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Evidence for two vortex phase transitions in $Tl_2Ba_2CaCu_2O_8$ thin films

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Consistent with various models of quasi-two-dimensional behavior in strongly anisotropic superconductors, evidence for two separate vortex phase transitions is observed for an epitaxial thin film of the high-temperature superconductor $Tl_2Ba_2CaCu_2O_8$. Based on electric field as a function of current density $E(J)$ behavior, the temperature dependencies of the resistivity $\rho(T)$ and dc magnetization $M(T)$, as well as the frequency dependence of the ac susceptibility $\chi_{ac}(f)$, there is strong evidence for a lower-temperature vortex-solid (glass) phase transition to a true superconducting state that is characterized by a loss of linear resistivity. In addition, $\rho(T)$ and $M(T)$ results provide evidence for a higher-temperature transition, or crossover, at which the vortex dissipation processes undergo distinctive changes. This transition or crossover may be associated with the coupling of unbound two-dimensional vortices (pancake-vortex liquid) into a liquid of vortex lines as temperature decreases.

The Tl-based high-temperature superconductor $Tl_2Ba_2CaCu_2O_8$ (Tl-2212) is one member of a group of high-temperature superconductor (HTS) systems that exhibit strong anisotropy. Various magnetic phase diagrams have been proposed for the anisotropic HTS, in both the clean and disordered limits, and a feature common to most of these models is a two-step transition from a true superconducting state to the normal state at magnetic fields large enough for the superconductor to show quasi-two-dimensional behavior.[1–3] In the model of Fisher, Fisher, and Huse, and that of Feigel'man, Geshkenbein, and Larkin, the quasi-two-dimensional regime is entered for fields larger than H_0 at which the wavelength of fluctuations along the c axis is less than the interplanar spacing d (Ref. 2) or, alternatively, the interaction energy between vortices in different layers is less than the dislocation mediated two-dimensional melting temperature of an individual layer.[1] Feigel'man *et al.* propose that what is a single transition below H_0 will become two separate transitions; a higher-temperature vortex-melting transition T_m and a lower-temperature vortex-glass transition T_g. Previous results in $Bi_2Sr_2CaCu_2O_8$ (Bi-2212) have suggested that the higher-temperature transition is merely a dimensional crossover,[4] whereas the results presented here suggest that this transition in Tl-2212 may actually be critical in nature.

Epitaxial c-axis-oriented Tl-2212 thin films were grown by off-axis magnetron sputtering of Ba-Ca-Cu-O onto (100) $LaAlO_3$ substrates followed by post-deposition annealing in air at about 850 °C in the presence of a powder mixing of $Tl_2Ba_2Ca_2Cu_3O_{10}$ and Tl_2O_3.[5] These films routinely have values of the superconducting transition temperature T_c between 102 and 105 K with low microwave surface resistance (150 $\mu\Omega$ at 77 K and 10 GHz) and high critical current density J_c ($>10^6$ A/cm^2 at 77 K). The recent measurement of $J_c>10^6$ A/cm^2 at 80 K over a full 1.8 m long, 12 μm wide line demonstrates the high quality and uniformity of these films.[6] The T_c of the 8000Å thick Tl-2212 thin film used in this study was approximately 102 K with a transition width of about 3 K when measured by χ_{ac} at 2 MHz. The film was patterned into a bridge with a length of 3200 μm and a width of 110 μm, and $\rho(T)$ and $E(J)$ measurements were performed with H applied perpendicular to the surface of the film by a method described previously.[7] A piece of the same Tl-2212 film was employed in dc magnetization as a function of temperature $M(T)$ measured both on warming after zero-field cooling (ZFC) and on cooling in an applied field (fcc). In addition, the temperature-dependent ac susceptibility χ_{ac} response was studied as a function of frequency.[7]

Figure 1 shows the strongly field-broadened $\rho(T)$ curves for a Tl-2212 thin film in fields ranging from 0 to 50 kOe.

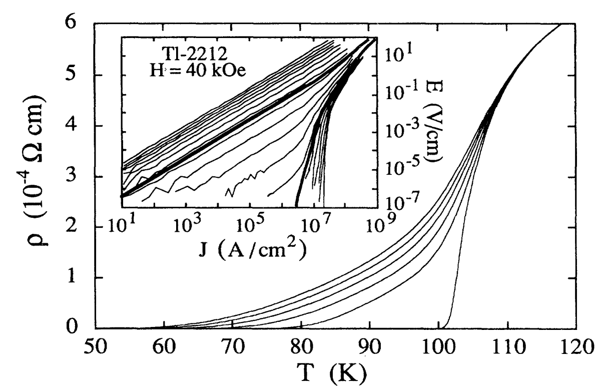

FIG. 1. Resistivity as a function of temperature in applied magnetic fields ranging from 0 to 50 kOe for an epitaxial Tl-2212 thin films. The inset shows electric field as a function of current density $E(J)$ isotherms at $H=40$ kOe for temperatures ranging from 10 K at the lower right to 100 K at the upper left in 5 K increments. The heavy lines in the inset correspond to the isotherms associated with the upper and lower transitions at T_{cH} and T_{cL}, respectively.

The transition width, as measured from the initial downturn of the resistive transition near T_c to the zero resistance point, is about 10 K for $H=0$ kOe and increases to more than 50 K for $H=50$ kOe. The inset of Fig. 1 is a plot of $E(J)$ isotherms ranging from $T=10$ K at the lower right to $T=100$ K at the upper left in increments of 5 K. These $E(J)$ curves are different than those observed for strongly disordered samples of $YBa_2Cu_3O_7$ (YBCO), where essentially only two types of curves exist: one type, at temperatures above the vortex-glass–phase-transition temperature T_g, having positive curvature and behavior that crosses from Ohmic to power law with increasing J, and a second type, observed at temperatures below T_g, exhibiting only negative curvature.[2,8] For Tl-2212 there are, in addition to these two types of curves, isotherms at intermediate temperatures which have an inflection point and thus contain regions of both positive and negative curvature. For epitaxial thin films of YBCO, each $E(J)$ curve can be scaled about a single temperature onto one of two universal curves using a universal scaling function.[2,8] In contrast to this, for Tl-based HTS materials, neither in this, nor previous studies, has it been possible to scale the $E(J)$ data about a single critical temperature.[9] However, the lower-temperature negative-curvature isotherms in the inset of Fig. 1 do exhibit the behavior characteristic of a true superconducting state with zero linear resistivity ρ_{lin}. In the vortex-glass[2] and collective-creep[10] models, this $E(J)$ behavior is characterized by equations of the form $E \sim \exp[(-A/J^{\mu})]$, where $\mu > 0$.

For a continuous superconducting phase transition at a temperature T^*, the linear resistivity should vanish as $\rho(T) \sim (T-T^*)^{\nu(z-1)}$ as T^* is approached from above, with the critical exponents ν and z characteristic of the transition's particular universality class.[2] In this regime near T^* a plot of $[d(\ln\rho)/dT]^{-1}$ as a function of temperature should be linear with a slope of $1/\nu(z-1)$.[10–12] A plot of this type is shown in Fig. 2 for the Tl-2212 film at $H=40$ kOe. In this case there are two large linear regions evident which is quite different from the single linear region previously observed in similar plots in YBCO (Ref. 12) and detwinned Bi-2212 crystals (Ref. 4). Extraction of the quantity $\nu(z-1)$ from each of these linear regions at all of the fields studied yields a value of 1.9 ± 0.1 associated with a possible high-temperature (T_{cH}) phase transition and 7.1 ± 1.1 associated with an apparent low-temperature (T_{cL}) transition. The value $\nu(z-1) = 7.1 \pm 1.1$ for T_{cL} agrees with the value of 7 ± 1 that has been reported for a three-dimensional vortex-glass transition in Bi-2212 crystals[4] and 8 ± 2 for YBCO thin films.[8] The temperature labeled T_{cR} in Fig. 2 identifies the crossover between the two linear regimes in these plots as defined by the intersection of linear extrapolations of the high- and low-temperature linear regimes. Shown in Fig. 3 is a phase diagram constructed from the values of T_{cH}, T_{cL}, and T_{cR} (denoted by the crosses) determined at all of the fields studied using plots similar to the one in Fig. 2.

There are several distinctive changes in physical properties at T_{cL} that are consistent with the formation of a vortex-solid phase which is characterized by a loss of linear resistivity. At all of the fields studied the ZFC/fcc dc-magnetization measurements[7] show the onset of irreversible (hysteretic) behavior to occur just below the value of T_{cL}

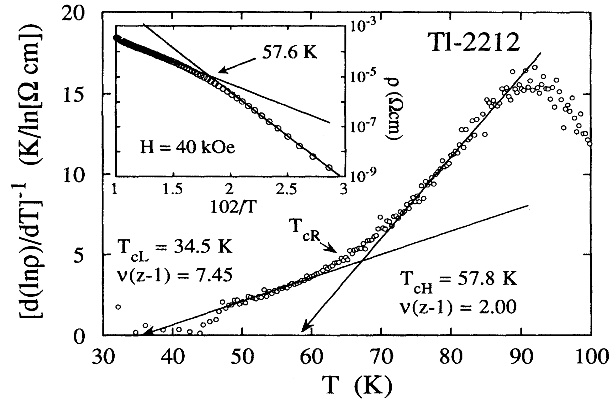

FIG. 2. Resistivity data plotted as $[d(\ln\rho)/dT]^{-1}$ as a function of temperature at $H=40$ kOe. Two linear regions in the plot correspond to critical regions of both an impending transition at higher temperatures (T_{cH}) and vortex-solid phase transition at lower temperatures (T_{cL}). The linear regions extrapolate to the respective critical temperature. The inset is an Arrhenius plot of the $\rho(T)$ data at $H=40$ kOe identifying the crossover that has been associated with a change in vortex-related dissipation.

determined from plots similar to that in Fig. 2. This change in behavior is evident in the $M(T)$ plot of Fig. 4 which shows reversible behavior down to about 54 K and then a strong upturn associated with the onset of irreversibility.[7] Identified in Fig. 4 are the values of T_{cL} and T_{cH} determined from the plot in Fig. 2 showing the close correspondence of the resistivity behavior and the upturn in magnetization at T_{cL} as well as a change in the reversible $M(T)$ dependence above T_{cH}. At all the fields studied, the reversible region between T_{cL} and T_{cH} is linear and crosses over to an upward curvature near T_{cH}. In addition, evidence for the onset of irreversibilities is found in the frequency dependence of the

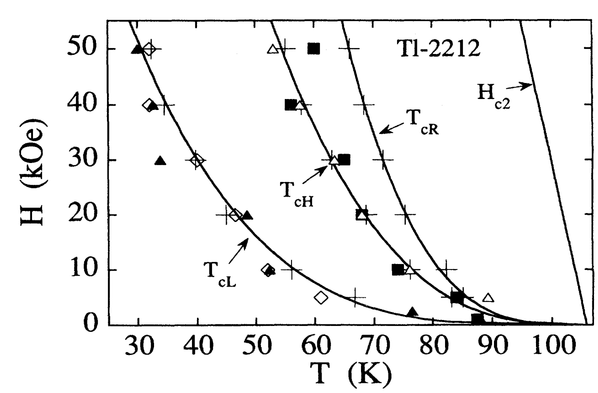

FIG. 3. Magnetic phase diagram for a Tl-2212 thin film. Curves are fits using the equation $H \sim (1-T/T_c)^n$ to T_{cL}, T_{cH}, and T_{cR} values, all indicated as crosses, determined from plots like that in Fig. 2. The solid triangles and hollow diamonds near T_{cL} determined from $\chi_{\text{ac}}(f)$ and $M(T)$ measurements, respectively, indicate the temperature where linear resistivity vanishes, while solid squares and hollow triangles near T_{cH} determined from $M(T)$ measurements and Arrhenius plots of the $\rho(T)$ data, respectively, indicate a change in vortex-liquid behavior.

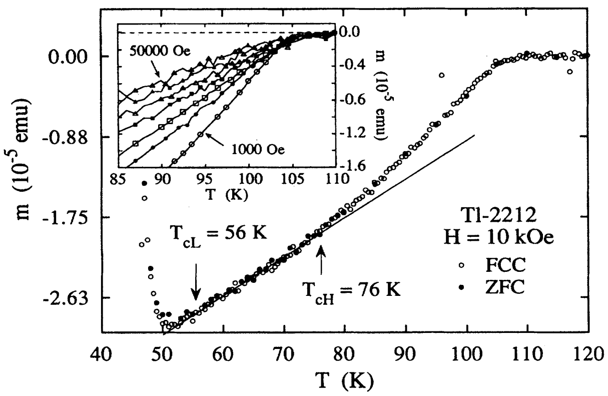

FIG. 4. Magnetization as a function of temperature at $H=10$ kOe. The values of T_{cL} and T_{cH} identified are those determined from the plot in Fig. 2, and are seen to set the range for a region of linear $M(T)$ behavior. The inset shows $M(T)$ data near T_c for $H=1$, 5, 10, 20, 30, 40, and 50 kOe.

onset temperature of the χ_{ac} response, $\chi_{ac}(f)$, which was found to extrapolate to T_{cL} at zero frequency. It was shown previously that such an extrapolation identifies the temperature at which the linear resistivity drops to zero.[2,7] Thus the $\chi_{ac}(f)$, $M(T)$ results, the low-temperature $E(J)$ behavior, and the $\rho(T)$ results all identify a drop of the linear resistivity to zero at T_{cL}. The transition temperature determined from the results of each of these various measurements is plotted in Fig. 3 and a close correspondence is observed for all of these methods. The large temperature regions over which linear behavior is observed in $[d(\ln\rho)/dT]^{-1}$ versus T plots, and the agreement with previously observed critical scaling coefficients, suggests that there is a continuous vortex phase transition at T_{cL} which is similar to the vortex glass transition in YBCO.[4,8,11,12]

There are also characteristic changes in the physical properties associated with T_{cH}. At all fields studied an upturn in the $M(T)$ behavior occurs above the T_{cH} value determined from $[d(\ln\rho)/dT]^{-1}$ plots. Further evidence for a change in properties is evident in the inset of Fig. 2, which is a plot of the resistivity (log scale) as a function of $1/T$ (Arrhenius plot). The temperature of the slope change for both the $M(T)$ curves, and the Arrhenius plots of each of the fields studies, is identified in Fig. 3. For all fields greater than 0 kOe there is a change in slope at T_{cH} that occurs between two approximately linear regimes. Often, the activation energy U for flux-vortex motion is determined from the slope in this kind of plot, and within this interpretation the slope change at T_{cH} would suggest a change in the dissipation mechanism associated with vortex motion. With increasing magnetic field there is a decrease in activation energy values determined from the slopes both above and below T_{cH}. Above T_{cH}, U ranges from 110 meV at $H=5$ kOe to 33 meV at 50 kOe while below T_{cH}, U ranges from 380 meV at $H=5$ kOe to 64 meV at 50 kOe. Above $H=10$ kOe, the ratio of U above to U below T_{cH} is approximately 2 and decreases slowly with increasing magnetic field, while below $H=10$ kOe, this ratio is greater than 3. The T_{cH} boundary in Fig. 3 was fit to an equation of the form $H\sim(1-T/T_c)^n$ and a value of $n=2.6\pm0.5$ was determined. This value is considerably larger than the value expected in the three-dimensional limit for either the vortex-glass transition ($n=1.5$–1.33) or the xy model ($n=1.33$).[2] However, it has been suggested that for the vortex-glass transition these dependencies will not be valid in magnetic fields large enough to drive the system into the quasi-two-dimensional regime where the vortex-glass and melting transitions begin to occur at two separate temperatures.[1]

The inset of Fig. 3 shows $M(T)$ isochamps for $H=1$ to 50 kOe near T_c. A two-dimensional scaling analysis[13] of the $M(T)$ data near T_c yields an initial upper-critical-field slope $dH_{c2}/dT=-5\pm1$ kOe/K and a value of the zero-temperature in-plane coherence length $\xi_{ab}(0)=25\pm5$ Å. The slope dH_{c2}/dT is plotted in the phase diagram of Fig. 3 and compares with previously reported values for Tl-2212 that range from -2.7 to -10 kOe/K.[9,14] Previously reported values of $\xi_{ab}(0)$ range from 11 to 31 Å.[9,15]

The changes in behavior observed at T_{cL} are all consistent with the existence of a continuous vortex solid (probably glass) phase transition in which the linear resistance drops to zero.[2,4,8,12] The behavior associated with T_{cH}, however, is considerably more difficult to characterize. A closer look at Fig. 2 reveals that the crossover temperature T_{cR} between the linear regions associated with T_{cH} and T_{cL} ranges from 4.4 K above T_{cH} at 5 kOe to 10.9 K above T_{cH} at 50 kOe. The higher-temperature linear behavior is consistent with an initial approach to a critical point (T_{cH}) of a particular universality class [having $\nu(z-1)\cong2$] that becomes preempted in the vicinity of T_{cR}. Within this scenario the system crosses over to a second universality class [having $\nu(z-1)\cong7$] and begins to approach to the critical point at T_{cL}. The temperature of the high-temperature critical point could be a function of some active parameter, such as Josephson coupling between superconducting layers, that changes the dimensionality of the system. The changes in physical properties appear to occur at T_{cH} rather than at T_{cR}, however, it is not possible to determine whether or not these changes occur outside of a crossover region, which may overlap T_{cH}.

The model of Feigel'man *et al.* appears to be fairly consistent with the observed results. This model describes a quasi-two-dimensional regime (for strongly anisotropic superconductors) in which occurs both a low-temperature three-dimensional continuous phase transition from a vortex-solid into a three-dimensional vortex-liquid phase as well as a high-temperature melting transition, or crossover, into a liquid of two-dimensional uncoupled pancake vortices.[1] The melting transition would correspond to the transition, or crossover, observed at T_{cH} and would separate a liquid of vortex lines for $T<T_{cH}$ from a liquid of unbound two-dimensional vortices for $T>T_{cH}$. In systems with weak disorder, decoupling transitions of this type have been predicted to be either first order or a crossover;[16] however, a continuous transition may be expected in the presence of strong disorder. Arrhenius plots similar to those in the inset of Fig. 2 were observed in measurements of $Mo_{77}Ge_{23}$/MoGe multilayers constructed in order to study the effects of Josephson coupling between separated superconducting layers.[17] Based on these results a pancakes-coupling-into-lines transition, or crossover, was suggested. This is the kind of transition expected at T_m in the model of Feigel'man *et al.* The lower bound of the quasi-two-dimensional regime in which T_g and T_m are separate transitions is given approximately by $H_0 \cong \phi_0/(d\gamma)^2$ (Ref. 2) or $4\phi_0/(d\gamma)^2$ (Ref. 1). Here d is the spacing between superconducting layers and γ is a measure of the anisotropy. Using values of $d = 11.55$ Å and γ ranging from 50 to 70, as previously reported for Tl-2212, values of H_0 range from about 25 to 3 kOe, respectively. The smaller estimate for this crossover is more consistent with the present observation of two separate transitions down to at least 1 kOe. Further evidence for a separate vortex-glass transition, and a possible dimensional-crossover boundary at fields greater H_0, has been found in Bi-2212 using plots of $[d(\ln\rho)/dT]^{-1}$ (Ref. 4).

In summary, evidence for two vortex-phase transitions, or crossovers, in a Tl-2212 thin film was observed. These observations are consistent with the existence of two different vortex-liquid phases separated by a phase boundary at T_{cH} and a true superconducting state at temperatures below T_{cL}. T_{cH} has been observed to be coincident with a change in the $M(T)$ behavior and a break between regions having different activation energies for vortex-related dissipation. T_{cL} has been observed to be coincident with the onset of irreversible $M(T)$ behavior, the extrapolation to the zero-frequency limit of the χ_{ac} response, and the onset of negative curvature in $E(J)$ curves. These results and the nonzero value of T_{cL}, as well as the quantity $\nu(z-1) = 7.1 \pm 1.1$, are all consistent with a continuous three-dimensional vortex-glass phase transition to a zero-linear-resistance state. The results are consistent with behavior expected for the quasi-two-dimensional regime of several models for anisotropic superconductors.

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