## Frequency independence of the vortex-glass transition in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> thin films

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The frequency dependence of the vortex-glass phase transition in a YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub> thin film was measured at frequencies ranging from  $10^{-1}$  to  $10^{5}$  Hz in applied fields H ranging from 0 to 50 kOe. The transition temperature  $T_g$  and the critical scaling parameter v(z-1) were determined from the scaling of both current-voltage characteristics and the resistivity measurements. The critical scaling parameters determined by different methods are found to agree  $[v(z-1)=7.2\pm0.3]$  and are consistent with the predictions of the continuous vortex-glass phase transition described by Fisher, Fisher, and Huse [Phys. Rev. B 43, 130 (1991)]. No frequency dependence is observed for the vortex-glass transition temperature  $T_g$ , for the range of parameters studied here. While the data do provide further support for the existence of a continuous vortex-glass phase transition, no evidence for the divergent time scale is observed, setting an upper limit of  $10^{-5}$  sec for this time scale at  $(T - T_g)/T_g \approx 0.02$ . Since ac-susceptibility data do not actually measure the "irreversibility line" (IRL), these results provide a fundamental measurement of the frequency dependence of the IRL. The observed frequency dependence is inconsistent with the fluxcreep-based picture of the IRL.

There is a continuing controversy about the nature of the so-called "irreversibility line" (IRL) in hightemperature superconducting materials. This controversy concerns the fundamental physics of the phase diagram in the H-T plane, in the presence of disorder which can pin the vortices. At least two possible scenarios have been proposed. In one picture, there is an actual thermodynamic phase transition between a vortex-liquid and a vortex-glass phase as the temperature is lowered.<sup>1</sup> In the other scenario, there is no actual phase transition; there is simply a crossover, as T is lowered, between two types of dynamical behavior.<sup>2</sup> Fairly recent experiments have provided evidence for critical scaling of both ac and dc transport measurement results,<sup>3,4</sup> providing strong support for a continuous phase transition in the highly disordered  $YBa_2Cu_3O_{7-\delta}$  (YBCO) materials. More recent<sup>1</sup> experiments have shown evidence for a first-order vortexlattice phase transition in YBCO samples with small amounts of disorder.<sup>5</sup> In both cases, the linear resistance appears to drop to zero at the phase transition, consistent with the model of Fisher, Fisher, and Huse (FFH).<sup>1</sup> This is in contrast to the persistence of a nonzero linear resistance that is expected in the dynamic crossover picture.

So why does the nature of the IRL remain an open question? One reason is based on the numerous reports of the frequency dependence of the IRL in YBCO when measured by ac susceptibility  $(\chi_{ac})^{.6,7}$  The temperature of the peak  $(T_{peak})$  in the out-of-phase first-harmonic  $\chi_{ac}$  response has often been used as an indication of the onset of irreversibility, and this as an indication of the IRL crossover or transition.<sup>7,8</sup> Using this method, many researchers have reported an approximate logarithmic frequency dependence  $T_{peak} \sim 1 - [B \ln (f_0 / f)]^{2/3}$  for the IRL at measurement frequencies f above 10 kHz, where  $f_0 \sim 10^8 - 10^{12}$  Hz is a characteristic attempt frequency and B is the magnetic induction.<sup>6,7</sup> This behavior is con-

sistent with the Anderson-Kim flux-creep model in which the zero-current activation energy is expressed as  $U_0(T,B,J=0)=(1-T/T_c)^{3/2}/B.^{7,9}$  In the dc limit, this model predicts that  $T_{\text{peak}}$  will approach 0 K as f approaches 0 Hz.<sup>1,6</sup> There has also been other evidence for a flux-creep-based crossover, including previous transport results.<sup>10</sup>

More recently, some groups have used the onset of nonlinearities in the  $\chi_{ac}$  response ( $T_{on}$ ) as an indication of the IRL.<sup>11,12</sup> However, it has been reported that both the first- and third-harmonic  $\chi_{\rm ac}$  responses in YBCO thin films are actually measures of ac magnetic-flux penetration (or equivalently, the ac magnetic-field penetration depth).<sup>13</sup> This suggests that the frequency dependence of the  $\chi_{ac}$  responses is directly related to the temperature dependence of the resistivity  $\rho(T)$  and is not a fundamental measure of irreversible magnetic behavior. It was also shown that the apparent frequency dependence of the IRL, as determined by  $T_{on}$ , is consistent with predictions of the vortex-glass model of  $FFH^1$  for the frequency dependence of ac-flux penetration into a sample.<sup>14</sup> However, rather than proving anything definitive, this recent progress in understanding  $\chi_{ac}$  measurements has only shown that the frequency dependence of the IRL has not yet been determined.

Since one key criterion for the existence of a true phase transition is the frequency independence of the transition temperature (at sufficiently low frequencies), we report here a fundamental measurement of the frequency dependence of the phase transition. Using resistivity measurements made over a frequency range from  $10^{-1}$  to  $10^5$  Hz, we examine the frequency dependence of the vortex-state transition at various values of the dc magnetic field *H*. We show the transition to be independent of frequency over a six-order frequency range and our ability to use an alternative scaling relation to fit our data provides further

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support for the idea that there is a true, continuous thermodynamic phase transition into the vortex-glass state in highly disordered YBCO materials.

Epitaxial YBCO thin films, with the c axis perpendicular to the film plane, were prepared by pulsed laser deposition onto heated (001) LaAlO<sub>3</sub> substrates using a method described previously.<sup>15</sup> The superconducting transition temperature ( $T_c$ ) for the 4000-Å thick film was 90 K, with transition width  $\Delta T_c \sim 2$  K as measured by ac susceptibility at 2.5 MHz.

The ac-transport and dc voltage-as-a-function-ofcurrent (V-I) measurements were performed on the 4000-Å thick YBCO thin film using a standard four-point probe technique. The transport bridge was approximately 70  $\mu$ m wide by 300  $\mu$ m long. Measurements were made after cooling in zero-applied field with data collected on warming. The dc magnetic field ranged from 0 to 50 kOe and was applied perpendicular to the plane of the film for all measurements. The ac excitation current was applied as a  $10^{-5}$  A rms square wave, generated by applying a voltage across the series combination of a 10-k $\Omega$ resistor and the YBCO film. The ac current value was monitored across the resistor. To avoid contact heating and eliminate contact potentials during the dc V-I measurements, the current was applied for two periods of a 1.3-Hz square wave and followed by a 5-s pause. For V-Imeasurements, the film was cooled in zero field and stabilized at the lowest recorded temperature. The field was then applied and constant temperature V-I data were taken in increasing temperature increments. The temperature reported for each V-I isotherm has a maximum temperature drift of 0.1 K. The dc resistance measurements were performed at  $10^{-6}$  and  $10^{-5}$  A, by a method described previously.<sup>13</sup>

Figure 1 shows the magnitude of the impedance from measurements made on a 4000-Å thick YBCO thin film as a function of temperature at various frequencies and applied magnetic fields. Magnetic field values of 0, 2.5, 12.5, 22.5, 32.5, and 50 kOe were used, and the frequencies of the square-wave excitation current were  $10^{-1}$ ,  $10^{0}$ ,  $10^1$ ,  $10^2$ ,  $10^3$ ,  $10^4$ , and  $10^5$  Hz. The normal-state phase shift of the impedance measurements during a temperature sweep was observed to be small and essentially independent of frequency. This suggests that the impedance is predominantly resistive. In order to be sure that measurements were made in the linear region of the V-I relations, the resistance was measured at several current values. The same resistance values were observed at all currents used and, in addition, dc resistance measurements gave the identical values of resistance (for a given H and T). The linear resistance  $R_{lin}(f, T)$  of YBCO at various H, shown in Fig. 1, appears to be independent of the frequency of transport measurements over the full frequency range.

In order for a vortex-glass transition to exist in a sample, it is necessary that the sample dimensions exceed the vortex-glass correlation length  $L_c \sim \{(10\varphi_0\xi)/[(4\pi\lambda)^2 J_c]\}^{1/2}$ , where  $\varphi_0$  is the flux quantum,  $\xi \sim 4 \times 10^{-8}$  cm is the superconducting coherence length,  $\lambda \sim 2 \times 10^{-5}$  cm is the penetration depth, and  $J_c \sim 10^7$  A/cm<sup>2</sup> is the critical current density at  $T \sim T_e$ .<sup>16</sup> For the



FIG. 1. Temperature-dependent resistance measurements at frequencies of 0.1, 1, 10, 100, 10000, 10000, and 100000 Hz. Each set of curves corresponds to a different applied magnetic field. Fields include 0, 2.5, 12.5, 22.5, 32.5, and 50 kOe.

4000-Å sample a value of  $L_c \sim 1000$  Å was determined, which is smaller than any dimension of the transport bridge under study. This suggests that a vortex-glass transition could exist in this sample. Evidence for a glass transition is found in the dc V-I isotherms which were observed to cross from positive curvature at higher temperature to negative curvature at lower temperatures, similar to previous reports.<sup>3</sup> The onset of negative curvature, with the concomitant loss of linear resistance, was observed at all the fields measured. The negative curvature onset temperature will be taken as the vortex-glass transition temperature  $T_g$ , and it is consistent with the transition to a phase characterized by zero linear resistance.

Below  $T_g$ , for J approaching 0, the FFH model predicts that the V-I isotherms will have the form  $V \propto \exp[-(J_0/J)^{\mu}]$ , where  $\mu < 1.^1$  The V-I measurements in this study exhibit this behavior having values of  $\mu$  between 0.2 and 0.3, consistent with a threedimensional vortex-glass transition. In general, for a three-dimensional continuous superconductor-to-normal phase transition, the dc electric field and the linear ac conductivity have the scaling relations

$$E(J,T) \sim J \xi^{1-z} E_{\pm} (J \xi^2 \phi_0 / k_B T) ,$$
 (1a)

$$\sigma(f,T) \sim \xi^{z^{-1}} S_{\pm}(f\xi^z) , \qquad (1b)$$

where J is the current density,  $\phi_0$  the flux quantum, and z the dynamic critical exponent. This model defines a coherence length  $\xi \sim |T - T_g|^{-\nu}$ , which diverges with a critical exponent v at the vortex-glass transition temperature  $T_g$ .  $E_{\pm}$  and  $S_{\pm}$  are the scaling functions above (+) and below (-)  $T_g$ . V-I data consistent with this model will collapse onto one of the two universal scaling functions  $E_{\pm}$  for the data at temperatures above and below  $T_g$ , when plotted as  $V/I|T - T_g|^{\nu(z-1)}$  versus  $J/|T - T_g|^{2\nu}$ . Using this relation, the dc V-I data measured at various H were scaled and the values of  $T_g$  and the critical exponents v and z are summarized in Table I. For a three-dimensional vortex-glass transition the predicted values for the critical exponents are 1 < v < 2 and



FIG. 2. Magnetic phase diagram using  $T_g$  determined from both scaled de V-I curves (solid diamond symbol) and using  $[d \ln(R_{\rm lin})/dT]^{-1}$  vs T plots. Each of the other symbols refers to one of the measured frequencies.

4 < z < 7,<sup>1</sup> consistent with the present, as well as most previous,<sup>3,5</sup> observations on YBCO thin films.

The scaling relation Eq. (1b) implies that the frequency-dependent linear resistance  $[R_{\rm lin}(f,T) \propto 1/\sigma(f,T)]$ , can be scaled by plotting it as  $|T - T_g|^{\nu(z-1)}/R_{\rm lin}(f,T)$  versus  $f/|T - T_g|^{\nu z}$ . All of the data in Fig. 1 were scaled using this method. This scaling was found to be sensitive to the choice of  $\nu(z-1)$  and  $T_g$ , allowing these parameters to be determined (see Table I). As shown in Fig. 3 for the H = 22.5 kOe data, with the correct choice of  $\nu(z-1)$  and  $T_g$  ( $T_g$  is fixed within the error bars of the dc V-I scaling results), the data in the critical regime will collapse onto the scaling function  $S_+$ . The portion of the  $R_{\rm lin}(f,T) \sim 1.3 \times 10^5$ . This implies that  $S_+$  is a constant, and  $R_{\rm lin}(f,T)$  is independent of frequency. This is consistent with the FFH theory, which predicts that  $S_+$  is a constant in the critical scaling regime ( $T - T_g |^{\nu(z-1)}/R_{\rm lin}(f,T) \sim 1.3 \times 10^5$ .

It was shown previously that  $R_{\text{lin}}^{s}$  can be rearranged to give  $[d \ln(R_{\text{lin}})/dT]^{-1} = [\nu(z-1)]^{-1}|T-T_g|.^{17}$  It follows from either Eq. (1a) or (1b) for  $T > T_g$  that  $R_{\text{lin}} \propto |T-T_g|^{\nu(z-1)}$ , since  $E_+$  and  $S_+$  are constants in the region of the critical scaling regime probed by the  $R_{\text{lin}}(f,T)$  data. As seen in Fig. 4 for  $R_{\text{lin}}(f,T)$  measured at H = 50 kOe and  $f = 10^3$  Hz, the data in the critical scaling regime will be linear when  $[d \ln(R_{\text{lin}})/dT]^{-1}$  is plotted as a function of T, having a slope of  $[\nu(z-1)]^{-1}$ and a temperature-axis intercept of  $T_g.^{17}$  This technique

TABLE I. A summary of  $T_g$ , v, z, and v(z-1) determined from scaling of the dc *V-I* isotherms and  $T_g$  and v(z-1) determined from scaling of ac resistance data, as in Fig. 3, for a 4000-Å thick YBCO thin film.

	V-I scaling				$R_{\rm lin}(f,T)$	
H (kOe)	$T_g$ (K)	z	v	v(z-1)	$T_g$ (K)	v(z-1)
2.5	84.2±0.2	5.2	1.65	6.9	84.0	7.0±0.5
12.5	79.7±0.2	5.0	1.75	7.0	79.9	7.1±0.3
22.5	$76.6 {\pm} 0.3$	5.0	1.75	7.0	76.9	7.2±0.4
32.5	74.1±0.3	5.1	1.80	7.4	74.4	7.8±0.4
50.0	$70.7{\pm}0.3$	4.8	1.85	6.9	71.0	7.8±0.5



FIG. 3. Scaling of the linear frequency-dependent resistance  $R_{\rm lin}(f,T)$  at 22.5 kOe for  $f=10^{-1}$ , 10<sup>0</sup>, 10<sup>1</sup>, 10<sup>2</sup>, 10<sup>3</sup>, 10<sup>4</sup>, and 10<sup>5</sup> Hz. Results are scaled as  $|T-T_g|^{\nu(z-1)}/R_{\rm lin}(f,T)$  vs  $f/|T-T_g|^{\nu z}$ .

is applied in Fig. 4 to the 4000-Å thick YBCO thin film. Note that the upper extent of the critical scaling regime in Fig. 4, which is indicated by the deviation from linearity, occurs between 79 and 81 K in agreement with that determined by the scaling of  $R_{\rm lin}(f,T)$ , mentioned above. The remaining results in Fig. 2 were determined from similar plots of  $[d \ln(R_{\rm lin})/dT]^{-1}$  as a function of T at each value of f and H measured. As seen in Table I, for each determination of  $T_g$  at a particular f and H, the values of  $T_g$  and v(z-1) were observed to be within experimental error of the values determined by scaling of the dc V-I data at the same applied field.

The highest temperature of the apparent critical scaling regime ranged from about 87 K at 2.5 kOe to about 81 K at 50 kOe, while the  $T_g$  values were 84.2 K at 2.5 kOe and 70.8 K at 50 kOe. The lowest temperature accessible in the critical scaling regime is limited by the noise associated with the measurement, which is on the order of  $10^{-3} \Omega$  with a  $10^{-5}$  A rms measuring current. The apparent critical scaling regime was found to be the same for all of the various measurements presented here, including *V-I* results. These results show the vortex-glass critical regime to be independent of frequency from



FIG. 4. Temperature dependence of  $[d \ln(R_{\rm lin})/dT]^{-1}$  for a 4000-Å thick YBCO thin film at H = 50 kOe and f = 1000 Hz. The linear fit is for the temperature range 74–79 K. The intercept of the linear region determines  $T_g = 71.1$  K while the slope determines  $\nu(z-1)=7.36$ .

 $f = 10^{-1}$  to  $10^5$  Hz. This is strongly suggestive that the flux-vortex phase transition is independent of frequency.

In the flux-creep-based picture of the IRL, there is a dynamic crossover in behavior.<sup>2</sup> In this model the critical current density is given by

$$J_{c} = J_{c0} [1 - (kT/U_{0}) \ln(f_{0}/f)], \qquad (2)$$

where  $J_{c0}$  is the critical current density without creep,  $U_0$ is the pinning potential, f is the measuring frequency, and  $f_0$  is a characteristic frequency. In this picture, the IRL occurs where the terms in brackets equal zero.<sup>2</sup> Some early results suggested that flux creep could account for the behavior, in particular, the frequency dependence inferred from ac susceptibility measurements.<sup>7</sup> The loss of a vortex-glass transition in very thin films<sup>18</sup> may explain some of the transport results suggestive of a flux-creep-based IRL,<sup>10</sup> while as mentioned above, ac susceptibility is not a valid measure of irreversibility onset and so its frequency dependence never was a measure of the frequency dependence of the IRL. More recent measurements have shown nonlogarithmic decay of the magnetization,<sup>19</sup> the nearly constant value of U/kT with temperature,<sup>20</sup> where U is a pinning potential, and the particular current density temperature dependence<sup>21</sup> are all more consistent with predictions of vortex-glass models than of single-vortex creep models. However, the negative curvature in V-I behavior and the scaling have probably provided the strongest support for a thermodynamic-phase-transition-based IRL.

The flux-creep model predicts a frequency dependence associated with Eq. (2), which is dependent on the value of  $f_0$ . For any choice of  $f_0$ ,  $T_{IRL}$  will approach zero at f=0. For  $f_0=10^9$  Hz,  $T_{IRL}$  would be expected to move by about 150 K for the 0.1 to  $10^5$  Hz frequency range employed here, when  $U_0$  is assumed to be independent of temperature. The frequency-dependent shift in  $T_{IRL}$  decreases when a larger  $f_0$  is assumed, but would still be about 65 K for  $f_0=10^{12}$  Hz. The lack of a frequency dependence for  $T_g$  measured from  $f=10^{-1}$  to  $10^5$  Hz therefore is fairly strong evidence against the flux-creep picture and is consistent with the expectations for a thermodynamic phase transition. The scaling and the associated values of v and z also support a continuous threedimensional vortex-glass phase transition at  $T_g$ . The value of v is larger than the mean-field expectation, but consistent with calculations for a spin-glass transition.<sup>1</sup> The value of z is also larger than mean-field expectation, but consistent with predictions for a highly disordered system.<sup>1</sup>

The FFH model describes a continuous phase transition to a vortex glass state and provides a scaling theory, with a divergent length scale  $(\xi \sim |T - T_g|^{-\nu})$  and time scale  $(\tau \sim |T - T_g|^{-\nu z})$ . The scaling of our data strongly suggests that, at the various values of f and H employed, we have been able to make  $T - T_g$  small enough to enter the critical scaling regime. If we write  $x \equiv f|T - T_g|^{-\nu z}$ so that  $\sigma(f,T) \sim \xi^{z-1}S_+(x)$ , our results indicate, for example, that the scaling function  $S_+(x)$  is a constant for xranging from  $10^{-8}$  to 1, when H = 22.5 kOe. This is consistent with the FFH picture, since these authors predict that  $S_+(x)$  is a constant for small values of x.<sup>1</sup> So, while our data provide support for the FFH picture, we as yet have no direct evidence of the divergent time scale entering  $S_+(x)$ , or that x is the correct scaling variable.

For our own range of fields (H=2.5-50 kOe) and  $|T-T_g|/T_g>0.02$  we can say that any diverging time scale is still shorter than about  $10^{-5}$  sec. This is consistent with the observations of Ollson *et al.*,<sup>4</sup> who were able to observe the divergence of the time scale at the considerably higher frequencies they employed in their experiments.

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