Experimental Evidence for a First-Order Vortex-Lattice-Melting Transition in Untwinned, Single Crystal $YBa_2Cu_3O_7$

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We report on current-voltage measurements in clean, untwinned $YBa_2Cu_3O_7$ single crystals with picovolt voltage sensitivity and millikelvin temperature resolution in magnetic fields ranging up to 7 T. We find evidence for a melting transition in the vortex lattice which is hysteretic in both temperature and magnetic field. The measured thermal and magnetic hysteresis widths are related by the local slope of the phase boundary. This strongly supports the picture that, in the clean limit, the melting transition of the Abrikosov vortex lattice is a first-order phase transition.

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The region of the magnetic phase diagram of the oxide superconductors where the vortex dynamics changes abruptly and where the resistivity drops very rapidly with decreasing temperature has proven to be both extremely interesting and very controversial [1]. The early and simplistic suggestions of a melting transition [2-4] have given way to more sophisticated theories of a vortex-glass phase transition [5]. Sensitive transport measurements in heavily twinned crystals have provided compelling support for such a phase transition as the explanation for the unusual dynamics [6]. Recent work has centered on the role of the dimensionality of the defects and its effect on the nature of the phase transition with a Bose-glass transition having been postulated for the case of spatially extended parallel defects [7].

In the vortex-glass picture [5] there exists a finite-temperature, second-order phase transition between the high-temperature vortex-liquid phase and the low-temperature vortex glass. The width of the vortex-glass critical region depends on the amount of disorder and, as shown both theoretically [5] and experimentally [6,8,9], as the amount of disorder decreases the width of the critical region becomes vanishingly small. In the clean limit (weak pinning of the vortex lines) the transition is expected to become first order [10]. This is then the true melting transition of the Abrikosov vortex lattice. Theoretical simulations [11] have found evidence for such a first-order transition in the clean limit. In the simulations, at the transition, the latent heat of melting is roughly $0.3k_BT$ per vortex line per layer for a magnetic field of order 10 T oriented normal to the layers with values of the parameters that model $YBa_2Cu_3O_7$ (YBCO). Transport and oscillator experiments [12,13] in untwinned YBCO have also seen tantalizing suggestions of a first-order transition, but no conclusive evidence has been found.

In this paper we report on measurements of the current-voltage response of untwinned YBCO single crystals with picovolt sensitivity as a function of temperature and magnetic field. We find that the resistive transition shows hysteretic behavior in both thermodynamic variables, temperature and magnetic field. The thermal and magnetic hysteresis widths are related by the slope of the phase transition line. This is strong evidence for a first-order melting transition in the vortex lattice in the mixed state of very clean type-II superconductors.

Our experimental apparatus is a SQUID picovoltmeter modified to allow operation at fields from 0 to 7 T and at temperatures from 4.2 to 100 K. The salient features of our apparatus as it applies to the present measurement are that, at 6 T, it has a voltage noise of $10^{-12}/\sqrt{\mathrm{Hz}}$ and a thermal reproducibility of ~5-10 mK. The temperature resolution in the short term is about 1 mK. This allows us to measure I-V curves with subpicovolt resolution in large fields with precise temperature resolution. As will be shown below, both features are necessary in order for us to observe the first-order features which are the central result of this paper.

Our samples are single crystals of $YBa_2Cu_3O_7$ grown by quenching the tetragonal phase during flux growth, mechanically separating them from the flux, and then oxygenating them without mechanical stress [14]. This method of producing untwinned crystals is known to produce very-high-quality, stress-defect-free samples as verified by x-ray diffraction studies. Samples from this batch have a sharp Meissner transition and exhibited complete diamagnetic shielding at low fields. Flux-lattice decorations [15] have found very-high-quality flux lattices with little evidence of strong pinning and defects. Sample 1 was a large, untwinned piece with dimensions of 0.33 mm×0.77 mm×0.02 mm. Sample 2 had similar dimensions, but consisted of two domains separated by a single twin. The c axis is along the small dimension as is usually the case in this system. For sample 1, the long dimension was along the a axis of the crystal and hence we measured ρ_{aa}. The samples were contacted on an a-b surface by evaporating ~1500 Å of gold and annealing for ~48 h at 400°C. Silver epoxy was then used to attach gold leads. The contacts were approximately 0.05 mm by 0.3 mm in size with a contact resistance of ~20 mΩ. The voltage leads were separated by ~0.25 mm with a standard four-terminal geometry used for the measurement. The normal-state resistivity of these samples was 50 $\mu\Omega$ cm for ρ_{aa} and 25 $\mu\Omega$ cm for ρ_{bb} as measured at 100 K. This is to be compared to a typical normal-state resistivity of a twinned crystal of $\rho_{ab}=65$ $\mu\Omega$ cm and is further evidence of the high quality of our sample. The zero-field resistive transition of sample 1 had the midpoint at 92.3 K and a width of 0.2 K (10%-90% criterion). Sample 2 had a similar transition temperature but with a slightly longer tail (~0.1 K) appearing below 5% of the normal-state resistance. By all the standards we can use to evaluate these crystals, these appear to be of very high quality with very little pinning disorder.

Figure 1 shows our main result. Plotted is the normalized linear response resistance for sample 1 as a function of temperature measured at 1.5 Hz with a current of 1 μA. A magnetic field of 6 T is applied parallel to the c axis. The hysteresis of ~30 mK in the resistance as a function of temperature is clearly visible. In addition to the overall transition, which is approximately 0.3 K wide, there can be seen a number of smaller, discrete jumps in the resistance which have widths at the limit of our temperature resolution of ~1 mK. We understand the data in Fig. 1 in the following way. The sample consists of a large number of Abrikosov crystallites which, due to large scale inhomogeneities, have melting temperatures spread out over ~0.3 K. Some of the crystallites are located in a region of the sample such that when they melt there is a discrete, visible jump in the resistance, while most are not. However, all crystallites supercool and superheat by the same amount, hence the uniform width of the hysteresis loop in temperature. The data in Fig. 1 are strong evidence for a first-order Abrikosov-vortex-lattice melting transition.

A number of experimental artifacts could mimic the above behavior. The most obvious are thermal gradients and thermal lags in our system. These have been carefully checked for and are not present at or above the level of 3 mK in our system. For example, the data shown in Fig. 1 are actually the result of three cycles of heating and cooling superimposed. Clearly the drifts and thermal lags in our system are significantly smaller than the effect which we see. Additionally, the zero-field resistive transition, which is as sharp as the transition shown in Fig. 1, is nonhysteretic. This rules out simple thermal effects. Similar results were obtained for sample 2.

Shown in Fig. 2 is a plot of the hysteresis width as a function of the applied field for both samples studied. As shown in the figure, with increasing field, the hysteresis widths increase monotonically and at our maximum field of 6.6 T we find a width of 38 mK for sample 1 and 18 mK for sample 2. Data were taken both with temperature sweeps, as shown in Fig. 1, and in fixed-temperature I-V curves with no change in the results. In addition, sweep rates which ranged from 0.1 to 1.0 K/h produced the same results. We understand the difference in the hysteresis widths in both samples as due to a different de-

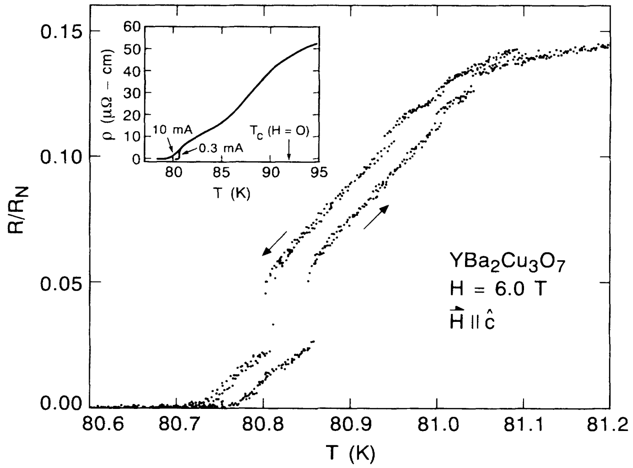

FIG. 1. Normalized linear response resistance as a function of temperature for sample 1 at an applied field of 6 T for data taken upon heating and cooling using the SQUID picovoltmeter. Note the hysteresis in the response. Inset: Data taken over a wider range using conventional electronics. The data shown in the inset, taken using conventional electronics, and the other data shown both here and in the other figures using the SQUID, were taken in two different runs and there is a ~0.1-K shift in the temperature scales.

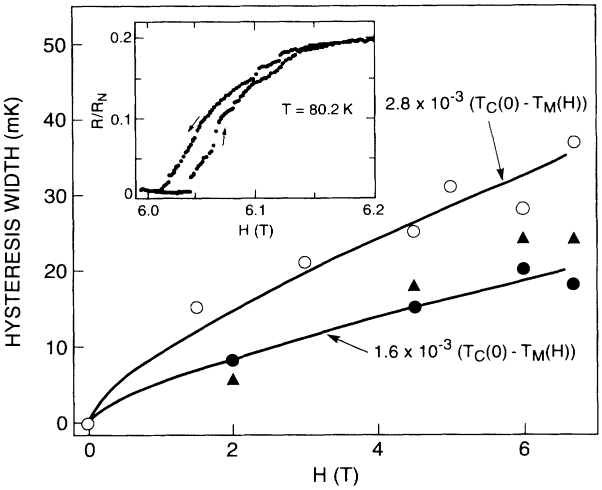

FIG. 2. Widths of the thermal hysteresis loops for sample 1 (open circles) and sample 2 (solid circles) as a function of applied field as measured with low applied currents. Also shown are the independently measured field hysteresis widths on sample 2 (solid triangles) converted using the slope of the phase boundary (see text). The hysteresis widths are roughly proportional to the suppression of the melting transition, $T_m(H)$, with field, as indicated by the solid lines. Inset: Magnetic-field sweep at a constant temperature of 80.2 K for sample 2 using a measuring current of 5 μA. Note the hysteresis in field.

gree of internal disorder. Other attempts [12] at observing the hysteretic behavior in untwinned single crystals using conventional electronics have failed, although those samples show a similar jump in the linear response resistance at the melting transition.

To provide further independent support for the intrinsic nature of the hysteresis we observe in the thermal sweeps at a fixed field, we have performed magnetic-field sweeps at a constant temperature. A result for sample 2 is shown in the inset of Fig. 2. We again found a noticeable hysteresis when crossing the vortex melting line; however, this time it was in field, not temperature. Changes in the sweep rate by a factor of 2 did not affect the measured hysteresis widths. The measured field hysteresis should be related to the thermal hysteresis by the slope of the phase transition line at a given field and temperature, since this line sets the scale between magnetic and thermal energy. We have calculated this local slope by fitting the phase boundary with a smooth polynomial. The result is a slope which goes from 1.8 K/T at $T=87$ K to 1.4 K/T at $T=80$ K. Shown in Fig. 2 are the magnetic hysteresis widths, measured on sample 2, converted by the slope of the phase boundary at each temperature and plotted on the temperature scale. The agreement is excellent. This constitutes independent and definitive evidence for a first-order melting of the vortex lattice in this system.

In Fig. 2 we have drawn solid lines through the data which are proportional to the distance between $T_m(H)$ and $T_c(0)$. We note that the temperature scale for the suppression of the melting transition (the width of the vortex-fluid regime) and the hysteresis width vary similarly with changing magnetic field for both samples although the prefactors are different. Shown in Fig. 3 is the current dependence of the hysteresis widths as measured in sample 1. Clearly, as the measuring current is increased, the hysteresis widths decrease. Even for the data at 6 T, the width is likely to be small in the 1-mA current ranges typically used for experiments with conventional electronics. A similar suppression of the hysteretic behavior with increasing current was observed in sample 2 in both field and temperature sweeps. The current scale generally increases with field in the same fashion as the hysteresis width, although our measurements are not sufficiently accurate to scale them as we have in Fig. 2.

We show in Fig. 4 the phase diagram for the melting transition as a function of applied magnetic field for sample 1. Results for sample 2 are the same and are not shown. To within our errors, our phase line appears to agree with that obtained by Farrell, Rice, and Ginsberg [13] as measured with a low-frequency mechanical oscillator on similar, untwinned YBCO single crystals.

The results presented here are very different from those found earlier in a similar measurement in strongly twinned YBCO single crystals [6]. In those measurements good agreement was found with the vortex-glass phase transition model [5]. As expected for a second-order phase transition, no hysteresis was observed. The present measurements show the importance of the twins in destroying the long-range positional correlations of the Abrikosov vortex lattice. Such an effect has been seen in decoration experiments but the relevance of such low-field measurements to these high-field ones is open to question [16].

In the vortex-glass scenario, for strong pinning, there is a finite-temperature, second-order phase transition at T_g. As the temperature is lowered from above T_g to the transition, the theory postulates a vortex-glass correlation length ξ_{vg} which diverges as $\xi_{vg} \sim (T-T_g)^{-\nu}$. The diverging length defines a current scale J_{sc} for linear response via the relation $\xi_{vg} = (ck_BT/\Phi_0 J_{sc})^{1/2}$. At $T = T_g$ the response becomes non-Ohmic at all current scales. Therefore, non-Ohmic response at all current

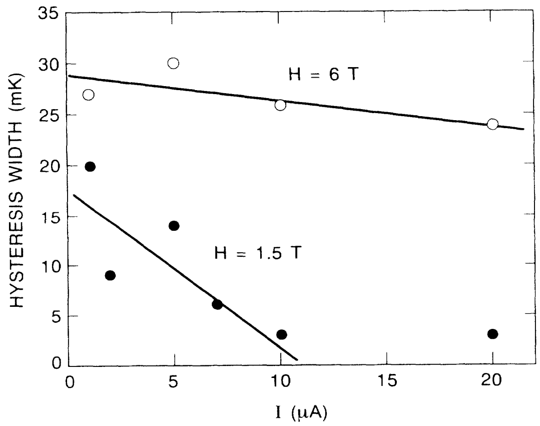

FIG. 3. The current dependences of the widths of the hysteresis loops for sample 1 for two different applied fields. The solid lines are guides to the eye.

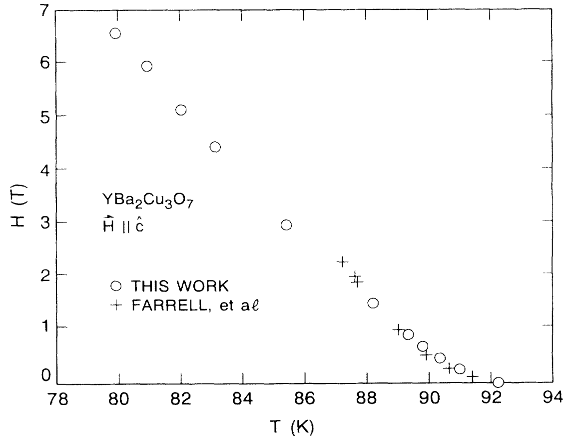

FIG. 4. The magnetic phase diagram for the first-order transition for sample 1 (open circles). For comparison, the data of Farrell, Rice, and Ginsberg (crosses) are also shown.

scales is the signature of the vortex-glass transition. However, there is another length scale in the problem, the Abrikosov vortex lattice positional correlation length ξ_{vl}. As the temperature is lowered through the vortex-liquid regime, this length also grows. As shown by Larkin and Ovchinokov [17], ξ_{vl} will always saturate at some finite value due to random pinning. The vortex-glass critical region occurs for temperatures such that $\xi_{\mathrm{vg}} > \xi_{\mathrm{vl}}$. As the samples get cleaner, and with the absence of pinning due to twins, ξ_{vl} gets longer and the vortex-glass critical region should become vanishingly small. As shown here, in this limit the transition ultimately becomes first order. In addition to experiments using the picovoltmeter, we have also measured our sample using conventional electronics. These data for 6 T are shown as an inset in Fig. 1 for sample 1. The sample's nonlinear response has been most extensively studied at 6 T and will be discussed here, but the response at other fields is similar. As seen in other measurements and shown in the inset, the shoulder in the resistivity at 81.1 K in Fig. 1 is where the resistance as measured with microvolt sensitivity becomes very nonlinear. Typical measuring currents in that regime are 1 mA. One finds that at all measuring levels, within the capability of conventional electronics, the shoulder becomes sharper as the measuring current is reduced. With such electronics, it is very difficult to detect the hysteresis. However, as we have shown above, if one probes the system with picovolt sensitivity, then one can readily see a resistance which is linear in current and is hysteretic with temperature as shown in Fig. 1. All the data shown in Fig. 1 were checked to be linear in response (at the picovolt level) over two decades in current and over the temperature range shown.

The question arises: Where is the vortex-glass transition which should be non-Ohmic at all currents, even those probed by the SQUID? We find that intrinsic nonlinearity in the I-V curves appears for $R/R_N \le 1\%$. For the data shown in Fig. 1, our response becomes non-Ohmic at approximately 80.7 K, which is at the lower end of the region of the first-order melting transition. Thus, for our sample, $T_g \le T_M$. This result is consistent with recent measurements of Worthington *et al.* [18], who see evidence for two separate transitions in samples where the pinning has been enhanced by radiation damage, with the higher-temperature one being interpreted as the remnant of the first-order transition we see clearly here and the lower being the vortex-glass transition which is where true superconductivity occurs.

In conclusion, we have reported on I-V curves measured on very clean, untwinned YBCO crystals. We find that the resistance is hysteretic with both temperature and field. The thermal and magnetic hysteresis widths are related by an energy scale set by the slope of the phase transition line. The hysteresis becomes more pronounced at higher fields but is easily reduced or even eliminated by very modest measuring currents. This provides strong evidence supporting the idea that in the clean limit the Abrikosov vortex lattice melts via a first-order phase transition.

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