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Irreversibility temperatures in c-axis-oriented powders of $YBa_2Cu_3O_7$, $Bi_2Sr_2CaCu_2O_8$, and $Bi_2Sr_2Ca_2Cu_3O_{10}$

Youwen Xu and M. Suenaga
Division of Materials Science, Brookhaven National Laboratory, Upton, New York 11973

Y. Gao and J. E. Crow
Department of Physics, Florida State University, Tallahassee, Florida 32306

N. D. Spencer
W. R. Grace & Company, Columbia, Maryland 21044
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The irreversibility temperatures $T_r(H)$ for the c-axis-oriented powders of pure and alloyed $YBa_2Cu_3O_7$, $Bi_2Sr_2CaCu_2O_8$, and $Bi_2Sr_2Ca_2Cu_3O_{10}$ were measured for $\mathbf{c}\|\mathbf{H}$. The results suggest that these temperatures are not the theoretically predicted vortex-lattice melting temperatures nor the vortex-glass-to-vortex-liquid transition temperatures.

The high degree of mobility for magnetic-flux lines in high-T_c superconductors, which is due to the short Ginzburg-Landau coherence lengths ξ ($\sim$1.5 nm) and the weak interlayer coupling, has been a much discussed subject since the first observation of this effect by Müller *et al.*[1] These authors showed that there is a significant temperature range below the critical magnetic field $H_{c2}(T)$ in which the magnetization of bulk La-Sr-Cu-O is reversible during a warming and cooling cycle in magnetic field. The low-temperature boundary of this reversibility range is called the irreversibility temperature $T_r(H)$. Based on the observation that $T_r(H)$ depends on H as $[1-T_r(H)/T_c(0)] \propto H^{2/3}$, they suggested the existence of a superconducting glass state. On the other hand, Yeshurun and Malozemoff,[2] who observed a similar relationship between $T_r(H)$ and H for a single crystal of $YBa_2Cu_3O_7$, argued that this magnetization behavior in these oxides could be described by a conventional flux-creep model.[3] Furthermore, they derived this relationship based on a flux-pinning argument.

These highly movable flux lines have also been studied by measurements of the broadening of the resistive superconducting transition T_c's under magnetic fields.[4-6] In addition, Gammel *et al.*[7] observed the temperatures of maximum dissipation in superconducting single crystals using a high-Q mechanical oscillator. They identified this $T_M(H)$ as the flux-line-lattice melting temperature. These results have stimulated theoretical studies.[8-12] Brandt[9] and Houghton *et al.*[10] calculated the melting temperatures from the nonlocal elasticity of the flux-line lattice within the anisotropic Ginzburg-Landau (GL) description. On the other hand, Fisher[11] argued that, in a type-II superconductor with random flux-pinning sites, there exists a disordered phase, a vortex-glass superconductor. He predicted a phase transition from a vortex-glass to a vortex-liquid phase with increasing temperature at a given magnetic field $H(>H_{c1})$, with the irreversibility line being the phase boundary. Subsequently, Koch *et al.*[13] showed that the anomaly in the I-V characteristics of a $YB_2Cu_3O_7$ film as a function of temperature can be interpreted as Fisher's glass-liquid transition. On the other hand, Griessen[12] and Esquinazi[14] argued that the Koch *et al.* data can be interpreted as due to thermally assisted flux flow, and that this vortex-glass–vortex-liquid line $T_g(H)$ is equivalent to the depinning line. Zeldov *et al.*[15] also showed that the resistive transition of a Bi 2:2:1:2 film can be interpreted as a consequence of flux depinning by incorporating a Lorentz force dependent pinning potential. Furthermore, Inui *et al.*[16] demonstrated that the resistive broadening in $Bi_2Sr_2CaCu_2O_8$ of Palstra *et al.*[4] can be explained by a single-vortex depinning model. Thus, it has not yet been determined whether this dissipative magnetic state is a consequence of a phase transformation in the flux-line lattice (or glass) or is simply due to thermally activated motion of flux lines. (See, for example, the review by Doniach.[17])

To provide additional information regarding the motion of the flux lines, we made a series of measurements of the irreversibility temperature $T_r(H)$ for a set of c-axis-oriented powders of nominal compositions $YBa_2(Cu_{1-x}M_x)_3O_7$, $Bi_2Sr_2CaCu_2O_8$, and $(Bi,Pb)_2Sr_2Ca_2Cu_3O_{10}$. (Here we refer to these oxides, respectively, as Y 1:2:3, Bi 2:2:1:2, and Bi 2:2:2:3.) We find that $T_r(H)$, as measured by the procedure described below, provides essentially the same information as $T_M(H)$ determined using the high-Q oscillator[7] and as the resistive critical temperature $T_R(H)$ obtained with a very low resistivity criterion [e.g., $\rho=10^{-4}$ $\mu\Omega$ cm (Refs. 4–6)]. We also studied the influence of substitutional elements in $YBa_2(Cu_{1-x}M_x)_3O_7$ on $T_r(H)$ and the temporal decay of magnetic hysteresis widths. These results will be discussed in terms of some of the theoretical ideas described above.

For the purpose of measuring the magnetic-field-dependent irreversibility temperature $T_r(H)$ and critical temperature $T_c(H)$, bulk sintered specimens of the desired compositions were prepared.[18-21] For alignment of the c axis of the powders along the field direction, the specimens were crushed to an average powder size of $\sim$10 μm and encapsulated in epoxy under a magnetic field of 80 kG.[22] The alignment was confirmed by transmission

Laué and powder x-ray-diffraction techniques.

A commercial superconducting quantum interference device (SQUID) magnetometer was used for the measurements of magnetic properties. Initially, the specimen was cooled to ~5 K in zero field and then a chosen magnetic-field value was set in several incremental steps. The sample was then slowly warmed up to a temperature well above $T_c(H)$ and then slowly field cooled. During a warm-up and cool-down cycle, the temperature was varied by 0.5-K steps for ~15-K spans below $T_c(H)$ and near $T_r(H)$. Between these regions the temperature was changed by 1 or 2 K per step. At each temperature, three measurements (three scans) were taken with a 2-cm scan length. As shown previously,[1] the trace of magnetic moment separates below $T_r(H)$ and two straight lines were drawn from the low-temperature side, and the intersection of these was defined as $T_r(H)$.

The magnetic-field dependence of the irreversibility temperature $T_r(H)$ for a Y 1:2:3 specimen is shown in Fig. 1 in a $\ln(H)$ vs $\ln[1-T_r(H)/T_c(0)]$ plot. The irreversibility temperature follows a power-law relationship, i.e., $H \cong a[1-T_r/T_c(0)]^n$, where a is a proportionality constant, and the value of n is approximately 1.5. Also shown in Fig. 1 are other transition temperatures determined by various methods. They are (i) the flux-lattice melting temperature T_M (Gammel *et al.*[7]), (ii) the vortex-glass–vortex-liquid transition temperature T_g (Koch *et al.*[13]), and (iii) the resistive transition temperature $T_R(H)$ at $\rho = 10^{-4}$ $\mu\Omega$ cm for a single crystal (Palstra *et al.*[5]) and 2.6×10^{-2} $\mu\Omega$ cm for a film (Iye *et al.*[6]) of $YBa_2Cu_3O_7$. Note that these transition temperatures all follow a similar power-law dependence on H. The values of n for the single-crystal data are ~1.5, for the films, ~1.2. The values of n among the crystal data and the present results are similar. This suggests that the irreversibility temperatures T_r measured here are comparable to T_g, T_M, and T_R determined for single crystals as well as for films. The differences in the values of the constants a and n are likely to be due to the variations in sample characteristics, e.g., flux-pinning strength, and in the level of measurement sensitivity.

In order to confirm this similarity among the transition temperatures, the magnetic-field dependences of the irreversibility temperatures, $T_r(H)$ for Bi 2:2:1:2 and Bi 2:2:2:3, are compared with T_M (Ref. 7) and T_R (Ref. 5) for Bi 2:2:1:2 single crystals in Fig. 2. For the Bi-based oxides, the relationship between $T_r(H)$ and H does not exhibit the same power-law relationship which was found in the Y 1:2:3 system.[23] However, the qualitative dependences of these transition temperatures on applied magnetic fields are nearly identical, similar to the situation in Y 1:2:3, although the exact temperatures for T_M, T_R, and T_r are different. Thus, we conclude that $T_r(H)$ as measured here is essentially the same as $T_M(H)$ and $T_R(H)$ and is also likely to be the same as $T_g(H)$, whether these transition temperatures are indications of the lattice melting or the depinning of the flux lines.

We also measured $T_r(H)$ for a set of specimens, $YBa_2(Cu_{1-x}M_x)_3O_7$, where M = Al, Fe, Ni, and Zn and $x = 0.02$. Again, the same power-law relationship was observed for all with $n \cong 1.5$, but the constant a varies. We measured the magnetic hysteresis width ΔM [at $T \cong 0.1$ and 0.5 $T_c(0)$ and $H = 10$ kG] for each specimen. We find that a decreases nearly proportionally with decreasing ΔM. It is clear that the pinning strength of a specimen has a strong correlation with the irreversibility temperature in the case of substituted $YBa_2Cu_3O_7$. In a recent report by Civale *et al.*,[24] a systematic shift in the constant a in the power law for $T_r(H)$ with increasing ΔM is reported for proton irradiated single crystals of Y 1:2:3. Large increases in $T_r(H)$ and ΔM for a neutron irradiated Bi 2:2:1:2 crystal[25] and Tl 2:2:2:3 ceramics were also reported recently.[26]

Among theories for the flux-lattice melting temperatures, only Houghton *et al.*[10] provide an explicit analytical expression for the transition temperature. We have attempted to find a best fit with their Eq. (3.9) in Ref. 10 al-

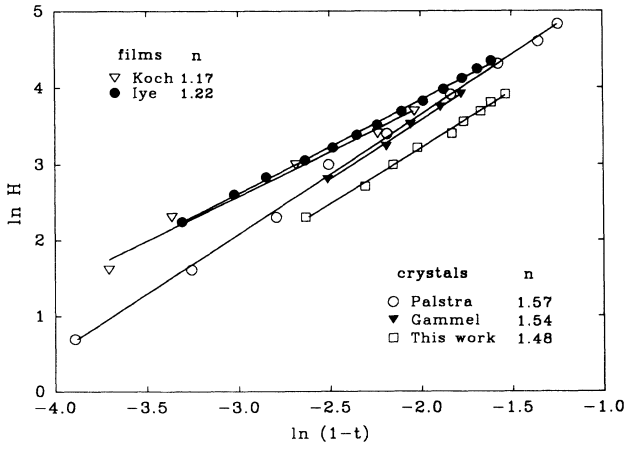

FIG. 1. $\ln[1-T_r(H)/T_c(0)]$ vs $\ln H$ for a pure $YBa_2Cu_3O_7$, and the transition temperatures $T_M(H)$ (Gammel *et al.*, Ref. 7), $T_g(H)$ (Koch *et al.*, Ref. 13), and $T_R(H)$ (Palstra *et al.*, Ref. 5 and Iye *et al.*, Ref. 6), which are determined by other techniques.

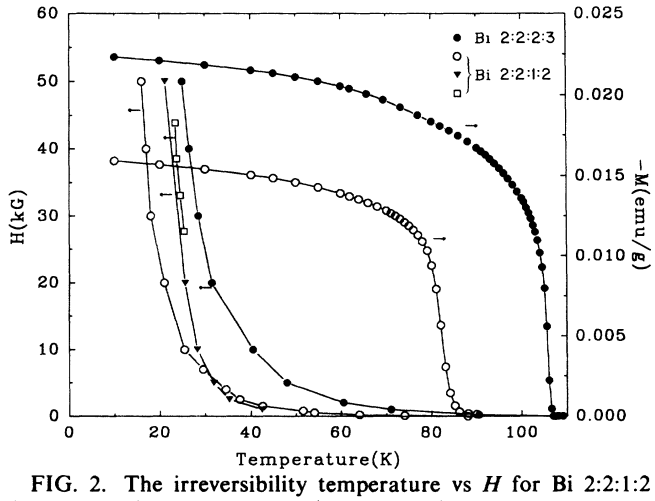

FIG. 2. The irreversibility temperature vs H for Bi 2:2:1:2 (open circles) and Bi 2:2:2:3 (solid circles) are compared with $T_M(H)$ (Ref. 7) (open squares) and $T_R(H)$ (Ref. 4) at $\rho = 10^{-4}$ $\mu\Omega$ cm (solid inverted triangles). The field-cooled (~1 Oe) results for Bi 2:2:1:2 (open circles) and Bi 2:2:2:3 (solid circles) are also shown to indicate the quality of these specimens.

lowing as a free parameter the value of c in the Lindemann melting criterion. However, since our data fit the power-law relationship with $n \cong 1.5$ quite well for all of Y 1:2:3, it was not possible to find a value of c to fit their expression, which predicts $n \cong 2$. Attempts to fit their expression for $T_r(H)$ for Bi 2:2:1:2 and Bi 2:2:2:3 were also unsuccessful. These poor fits may be due to the fact that either our measured $T_r(H)$ [as well as $T_M(H)$, $T_R(H)$, and $T_g(H)$] are not the melting temperatures for the flux lattice, or that these theories do not take into account the flux pinning.

Since the irreversibility temperature line does not appear to be a vortex-lattice melting line, the question reduces to whether this is a depinning line or a glass-liquid phase-transition temperature. Several observations support the depinning argument.[2,12,14] $T_r(H)$ is strongly dependent on the rate of temperature changes, on the frequency of magnetic field modulation,[27,28] on the size of the specimen, and on the pinning strength, ΔM as shown above. At the same time, many of the above observations, such as the frequency dependence and the pinning-strength effect on $T_r(H)$, can also be shown to support the glass-liquid transition model.[11,28] Thus, it is still difficult to conclusively argue for one model or the other. With the hope of clarifying this situation, we have examined the temporal decay of the hysteresis width in these oxides. The detailed results are discussed elsewhere and pertinent results are briefly summarized below.

Fisher predicted that the critical current of a superconductor at a temperature below $T_g(H)$ should decay, $J_c \sim \Delta M \sim (\ln t)^{-1/\mu}$ as time $t \rightarrow \infty$, where $\mu \leq 1$. In order to examine this prediction, we followed the procedure described earlier for flux-creep measurements.[29] Several measurements of $\Delta M(t)$ were made near the transition temperature (2–5 K below T_r) at 0.5 and 1 kG for Bi 2:2:1:2 and Bi 2:2:2:3, and at 10 and 20 kG for the specimens of pure Y 1:2:3. $\Delta M(t)$ was measured for times up to $\sim 1.4 \times 10^4$ s.

As shown in Figs. 3(a) and 3(b), for the Bi oxide specimens at these low fields, we found that $\Delta M(t)$ decayed with a logarithmic and then an exponential time dependence for the intermediate period and for $t \rightarrow \infty$, respectively. These are essentially the forms which are predicted by Griessen *et al.*[30] for thermally activated flux creep, i.e., $\Delta M \propto \ln t$ for small t and $\propto e^{-t}$ for $t \rightarrow \infty$. On the other hand, since, as shown in Fig. 3(a), $\Delta M \rightarrow 0$ for large t, we consider that this satisfies the condition $t \rightarrow \infty$ for the Fisher relationship to be applicable. However, as clearly shown in Fig. 3(c), the moment does not decay as predicted by the glass-liquid transition model, i.e., $\Delta M \propto (\ln t)^{-1/\mu}$. Thus, it appears that the decay of $\Delta M(t)[\sim J_c(t)]$ follows the conventional flux-creep model for the Bi oxides at this temperature and field regime. However, for Y 1:2:3, it was very difficult to achieve the condition $\Delta M(t) \rightarrow 0$ within a practical period, and it was difficult to determine whether Fisher's or the flux-creep model's predicted relationship was obeyed.

In summary, our measurements of the irreversibility temperatures $T_r(H)$ for Bi 2:2:1:2, Bi 2:2:2:3, and pure and substituted Y 1:2:3 oriented powders confirm that $T_r(H)$ is essentially the same as the resistive transition

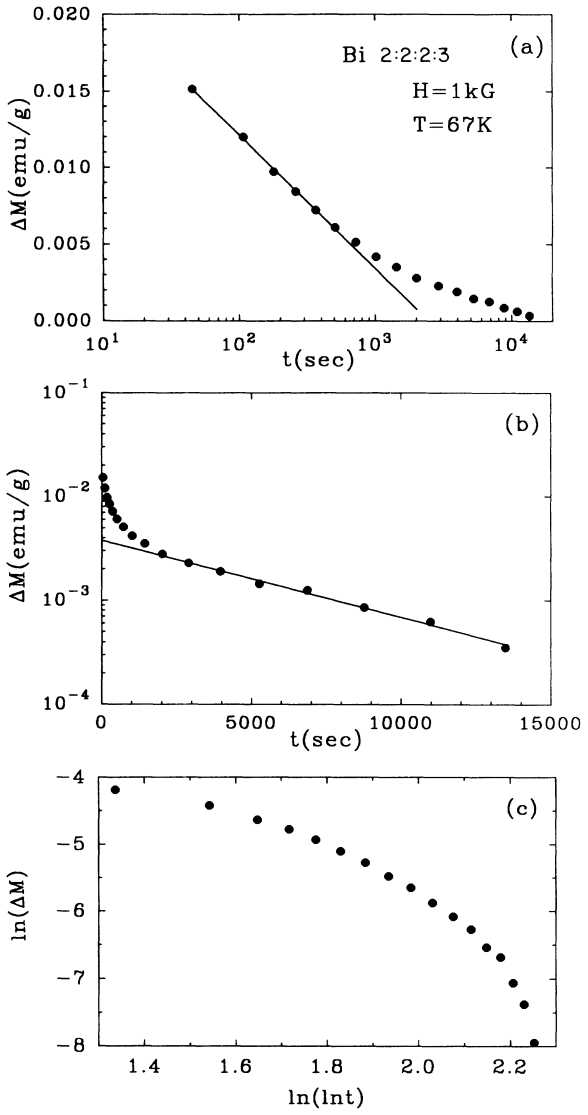

FIG. 3. An example of the temporal dependence of the magnetic moment $\Delta M(H,T)$ for the Bi 2:2:2:3 at $T \cong T_r(H)$ ($T = 67$ K and $H = 1$ kG). (a) and (b) are the same data plotted to show a logarithmic and an exponential temporal dependence, respectively. (c) is also the same data as in (a) and (b), but it is compared to the prediction by Fisher (Ref. 11), i.e., $\Delta M \propto (\ln t)^{-1/\mu}$.

$T_R(H)$,[4–7] the melting $T_M(H)$,[7] and the glass-liquid transition $T_g(H)$ (Ref. 13) temperatures. The results suggest that these temperatures are a manifestation of the depinning line in the H-T plane rather than of the lattice melting or the glass-liquid transition temperature predicted by theories.

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