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Static and Dynamic Vortex Transitions in Clean $\text{YBa}_2\text{Cu}_3\text{O}_7$

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*The evidence establishing first order melting of the vortex lattice in clean
 $\text{YBa}_2\text{Cu}_3\text{O}_7$ is reviewed. Dynamic transitions in the moving vortex system
are demonstrated experimentally through resistivity and magnetization
measurements.*

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1. INTRODUCTION

The equilibrium phase diagram of the vortex state of high temperature superconductors is remarkably rich in its phases and phase transitions. Liquid, lattice, and glassy phases have been postulated theoretically, with first or second order transitions separating the liquid from the lattice or glass, respectively.¹ Experimentally, these transitions have been probed with resistivity, magnetization, and very recently, calorimetric measurements. Each of these probes examines a different aspect of the transition, and all are needed to develop a full picture of the vortex behavior. The measured voltage in a transport measurement is sensitive only to *moving* vortices, proportional to the product of their number and velocity. In contrast, magnetization and calorimetric experiments probe vortices nominally at rest. Provided the system is reversible, these experiments probe *equilibrium* behavior, giving the vortex density and latent heat or heat capacity, respectively. This quantitative information is necessary to establish the thermodynamic character of the transition.

In addition to equilibrium phase transitions, there is a second class of transitions which occurs in *moving* vortex systems. Moving vortices are dissipative, so that energy is not conserved and cannot be used to define the stability of phases or the order of phase transitions between them. Nevertheless, there are well defined steady states of vortex motion which can be distinguished in numerical simulations²⁻⁵ and in experiments.^{6,7}

Dynamic transitions between these states can be studied with the same experimental tools used to study equilibrium phase transitions, resistivity and magnetization. A growing variety of dynamic states have been identified, loosely categorized into plastic, where the neighbors change throughout the motion, and elastic, where the neighbors remain the same. In general, there is a strong interplay between dynamic transitions and equilibrium phase transitions, because the character of the motion is determined in part by the same thermal fluctuation and interaction energies among vortices and pin sites which control the equilibrium phases.

In this paper we review recent resistivity, magnetization and calorimetric experiments which probe and establish the equilibrium first order vortex melting transition in clean $\text{YBa}_2\text{Cu}_3\text{O}_7$. We then apply these tools to dynamic transitions between plastic and elastic flow which naturally extend the equilibrium phase diagram.

2. DEVELOPMENT OF THE SHEAR MODULUS

The shear modulus is the fundamental elastic property distinguishing liquids from solids. In the liquid a given vortex can be pinned while its un-

pinned neighbors are free to slide past it at a rate governed by the viscosity of the liquid. In the solid, the shear viscosity is replaced by shear elasticity, which prevents neighbors of a pinned vortex from moving as long as the driving force is less than the elastic shear restoring force. Thus the same set of pinning sites is more effective in pinning the solid than it is the liquid.

In transport experiments, the enhanced pinning of the solid appears as a drop in the resistivity as the temperature is lowered below the freezing line. If the transition is first order, the drop is expected to occur suddenly, simultaneously with the discontinuous onset of the shear modulus. In addition, the resistivity drop may display hysteresis if the first order transition itself is hysteretic. The resistivity drop is largest at low currents, below the critical current of the solid, where it reflects the velocity difference between the moving

Fig. 1 ρ vs θ above and below the vortex freezing transition for vortices flowing in channels created by twin boundaries. The insets illustrate the channel geometry and velocity profile in the liquid state, and the effect of tilting the field away from the plane of the channel walls.

of the solid, where it reflects the velocity difference between the moving

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liquid and the stationary solid. An analysis of the resistive melting behavior at low current is given in Section 3.

At driving currents which exceed the critical current of the solid, the onset of the shear modulus manifests itself in a different way. The drop in resistivity between liquid and solid is reduced because the solid is no longer at rest. Moreover, the nature of the moving state itself may be quite different in the liquid and solid. This dynamic difference can be seen in channel geometries where the channel width is greater than the velocity correlation length of the liquid due to its viscosity, but less than that of the solid due to its shear elasticity.⁸ Because the vortices are lines which interact strongly with the channel walls for parallel fields, the *anisotropy* of the resistivity in the driven state is opposite for the liquid and solid. This is illustrated in Figure 1 for channels formed by twin boundaries in a single crystal of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO).⁸

For the field parallel to the twin boundary walls, the vortices in the walls are strongly pinned and move with small or zero velocity. For the liquid, the viscous correlation length is less than the channel width, leaving many unpinned vortices which move freely. If the field is tipped sufficiently far from the parallel direction, even vortices which are near the center of the channel intersect the walls and feel a pinning force. Thus the parallel direction is a maximum in the resistivity of the liquid. For the solid, where the velocity correlation length due to shear elasticity is longer than the channel width, the vortices feel the pinning effect of the walls everywhere in the channel. The dominant feature is the strength of the pinning force exerted by the walls, which falls off as the field is tipped away from the parallel direction. Thus in the solid, the resistivity is a minimum for the field parallel to the walls. This experiment illustrates dramatically a *dynamic* difference between the liquid and solid. Obvious experimental consequences mark the replacement of shear viscosity with shear elasticity, indicating that a *dynamic* transition has occurred. Further examples and discussion of dynamic transitions at high transport current are given in Section 5.

3. LOW CURRENT MEASUREMENTS OF MELTING

The melting transition as seen in transport experiments is illustrated in Figure 2 for a clean untwinned crystal of YBCO in magnetic fields of 0 - 8 T perpendicular to the CuO_2 planes.⁹ The superconducting transition is marked by the sharp drop to zero resistivity at zero field. In finite field, the resistivity is rounded, due to fluctuations which suppress the second order phase transition between the vortex and normal states which is expected from mean field theory. At the low driving currents used in Figure 2, vortex motion in the liquid phase occurs easily, while the enhanced pinning of the solid effectively immobilizes the vortices. The freezing transition appears as a sharp drop to zero resistivity, labeled T_m in Figure 2. From the position of the drop, a phase diagram for the melting line in the H-T plane can be constructed as shown in the inset. The melting line determined

from the resistive drop is remarkably reproducible, even for samples of different quality grown in different laboratories. The melting curve determined from Figure 2

Fig. 2 ρ vs T for an untwinned $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ crystal for $H \parallel c$. Inset: Phase diagram determined from resistivity.

is in excellent quantitative agreement with the earlier work of Safar *et al.*¹⁰

The sharpness of the resistive drop at T_m , strongly suggests a first order freezing transition. This suggestion is further reinforced by substantial hysteresis which is observed in both the temperature and field dependence of the transition. Figure 3

shows the development of hysteresis in field as a function of melting temperature. The largest hysteresis occurs at intermediate fields, about 5 T

Fig. 3 Field dependence of the resistivity, showing hysteresis behavior near H_m at several temperatures.

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for the crystal shown. At lower and higher fields, the hysteresis decreases, though it is evident wherever an incomplete sharp drop in the resistivity occurs.

Figure 4 shows the partial hysteresis loops which occur for another untwinned crystal if the field is reversed before the freezing or melting transition is complete. There are two features in the partial loops which are typical of first order phase transitions. First, the sharp freezing transition occurs at slightly different fields for successive trials. This is characteristic of first order nucleation which occurs

locally wherever the thermodynamic conditions are fulfilled and an appropriate fluctuation is present. Because each field sweep in Figure 4 has a different history, nucleation may occur at different fields.

The second feature characteristic of first order transitions is the similarity in the shape of the partial loops to that of the full loop. This occurs in first order transitions because the fraction of material which transforms in a partial transition follows the same thermodynamic pattern as the full system. In Figure 4 the full loop is characterized by strong asymmetry, with sharp freezing and gradual melting. The partial loops show the same pattern. Following each of the three reversals on incomplete freezing and the two reversals on incomplete melting, the resistivity abruptly switches from one branch of the hysteresis curve to the other. The alternate behavior, retracing the same curve on reversing the field after a partial transition, would have indicated that the hysteresis is not associated with a first order transition.¹¹

Fig. 4 Field dependence of the resistivity at $T=82.0\text{K}$ showing evidence for hysteresis sub-loops in the magnetic field sweep cycles.

4. THERMODYNAMIC MEASUREMENTS OF MELTING

Although transport measurements accurately locate the position of the melting line and strongly suggest a first order transition, they cannot provide definitive proof of such a transition because they are sensitive only to moving vortices rather than to those in thermal equilibrium. Two thermodynamic consequences of a first order transition can provide such proof: a discontinuity in the magnetization $M = \partial F / \partial H$ or in the entropy $S = -\partial F / \partial T$ (where F is the free energy) at the transition. In principle, the former can be seen in a magnetization experiment and would imply a discontinuity in the vortex density at melting, and the latter would appear in a calorimetric experiment as a latent heat $T\Delta S$. Conventional wisdom held that both

effects were too tiny to be seen in experiments, especially given the small size of the relatively clean crystals which show sharp melting behavior in transport. However with careful experimental technique, both experiments have now been successfully carried out.

The magnetization at 84 K near the melting field is shown in the inset

Fig. 5 Jump in magnetization at the melting field. Inset: reversible magnetization behavior

of Figure 5, for up and down field sweeps.¹² Within the experimental noise of ~ 0.05 gauss, the magnetization is reversible. There is a well defined increase in magnetization at the melting field as the sample enters the vortex liquid phase. This jump is more obvious when a linear extrapolation of the solid phase magnetization is subtracted, indicated by the dashed line in the inset. The difference in

magnetization between the solid and liquid is shown in the main panel of Figure 5 for several temperatures.

Simultaneous measurements of the magnetization and resistivity¹³ demonstrate that the two transitions occur together throughout the measured range in the H - T plane. Magnetization measurements like those in Figure 5 and the earlier work of Liang *et al.*¹⁴ provide the first thermodynamic evidence that the melting transition in YBCO is indeed first order.

The magnetization of the liquid is larger than the magnetization of the solid, indicating that the liquid has a higher density of paramagnetic vortex lines. Melting transitions with denser liquids than solids are unusual in condensed matter. Ice is the most familiar example, with a density discontinuity of order 10%. In YBCO, the vortex density difference is much smaller, of order 5×10^{-6} . Ice-like melting is required by the slope of the melting line. This can be seen most directly from the Clausius-Clapeyron equation for magnetic systems, $dH_m/dT = -\Delta S/\Delta M$, where H_m is the melting field and ΔS and ΔM are the entropy and magnetization discontinuities, respectively. The melting line has negative slope, requiring ΔS and ΔM to have the same sign. Thermodynamics requires the high temperature phase to have the higher entropy, so the magnetization of the liquid must also be higher than that of the solid. The thermodynamic equivalence of negative slope of the melting line and a denser liquid phase means that both effects have the same physical origin. They arise from the relatively weak spatial dependence of the interaction between vortex lines,¹⁵ $\sim \ln(\lambda/r)$ for the fields of interest here.

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The entropy discontinuity on melting can be derived from the magnetization experiments via the Clausius-Clapeyron equation using the measured slope of the melting line and the magnetization discontinuity. The results are shown in Figure 6. The entropy jump is approximately constant below 88 K. At higher temperature, the entropy falls sharply, and neither the magnetization nor entropy jumps can be seen above 90 K. This is at

Fig. 6 The temperature dependence of the jump in the magnetization and the entropy.

least 2 K below T_c , and it is an interesting open question why the first order transition disappears. It is possible that the melting transition is re-entrant, turning to follow H_{c1} at low field.¹⁶ Alternatively the solid phase may become glassy at low fields as the number of vortices decreases while the

number of residual pinning sites remains constant. The freezing transition to a glass is expected to be second order or continuous,¹⁶ which would require the first order discontinuities to disappear.

The magnetic behavior of YBCO at melting shows important similarities and differences from that of BSCCO. Despite much weaker coupling between CuO_2 layers and consequent greater flexibility of the lines, BSCCO displays approximately the same entropy jump on melting at low temperature as YBCO.^{17,18} However, near T_c the entropy jump in BSCCO increases dramatically, in contrast to its decrease and eventual disappearance in YBCO. At low temperatures, BSCCO displays a critical point where the first order transition is suppressed. There is no magnetic evidence for such a critical point in YBCO, though the high melting fields in YBCO limit SQUID measurements to the temperature range above about 80 K. Transport measurements of the melting line indicate a rounding of the drop to zero resistivity as shown in Figure 3 and elsewhere,¹⁹ which may indicate the existence of a critical point.²⁰

The latent heat measured in calorimetry provides independent thermodynamic evidence of first order melting. This experiment requires unusual sensitivity, because the latent heat amounts to a tiny fraction of the heat content of the superconductor itself at the vortex melting temperature. Nevertheless, sufficiently sensitive differential techniques have been developed by Schilling et al.²¹ and applied to a crystal on which the magnetization jump had also been measured.²² The entropy jump measured from magnetization is shown in Figure 6 and has recently been confirmed with conventional calorimetric techniques.²³ The excellent thermodynamic

agreement of the vortex density and entropy discontinuities at melting dramatically confirms the first order transition first suggested by theory and by transport measurements.

5. TRANSITIONS IN MOVING VORTEX SYSTEMS

In moving vortex systems energy conservation is destroyed by dissipation, precluding the use of equilibrium concepts based on the thermodynamic potentials to analyze the stability of phases and the order of phase transitions. Nevertheless, there is interesting steady state behavior driven by the same vortex-vortex and vortex-pin site interactions which control the equilibrium states. In equilibrium systems without pinning the stable solid phase is the lattice, where elastic forces among the vortices induce long range spatial coherence in the vortex positions. In moving systems without pinning the same elastic forces induce *elastic* motion which preserves long range spatial coherence. Pinning can destroy elastic flow by introducing local forces opposing the motion. If the driving and pinning forces are strong enough, long range spatial coherence is destroyed by rifts which appear in the moving system, separating regions of vortices which move at different average velocities. The neighbors change across these rift lines, and the motion is therefore *plastic*. In terms of spatial coherence, the elastic and plastic moving states are natural extensions of the lattice and glassy vortex states in equilibrium.

Figure 7 shows simultaneous resistivity and magnetization measurements in a field of 4.2 T perpendicular to the CuO_2 planes for a clean YBCO crystal which undergoes an equilibrium first order melting transition.¹³ At low driving current, where the lattice is stationary, the usual sharp drop in the resistivity on freezing is observed. Simultaneously, the magnetization discontinuity is observed, indicating a first order change in the vortex density. At higher current, the vortex lattice begins to move, as indicated by the finite resistivity below T_m . Remarkably, the discontinuity in magnetization is unchanged, indicating that the same vortex density change occurs in the moving system as at rest. At the same time, the discontinuity in resistivity between moving liquid and moving solid is significantly reduced with increased driving force, almost disappearing at the highest current (see inset of Figure 8).

The dynamic resistive and magnetic data indicate plastic and elastic motion of the liquid and solid states. The liquid state always moves in plastic flow, the spatial coherence being destroyed by thermal fluctuations even in equilibrium. At high driving forces, the small difference between the liquid and solid resistivity indicates that pinning has little effect, as expected for the small pinning forces in this relatively clean sample. This is the limit in which the elastic forces of the solid are expected to induce elastic flow. The vortex density discontinuity in the moving system is the same as in equilibrium within experimental uncertainty, consistent with the long range spatial coherence expected for a moving lattice. At lower driving current, the resistivity of the solid falls below that of the liquid, indicating

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Fig. 7 Simultaneous temperature dependent magnetization and resistivity measurements near the melting transition. 13

that pinning forces are effective in opposing the driving force.

The resistive and magnetic data may be summarized on an experimental dynamic phase diagram for clean systems as shown in Figure 8. The

Fig. 8 Experimental dynamic phase diagram for clean $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. The vertical axis gives $J\Phi_0$, the driving force/unit length on a single vortex. Inset shows the difference in magnetization and resistivity at the onset and completion of the dynamic transition.

horizontal axis represents the equilibrium behavior at zero driving force. The approximately vertical lines mark the positions of the sharp resistive and magnetic changes at finite driving force. In equilibrium, these lines define a coexistence region between liquid and lattice. In the moving system, they represent the onset and completion of the dynamic transition from plastic to elastic flow. For clean systems this dynamic transition is controlled by thermal fluctuations only, as indicated in Figure 8. For systems with sufficient quenched disorder that the

equilibrium lattice is replaced by a glass, the motion at low driving force is

expected to be plastic, while that at high drive is expected to be elastic.^{3,4} This implies a complex dynamic phase diagram where the plastic-to-elastic transition is governed by the balance between driving and pinning forces as well as by thermal fluctuations. The clear magnetic and resistive signatures of the dynamic transition demonstrated here for clean systems provide experimental tools for exploring this potentially rich dynamic behavior.

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REFERENCES

1. G. Blatter *et al.*, *Rev. Mod. Phys.* **66**, 1125 (1994).
2. H. J. Jensen *et al.*, *J. Low Temp Phys* **74**, 293 (1989).
3. A. Koshelev and V. Vinokur, *Phys. Rev. Lett.* **73**, 3580 (1994).
4. T. Giamarchi and P. Le Doussal, *Phys. Rev. Lett.* **76**, 3408 (1996).
5. F. Nori, *Science* **271**, 1373 (1996).
6. M. J. Higgins and S. Bhattacharya, *Physica C* **257**, 232 (1996).
7. J. Harris *et al.*, *Phys. Rev. Lett.* **74**, 3684 (1995).
8. W. K. Kwok *et al.*, *Phys. Rev. Lett.* **76**, 4596 (1996).
9. W. K. Kwok *et al.*, *Rev. Lett.* **72**, 1092 (1994).
10. H. Safar *et al.*, *Phys. Rev. Lett.* **69**, 3370 (1992).
11. W. Jiang *et al.*, *Phys. Rev. Lett.* **74**, 1438 (1995).
12. U. Welp *et al.*, *Phys. Rev. Lett.* **76**, 4809 (1996).
13. J. A. Fendrich *et al.*, *Phys. Rev. Lett.* **77**, 2073 (1996).
14. R. Liang *et al.*, *Phys. Rev. Lett.* **76**, 835 (1996).
15. David Nelson, Proceedings of the Indo-US Symposium on Liquid Crystals and Supramolecular Order, Bangalore, India, Jan. 2-5, 1996, to appear in *Molecular Crystals and Liquid Crystals*.
16. D. S. Fisher *et al.*, *Phys. Rev. B* **43**, 130, (1991).
17. E. Zeldov *et al.*, *Nature (London)* **375**, 373, (1995).
18. T. Hanaguri *et al.*, *Physica C* **256**, 111 (1996).
19. W. K. Kwok *et al.*, *Phys. Rev. Lett.* **69**, 3370 (1992).
20. H. Safar *et al.*, *Phys. Rev. Lett.* **70**, 3800 (1993).
21. A. Schilling and O. Jeandupeaux, *Phys. Rev. B* **52**, 9714 (1995).
22. A. Schilling *et al.*, *Nature (London)* **382**, 791 (1996).
23. M. Roulin, A. Junod, E. Walker, and A. Erb, these proceedings.