat{c}$, the vortex mobility transition occurs at H_{c2}, as in three-dimensional superconductors. For $\mathbf{B}\parallel\hat{c}$, the vortex mobility transition occurs 3 K below H_{c2} (indicated by the dashed line). The shaded region is the vortex fluid.

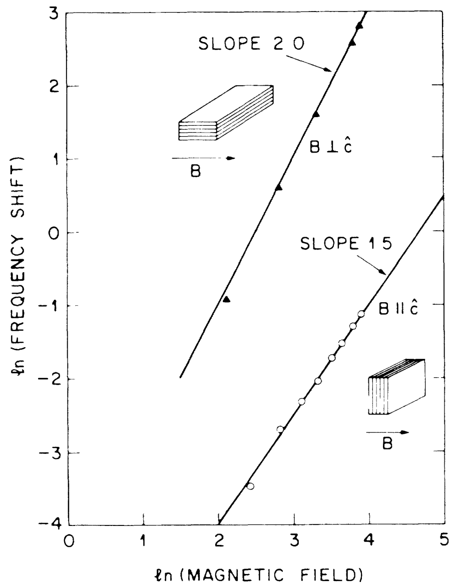

FIG. 3. The magnitude of the frequency shift for H∥ĉ and H⊥ĉ as a function of applied field.

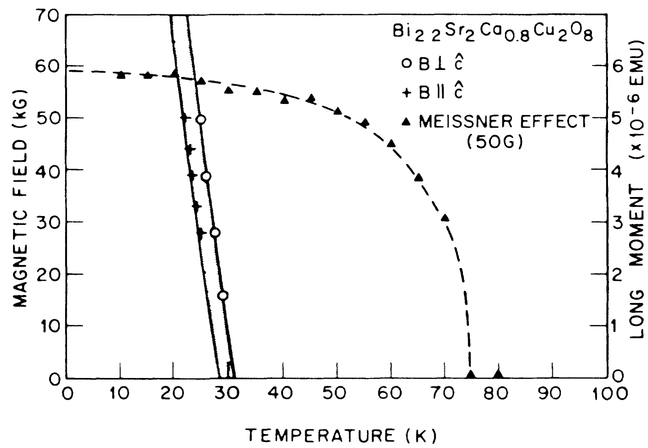

FIG. 4. The Meissner signal and the melting temperatures for single-crystal BSCCO for both field orientations.

measurements, in conjunction with the decoration experiments, provide evidence for melting and not merely flux line depinning.

Shown in Fig. 3 are the values of the frequency shift as a function of magnetic field for YBCO. We find that for $\mathbf{H}\perp\hat{\mathbf{c}}$ the response is significantly stiffer than for $\mathbf{H}\parallel\hat{\mathbf{c}}$ and that it varies as H^2. For $\mathbf{H}\parallel\hat{\mathbf{c}}$, we find that the resonse is soft and varies as $H^{1.5}$. For a conventional superconductor, the real response can be obtained from the total field energy and is proportional to H^2. For $\mathbf{H}\perp\hat{\mathbf{c}}$ we find this behavior. However, the weaker dependence for $\mathbf{H}\parallel\hat{\mathbf{c}}$ and the smaller magnitude suggests a much more disordered lattice with shorter-range order and weaker pinning consistent with the flux-lattice decoration images.[3]

We will now discuss BSCCO in which the coherence length is 4 times smaller[13] than the interplanar spacing and the effect on the melting temperatures is even more pronounced. Flux-lattice decoration experiments[14] in BSCCO at 4.2 K show similar patterns to those in YBCO. The flux quantum is $hc/2e$ and there is only short-range hexagonal order as was seen in YBCO. The mechanical measurements show an identical feature at the melting transition as was shown for YBCO. However, the transition temperature is much lower.

Shown in Fig. 4 is the phase diagram for flux-lattice melting in BSCCO. Also plotted is the Meissner signal for the same sample. Note that while bulk superconductivity sets in at 75 K the flux-lattice melting temperature is $\sim$30 K for both field orientations with $T_M(\mathbf{H}\parallel\hat{\mathbf{c}}) \le T_M(\mathbf{H}\perp\hat{\mathbf{c}})$. This strong reduction in the melting temperature for a material with a coherence length much shorter than the interplanar spacing suggests that this might be the relevant parameter in determining whether the flux-lattice melting is described by two- or three-dimensional physics. This conclusion has severe implications for potential applications. If, as is widely believed, the high transition temperatures are related to the two-dimensional character of the planes, then, as the interplanar coupling decreases, the flux-lattice melting temperature will go down making many applications impossible for very high-temperature superconductors. The similarity of the response for both field orientations is quite surprising given the strong anisotropy of the system. Varma[15] recently has made arguments based on entropy considerations in which the transition for $\mathbf{H}\parallel\hat{\mathbf{c}}$ is from two to three dimensions and for $\mathbf{H}\perp\hat{\mathbf{c}}$ from one to three dimensions with the expectation that the transition temperatures should be similar. Clearly this surprising result needs to be better understood.

There are other experiments which also suggest that the flux lattices can melt well below T_c. In ceramic samples of YBCO, the critical currents[16] have been found to be extremely low within 2 K of T_c. In BSCCO in finite magnetic fields, it has been found that J_c drops abruptly[17] at $\sim$30 K. In YBCO, SQUID noise is found to diverge below T_c.[18] We conclude that these observations are the result of the flux-lattice melting behavior that we have observed in the oscillator experiments and are consistent with our results. For example, we predict that flux noise will be much less for devices in which $\mathbf{H}\perp\hat{\mathbf{c}}$ in comparison with those for $\mathbf{H}\parallel\hat{\mathbf{c}}$ as is standard at the moment. Therefore, superconducting devices should be made from films with the $\hat{\mathbf{c}}$ axis in the plane.

At the moment it is difficult to give an unambiguous theoretical interpretation to our results. The vortex entanglement theory of Nelson[2] predicts new phases which are primarily low-field phases (near H_{c1}). It is unclear whether such phases could exist in the high-field regime where we see our transitions. The Kosterlitz-Thouless vortex unbinding theory is appealing.[6] However, the large suppression for $\mathbf{H} \perp \hat{\mathbf{c}}$ in BSCCO is hard to reconcile with this idea. The final possibility is simply that the small coherence lengths and high transition temperatures make thermal-fluctuation or Lindeman-induced melting much more likely.

We are currently studying larger samples at low fields to search for any additional low-field phases as suggested by Nelson. Neutron-scattering studies of flux-lattice melting would be most interesting as it would provide real structural information about these phases. A limitation of our technique is that we can only identify transitions between flux-lattice phases and obtain very little information about the structures of the phases themselves.

Finally, we note that in very recent measurements[19] on single-crystal thallium compounds with a T_c of 110 K that melting occurs at approximately 40 K, strengthening our surmise that this is a general feature of these compounds.

In conclusion, we have studied flux-lattice melting in single-crystal YBCO and BSCCO. Unlike in conventional three-dimensional superconductors which melt at T_c, these materials show flux-lattice melting well below $T_c(H)$. We argue that this is a general feature of these materials with severe implications for potential applications. At the moment we have no clear theoretical understanding of the phenomena, although some type of fluctuation-induced melting transition enhanced because of the short coherence lengths seems possible.

We would like to thank D. Nelson, C. M. Varma, B. Batlogg, B. van Dover, D. S. Fisher, W. F. Brinkman, B. I. Halperin, and A. P. Ramirez for numerous helpful discussions.

[1]M. Rice, Z. Phys. B **67**, 141 (1987), and references therein.

[2]D. R. Nelson, Phys. Rev. Lett. **60**, 1973 (1988), and references therein.

[3]P. L. Gammel, D. J. Bishop, G. J. Dolan, J. R. Kwo, C. A. Murray, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. **59**, 2592 (1987).

[4]M. Tinkham, Helv. Phys. Acta (to be published).

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

[6]D. S. Fisher, Phys. Rev. B **22**, 1190 (1980).

[7]D. R. Nelson, private communication.

[8]R. N. Kleiman, P. L. Gammel, E. Bucher, and D. J. Bishop, to be published.

[9]L. Gor'kov, to be published.

[10]R. N. Kleiman, G. K. Kaminsky, J. D. Reppy, R. Pindak, and D. J. Bishop, Rev. Sci. Instrum. **50**, 2088 (1985).

[11]L. F. Schneemeyer, J. V. Waszczak, T. Siegrist, R. B. van Dover, L. W. Rupp, B. Batlogg, R. J. Cava, and D. W. Murphy, Nature (London) **328**, 601 (1987); L. F. Schneemeyer, R. B. van Dover, S. H. Glarum, S. A. Sunshine, R. M. Fleming, B. Batlogg, T. Siegrist, J. H. Marshall, J. V. Waszczak, and L. W. Rupp, Nature (London) **332**, 423 (1988).

[12]T. K. Worthington, W. J. Gallagher, and T. R. Dinger, Phys. Rev. Lett. **59**, 1160 (1987).

[13]T. T. M. Palstra, B. Batlogg, L. F. Schneemeyer, and R. J. Cava, to be published.

[14]P. L. Gammel, L. F. Schneemeyer, J. V. Waszczak, and D. J. Bishop, to be published.

[15]C. M. Varma, private communication.

[16]M. A. Dubson, S. T. Herbert, J. J. Calabrese, D. C. Harris, B. R. Patton, and J. C. Garland, Phys. Rev. Lett. **60**, 1061 (1988).

[17]B. van Dover, L. F. Schneemeyer, E. M. Gyorgy, and J. V. Waszczak, to be published.

[18]J. C. Clarke, private communication.

[19]P. L. Gammel, D. J. Bishop, D. Ginley, and J. Schirber, to be published.