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# Identifying the loss of critical current density in $YBa_2Cu_3O_7$ thin films

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The magnetic and magnetotransport behavior of a $YBa_2Cu_3O_7$ thin-film sample was studied by a variety of methods that have been used previously to determine the irreversibility line (IRL). From transport measurements it is possible to identify at least two regions separated by a boundary identified as a vortex-glass transition, with the region below the boundary having zero linear resistance and that above having a low-current linear resistivity which seems to be associated with thermally activated flux motion. Evidence for flux pinning above the glass transition suggests that there is a loss of critical current density ($J_c$) rather than an onset of reversible magnetic behavior at the glass transition. The complications of using first-harmonic ac susceptibility ($\chi_{ac}$) to define the IRL are discussed. In addition, the third-harmonic $\chi_{ac}$ is shown to measure the same property that the first-harmonic $\chi_{ac}$ measures: ac magnetic-field penetration rather than the loss of $J_c$. Results and a theoretical model showing that there are significant differences between field-cooled dc magnetization measurements when the data are collected on warming versus collecting data on cooling are presented.

One of the more surprising features of high-temperature superconductors has been the loss of critical current density ($J_c$) in modest applied magnetic fields at temperatures well below the apparent superconducting transition temperature ($T_c$). This loss of $J_c$ has serious implications for those attempting to use these materials in many types of applications. Many models have been put forth to describe this so-called "irreversibility line" (IRL). Some of these models described the IRL as a dynamic crossover,[1] while others have considered it to be a thermodynamic transition.[2,3] The models based on standard flux creep theories suggested that a dissipative state always existed, but the results of Koch *et al.* provided strong support for the vortex-glass transition models in which, below a phase transition ($T_g$), a nondissipative state exists.[4] Further evidence for this transition has made the prospects for applications much more promising.

The measurement of the IRL has been a fairly controversial problem. The measurement of the IRL by Müller, Tagashige, and Bednorz using dc magnetization measurements seemed to provide a reliable method.[5] However, we will show that a commonly used variation of this method does not provide reliable results. The use of fundamental-frequency ac susceptibility ($\chi_1$) became a popular method to measure the IRL, even after the warnings of Shaulov and Dorman.[6] More recently, the realization[7] that $\chi_1$ is probably a measure of ac flux penetration[8,9] rather than irreversibility has led many to choose the third-harmonic ac susceptibility response to determine the IRL. The basis for using this method is that the existence of irreversibility will produce harmonics.[10] However, we will show that there is a coincidence of both the fundamental and third-harmonic response onsets that results from ac flux penetration occurring in a region in which the current-voltage behavior is nonlinear.

Epitaxial Y-Ba-Cu-O thin films, with the $c$ axis perpendicular to the film plane, were prepared by pulsed laser deposition onto heated (001) $LaAlO_3$ substrates using a method described previously.[11] The superconducting transition temperatures ($T_c$) ranged from 89.8 to 90.3 K, and the transition width $\Delta T_c$ was about 2 K as measured by ac susceptibility at 2.5 MHz.

Voltage as a function of current ($V$-$I$) measurements were made on films that had been patterned using laser ablation, with typical bridge dimensions of 70 $\mu$m wide by 300 $\mu$m long. A standard four-point probe technique was used for the $V$-$I$ measurements. For all measurements, the dc magnetic field for the measurements ranged from 0 to 55 kOe and was applied perpendicular to the plane of the film. To avoid contact heating and eliminate contact potentials, the current was applied for 2 periods of a 1.3-Hz square wave and followed by a 5-s pause. The temperature associated with each $V$-$I$ curve is an average of the temperatures recorded at each data point of the curve with maximum temperature drift of 0.1 K observed. All measurements were zero-field cooled, and data were collected on warming.

ac susceptibility ($\chi_{ac}$) measurements of the fundamental frequency and the third harmonic were made on the same films used in the $V$-$I$ measurements. The $\chi_{ac}$ measurements utilized a single-coil self-inductance technique for frequencies above 20 kHz and a two-coil mutual inductance technique for lower frequencies. The ac field was applied parallel to the dc field. The fundamental and the third-harmonic susceptibilities were measured in separate temperature sweeps and the in-phase and the out-of-phase components of each were recorded.

dc magnetization as a function of temperature was measured using a Quantum Design superconducting-quantum-interference-device (SQUID) magnetometer. The same sample used for ac susceptibility and for transport was measured. A scan length of 2.5 cm was used for the results presented and the temperature was stabilized to ±0.05 K before data collection. The samples were initially cooled and stabilized at the lowest temperature [zero-field cooled (ZFC)] and the field was then applied followed by collecting data on warming. The sample was then cooled from $T=99$ K in the same field and data again were collected on warming for the "field cooled with data collected on warming" (FCW) experiments. For the "field cooled with data collected on cooling" (FCC) experiments, data were collected as the sample was cooled from 99 K in a magnetic field using small temperature increments to avoid temperature undershooting.

Figure 1 is a plot of electric field ($E$) versus current density ($J$) for a 3000-Å film in an applied magnetic field $H=10$ kOe. The $E$-$J$ curves are isotherms, ranging from 75 K at the lower right to 92 K at the upper left. As observed previously,[4] three regions of differing curvature can be distinguished. At the highest temperatures an Ohmic region is evident. As the temperature is decreased, the curvature becomes more positive with individual isotherms in this region showing a crossing from Ohmic to power-law behavior, as current increases, which is characteristic of thermally activated flux motion.[12] In this region of positive curvature, and at higher temperatures, the dc magnetic behavior appears to be reversible. The behavior in this region of the $E$-$J$ curves is due in part to flux flow. The temperature of crossing (denoted $T_{\mathrm{ohm}}$) from the Ohmic region to this positive-curvature region, at various applied fields, is identified in Fig. 2, where $T_{\mathrm{ohm}}$ is defined as the lowest-temperature $E$-$J$ isotherm that is linear over its entire current range. As the temperature is decreased further, the $E$-$J$ curvature becomes negative. This change from positive to negative curvature is consistent with a vortex-glass phase transition.[2,3] The transition temperatures ($T_g$) at various fields were determined by scaling the $E$-$J$ curves to $V/I \sim |T-T_g|^{\nu(z-1)}$ and $I \sim |T-T_g|^{2\nu}$. Plotted in Fig. 2 is $T_g(H)$ along with a curve identified as $H_{c2}$ based on previous Y-Ba-Cu-O single-crystal results.[13] The other curves in Fig. 2 are determinations of the irreversibility line based on dc magnetic and ac susceptibility measurements.

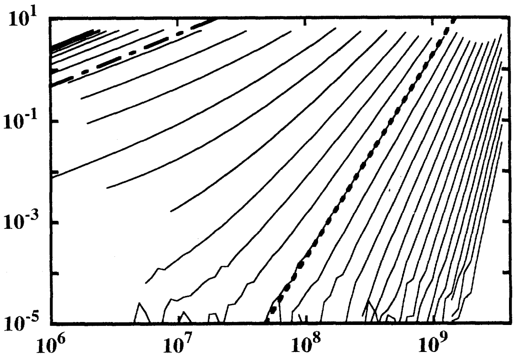

FIG. 1. Plots of $\log_{10}E$ vs $\log_{10}J$ for a 3000-Å-thick Y-Ba-Cu-O thin film in an applied magnetic field $H=10$ kOe. Each curve is an isotherm, ranging from 75 K at the lower right to 92 K at the upper left, in increments of 0.5 K. The broken line at higher temperatures identifies the isotherm $E(J,T=T_{\mathrm{ohm}}=87$ K), while the dashed curve is the glass transition isotherm $E(J,T=T_g=82.5$ K) calculated from parameters determined from the scaling of the $E$-$J$ results.

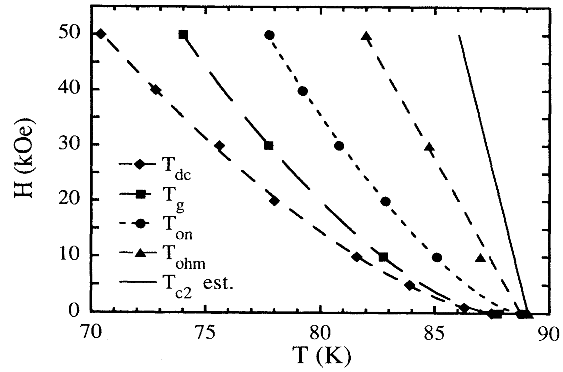

FIG. 2. The irreversibility line as determined by several methods on the same 3000-Å-thick Y-Ba-Cu-O film. Included are lines determined by dc magnetization, third-harmonic $\chi_{\mathrm{ac}}$, and magnetotransport measurements. The line defining $H_{c2}$ is an estimate determined from earlier measurements on single crystals and the line identified as $T_{\mathrm{ohm}}$ is the temperature above which the curves are Ohmic over their entire current range.

The results of dc magnetization measurements on the same 3000-Å-thick film are shown in Fig. 3. The curves in Fig. 3 correspond to three different types of measurements: (1) zero field cooled (ZFC), (2) field cooled with data collected on warming (FCW), and (3) field cooled with data collected on cooling (FCC). For both the ZFC and FCW curves there is a distinctive negative change in

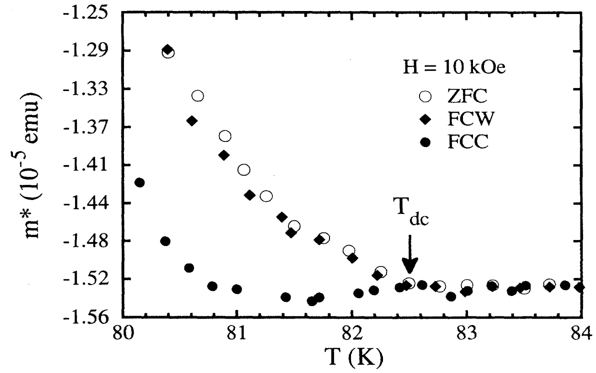

FIG. 3. Typical dc magnetization curves for zero-field cooled (ZFC), field cooled with data collected on warming (FCW), and field cooled with data collected on cooling (FCC) methods. At $T_{\mathrm{dc}}$ the FCC curve splits away from the coincident ZFC and FCW curves. The sample is the same 3000-Å thin film of Y-Ba-Cu-O measured at $H=10$ kOe.

the magnetization at a point we will denote $T_{dc}$. The large values of the apparent magnetic moment ($m^*$) and the anomalous SQUID output signals below this point are consistent with the disappearance of irreversibility at $T_{dc}$.[14] In Fig. 3 the ZFC and FCW curves are coincident down to about 10 K below $T_{dc}$ where they then split into two separate curves. On the other hand, as the temperature is decreased from above $T_{dc}$, the FCC curve initially makes a negative deviation at $T_{dc}$ before turning positive. The FCC curve does not coincide with the FCW and ZFC curves except at $T_{dc}$ and above. Values of $T_{dc}$ determined in various applied fields, are plotted in Fig. 2. The curve is seen to fall below that determined by transport measurements.

Shown in Fig. 4 are the voltages proportional to both the in-phase part of the fundamental $\chi_{ac}$ response, $\chi_1'$, and the amplitude of the third-harmonic $\chi_{ac}$ response, $|\chi_3|$, as a function of temperature measured using a fundamental frequency $f=70$ kHz in an applied dc field $H=10$ kOe. Clearly, both the fundamental and third-harmonic onsets are coincident. We have found a coincidence of both the fundamental and third-harmonic $\chi_{ac}$ responses at fundamental frequencies ranging from 50 Hz to 70 kHz. This is consistent with the observations of other groups.[15] The temperature of the onset of both $\chi_{ac}$ responses, in various applied dc fields, is plotted in Fig. 2 showing that at $f=70$ kHz this curve falls well above $T_g(H)$. The $\chi_{ac}$ onset temperature ($T_{on}$) increases with $f$ and in an applied dc field $H=50$ kOe has a dependence given by $T_{on}=74+0.87\ln f$.

For a superconductor containing sufficient disorder, recent vortex-glass transition models describe a phase transition from a vortex-liquid state above $T_g$ into a vortex-glass state. [2,3] As in Fig. 1, these models predict that below $T_g$ the $E$-$J$ behavior will have a negative curvature that results from the divergence of the pinning potential $U$ in the small-current-density limit. In this state the linear resistivity is zero. Above $T_g$ and below $T_{ohm}$, the $E$-$J$ behavior is characteristic of thermally activated flux motion and manifests a crossing from Ohmic behavior at small $J$ to power-law behavior at larger $J$ values, with a nonzero linear resistance at all $J$. Even though the $E$-$J$ behavior is nonlinear, it is not necessary that the $M(H)$ behavior be reversible, except in the small $E$-$J$ limit where the $E$-$J$ behavior is Ohmic. It is, however, true that measurements on a sufficiently long time scale will show evidence of reversible behavior resulting from the resistive decay of $J$ into the Ohmic portion of the $E$-$J$ curve. Although the IRL has been identified with the crossing at $T_{ohm}$, it follows that the IRL is actually the vortex-glass transition, and that this transition is characterized by the loss of low-current linear resistance and the resulting appearance of a $J_c$ that persists to infinite time due to the divergence of $U$ with decreasing $J$.

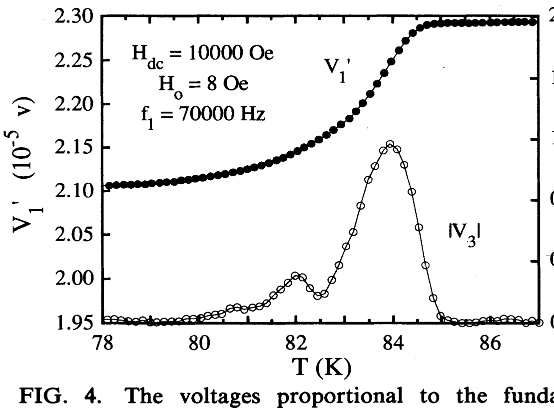

FIG. 4. The voltages proportional to the fundamental-frequency in-phase component of the ac susceptibility $\chi_1'$ and the magnitude of the third-harmonic ac susceptibility $|\chi_3|$ as a function of temperature for the same 3000-Å thin film of Y-Ba-Cu-O. The fundamental frequency is 70 kHz, the ac amplitude $H_0=8$ Oe, and the applied dc field $H_{dc}=10$ kOe. The onset of the first and third harmonics occurs at the same temperature.

The fundamental-frequency response $\chi_{ac}$ measures the extent to which ac fields penetrate into the sample.[8,9] It has been shown that $\chi_{ac}$ depends strongly on temperature near $T_c$. The peak in the out-of-phase signal ($\chi_1''$) occurs at a temperature $T_p$, which can be identified as the lowest temperature at which the ac magnetic field fully penetrates the sample. The position of this peak ($T_p$) depends on the sample size, the measuring frequency, and the sample resistivity. The $\chi_1$ response has been a standard method for measuring the IRL. At higher frequencies this $\chi_{ac}$ response measures the penetration of the ac field into the superconductor[8,9] with a characteristic penetration depth (Ref. 16), $\lambda_f=2\delta_f^2/d_f$, where the flux-flow skin depth, given by $\delta_f=(\rho_f/\mu_0\pi f)^{1/2}$, is used. Here $\mu_0$ is the permeability, $\rho_f$ is the flux-flow resistivity, and $d_f$ is the film thickness. In this case, the out-of-phase peak, $\chi_1''$, occurs at a temperature above $T_g$ at a value of $\rho$ such that $\rho_{peak}\sim 0.25\mu_0 2\pi f R d_f$, where $R$ is the film radius. At sufficiently low $f$ values, this high-frequency relation is no longer valid and $\rho_{peak}$ will occur below $T_g$. Now the penetration depth is proportional to $H_0$, and inversely proportional to $J_c$. Therefore, in the limit of small $f$ and small $H_0$ the temperature of the onset of $\chi_1'$ should be close to $T_g$. By extrapolating our observed $f$ dependence, we find that $T_g$, as measured by transport, is intercepted at $f\sim 1$ Hz.

The coincidence of the fundamental and third-harmonic responses is not unexpected given that $T_p$, and the $\chi_1$ and $\chi_3$ onsets, all occur within a temperature regime (between $T_g$ and $T_{ohm}$) in which the $E$-$J$ behavior is nonlinear. This coincidence occurs because nonlinear $E$-$J$ behavior will result in the generation of harmonics [a nonlinear $M(H)$ response]. At low enough $H_0$ and low enough frequency, the $E$-$J$ behavior is Ohmic suggesting that the third-harmonic could be eliminated. Some of our preliminary results suggest that this may be possible. Both the $E$-$J$ behavior and the presence of a third-harmonic $\chi_{ac}$ response are consistent with the existence of flux pinning in this region. However, because of the nonzero linear resistance a critical state can only exist transiently above $T_g$.

It is clear from Fig. 3 that the FCC and FCW methods do not give the same results. As shown by Clem and Hao for a cylinder in a parallel field,[17] this is because of differences in the flux-density profiles for the different temperature histories. During an FCC measurement, the flux density drops at the surface as it attempts to remain in equilibrium with the externally applied field. Flux is gradually expelled from the sample and the slope of the flux-density profile increases in magnitude. However, at each temperature point, the sample has a flux-density profile in which, as a function of distance from the center, the local flux density decreases monotonically out to the surface. On the other hand, during an FCW measurement, only at the lowest temperature does the flux-density profile decrease monotonically out to the surface. As the temperature increases, the flux density at the surface increases, and a flux-density front (having a higher flux density at the surface than at some point farther in) advances inward. At temperature $T^*$ this flux-density front reaches the center of the sample, and for temperatures $T > T^*$, as a function of distance from the center, the local flux density increases monotonically out to the surface. Flux continuously enters the sample and the slope of the flux-density profile decreases, eventually becoming perfectly flat at $T_g$.

During a ZFC measurement a flux-density front (having a higher flux density at the surface than at some point farther in) appears at the surface when the external field is first applied, and it advances inward as the temperature gradually increases. At some temperature this flux-density front reaches the center of the sample. As the temperature further increases, the slope of the flux-density profile decreases, eventually becoming perfectly flat at $T_g$. Note that the flux-density profiles for FCW and ZFC are identical for temperatures $T > T^*$. Thus the magnetization curves for the two processes must be identical in the temperature range $T^* < T < T_g$. It is clear that $T^*$ has nothing to do with the irreversibility temperature, since the joining of the magnetization curves at $T^*$ follows from the critical-state model, which is describing the irreversibility of the sample. Therefore, FCW in combination with ZFC, which is a commonly used method for determining the IRL by dc magnetic measurements, is *not* an accurate procedure for determining $T_g$.

Without further measurement, it is not possible to confirm that $T_{\mathrm{dc}}$ is an approximate determination of $T_g$. However, it would seem very likely to be so. At some point the magnetic moment associated with the irreversible magnetization falls below the resolution of the magnetometer, leading to an underestimate of the transition temperature. Therefore, a determination of the vortex-glass transition by dc magnetization measurements is expected to lie at a temperature lower than that determined by transport measurements.

In summary, it has been our purpose to show that some of the discrepancies in measuring the IRL are a result of using inappropriate methods. There appears to be only one IRL which is the same as $T_g$, and the characteristic property change at $T_g$ is the onset of a linear resistance rather than the onset of reversibility. We have shown that because ac field penetration occurs in a region in which the $E$-$J$ behavior is nonlinear, there is a coincidence of the fundamental-frequency and third-harmonic $\chi_{\mathrm{ac}}$ response onsets. Therefore, neither $\chi_{\mathrm{ac}}$ response is an accurate measure of $T_g$, except in the limit of low frequency and ac amplitude. We have also shown that to measure $T_g$ using dc magnetization requires that the combination of ZFC and FCC methods be used because the commonly used combination of ZFC and FCW does not determine $T_g$. The temperature determined by the ZFC-FCC method will always be lower than $T_g$ due to technical limitations.

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