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Diverging time scales for onset of irreversibility in high-temperature superconductors

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Abstract

The onset temperature, T_{irr}, for magnetic irreversibility in high-temperature superconductors has been investigated as a function of frequency ($0.2\ \text{Hz} \leqslant f \leqslant 65\,000\ \text{Hz}$) and DC magnetic field ($0 < H < 4000$ Oe). In Y–Ba–Cu–O crystals T_{irr} is essentially independent of the frequency, while in Bi–Sr–Ca–Cu–O and Tl–Ba–Ca–Cu–O crystals T_{irr} exhibits a strong frequency dependence. For all three systems $T_{irr}(H)$ converges to a finite temperature as the frequency approaches zero. Analysis of the experimental data shows that conventional theories of thermally-activated vortex motion cannot explain the measured frequency dependence of T_{irr}. Rather, the data indicate that T_{irr} for $f \to 0$ can be interpreted as a transition temperature separating the reversible vortex-liquid and the irreversible vortex-glass phases.

1. Introduction

Recent studies of high-temperature superconductors (HTSC's) focus on the disordered vortex structure in these materials [1,2]. According to these studies, as the temperature is lowered, the vortex system undergoes a phase transition from a "vortex-liquid" state, where the vortices are highly mobile, into a "vortex-glass" state, where the vortices are immobile and arranged in a sample-specific configuration. For a second-order phase transition, one expects a universal critical scaling behavior near the vortex-glass phase-transition temperature T_g. Critical slowing of the dynamics is also expected near T_g with a power-law divergence of the relaxation time [1,2]. In contrast to the vortex-glass phase-transition approach, the conventional theories of thermally activated vortex motion [3–6] predict that the mobility of vortices freezes gradually as the temperature is lowered. The vortex mobility is characterized by an attempt frequency, f, for hopping above the pinning barrier U, according to the Arrhenius law: $f = f_0 \exp(-U/k_B T)$.

The controversy regarding the two approaches de-

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scribed above, a phase transition versus a gradual freezing, recalls an echo of a similar debate, during the last two decades, on the nature of the transition into the spin-glass state [7]. In the latter, valuable experimental information on the spin dynamics was obtained from the response of the material to alternating magnetic fields at various frequencies [8]. In the present work we have adopted a similar experimental approach to study the vortex dynamics in HTSC's. Evidence for a vortex transition in HTSC's has been mostly based on magnetoresistence [9–15] and mechanical-oscillation [16] measurements. Recent work by Deak et al. [17] utilized third-harmonic AC measurements in studying the vortex-glass phase transition in YBCO thin films. Other studies of the frequency dependence of the AC magnetic response concentrated on measurements of the peak location of the out-of-phase fundamental susceptibility [4,18]. With few exceptions [18–20] the results were interpreted in the framework of the flux-creep model, identifying the peak location with the onset temperature, T_{irr}, for magnetic irreversibility. However, this interpretation has been highly debated [21–23].

In this work we describe measurements of the frequency dependence of the onset temperature T_{irr} for magnetic irreversibility in different HTSC crystals. This temperature is probed by measuring the onset of a nonlinear magnetic behavior, as indicated by the appearance of high-harmonic components in the response to alternating magnetic fields [21]. Special precautions were taken to avoid the Campbell regime [24], assuring that the amplitude of the alternating field is sufficiently large to drive fluxons in and out of their potential wells. This was accomplished by identifying the range of amplitudes for which the nonlinear response can be described by the critical-state model [25]. Using AC amplitudes in this range, we have interpreted the onset of a nonlinear response as the onset of magnetic irreversibility.

The phase-transition and the gradual-freezing models outlined above, both predict an onset temperature for irreversible magnetic behavior. However, these models yield different predictions for the frequency dependence of this temperature. According to the thermally-activated flux motion approach, the magnetic irreversibility results from gradual freezing of fluxons in their pinning centers. Above the irreversibility temperature T_{irr}, thermal activation permits depinning of fluxons to occur within the time scale of the experiment, leading to $T_{irr}=0$ for time scales approaching infinity, i.e. $f\to 0$. In contrast, within the vortex-glass phase-transition model, one may interpret T_{irr} for $f\to 0$ as the transition temperature T_g, separating the reversible vortex-liquid state and the irreversible vortex-glass state. We have attempted both approaches in explaining our experimental data for the frequency dependence of T_{irr} in different HTSC crystals. Our analysis indicates a clear discrepancy with the thermally activated flux-motion model and a good agreement with the vortex-glass phase-transition model.

2. Experimental

Single crystals of three high-T_c superconducting systems were investigated: $YBa_2Cu_3O_7$ (YBCO), $Bi_2Sr_2CaCu_2O_8$ (BSCCO), and $Tl_2Ba_2CaCu_2O_8$ (TBCCO). Three different YBCO samples were measured, referred to as Y1, Y2 and Y3 ($T_c=92.5$ K, 91.2 K and 89.2 K, respectively). The sample preparation is described in Ref. [26]. One of these samples (Y3) was irradiated at low temperature (20 K) by 2.5 MeV electrons in a fluence of 0.138 C/cm^2 [27]. The preparation of the BSCCO and TBCCO samples is described in Refs. [28] and [29], respectively.

The experimental set-up is based on a miniature InSb Hall probe [30] with a sensitivity of 50 μV/G, capable of measuring the response of the crystal to either DC or AC magnetic fields. The Hall probe ($80\times 100\ \mu m^2$) is attached to the c-face of the crystal using a thin layer ($\approx 5\ \mu m$) of vacuum grease. The crystal and the probe are placed on a temperature-controlled sample holder inside two concentric coils providing the AC and DC fields parallel to the c-axis. The maximum DC field achievable in this set-up is limited to 4 kOe. The DC signal from the probe is read by a 1271 Datron, $8\frac{1}{2}$ digits DVM, allowing a resolution of better than one flux line over the active area of the probe. For the AC measurements, a two-channel 650 Wavetek synthesizer is used, where the first channel provides the current for the AC field generation and the second one provides a reference signal for an Ithaco 3961B lock-in amplifier, by which the in-phase and out-of-phase components of the

harmonic signals are measured. The minimal detectable shielding current using this set-up is of the order 0.1–10 A/cm^2. This is equivalent to picovolts sensitivity in transport measurements [31].

Our experimental technique is based on measuring the onset of a nonlinear response to sinusoidal fields of various frequencies. This is interpreted as indicating the onset of irreversible behavior of the magnetization. As was pointed out by Konczykowski et al. [31], in this type of measurement, one should carefully select the proper amplitude, H_{AC}, of the driving field to avoid the Campbell regime. Campbell [24] showed that low-amplitude alternating fields might cause reversible oscillations of flux lines inside their potential wells, giving rise to a linear response even in the irreversible state. In addition, a nonlinear response may result from oscillations of fluxons inside anharmonic potential wells. Clearly, a nonlinear response of this origin is irrelevant to this study. In order to identify the range of amplitudes which give rise to a flux gradient and the nonlinear response predicted by the critical-state model [25,32–34], we measured the third-harmonic signal as a function of the amplitude H_{AC}, at different temperatures.

Fig. 1 shows the amplitude dependence of the normalized third-harmonic signal, V_3/H_{AC}, measured in sample Y1 at a frequency 177 Hz, in a DC field of 1000 Oe at various temperatures. The sample was field-cooled from above T_c to the measurement temperature which was kept stable within 20 mK during the measurement. In the range of low amplitudes, Fig. 1 shows a nonlinear increase of V_3/H_{AC} with amplitude, in contrast with the prediction of the critical-state model. However, one notices that this range of discrepancy shrinks as the temperature increases towards T_c. For example, the data obtained at 90 K is well described by the critical-state model, over a wide range of amplitudes, as shown by the solid curve in Fig. 1 [32]. Our measurements of T_{irr} were concentrated in a close vicinity of T_c and the amplitude of the alternating field was selected to be in the range where the experimental data agree well with the critical-state model. We note that the "problematic" Campbell regime appears on the ascending part of the curves in Fig. 1, while the onset of $V_3(T)$, in the process of cooling, should appear on the descending branch (or the "tail") of such curves. In this region demagnetization effects can be neglected, since the magnetization is very small. We have further verified that in the selected range of amplitudes, the onset of the third-harmonic signal is independent of the amplitude. This is demonstrated in Fig. 2 which describes the temperature dependence of V_3 for amplitudes of 0.5, 2.5 and 5 Oe, at a frequency of 177 Hz for the same YBCO crystal.

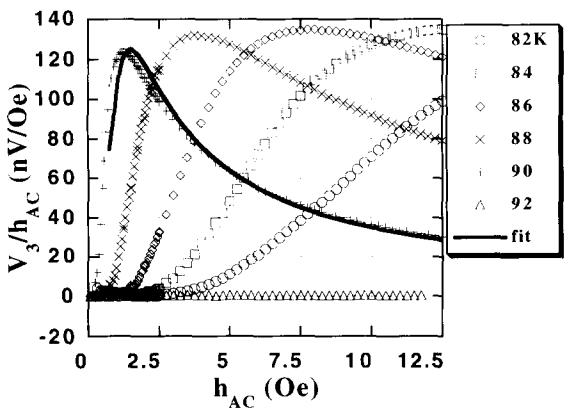

Fig. 1. Amplitude dependence of the third-harmonic signal measured in YBCO sample Y1 at frequency 177 Hz and DC field 1000 Oe at the indicated temperatures. The signal is normalized to the amplitude of the alternating field. The solid line is a fit to the critical-state model for $T=90$ K.

The temperature dependence of the third-harmonic signal was measured in the frequency range 0.2–65 000 Hz and DC field range 0–4000 Oe. Typical data for YBCO are shown in Fig. 3 for $H=300$ Oe, at several representative frequencies. Three observations can be made:

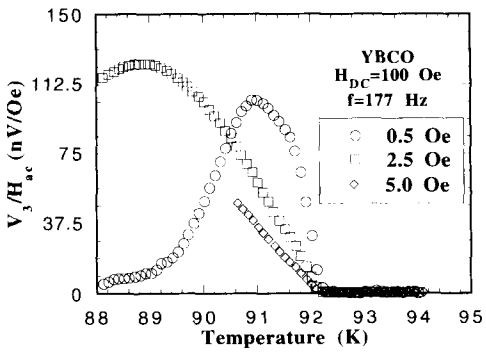

Fig. 2. Temperature dependence of V_3 in YBCO crystal Y1 measured at frequency 177 Hz and different amplitudes of the alternating field.

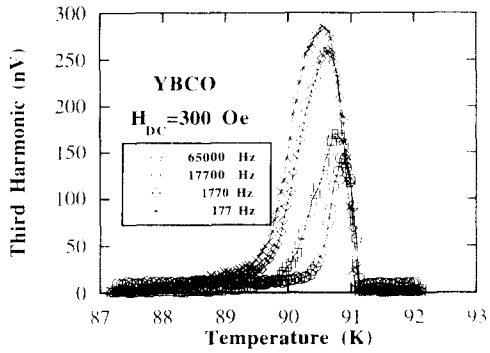

Fig. 3. Temperature dependence of third-harmonic signal for YBCO crystal Y2 measured at the indicated frequencies in a DC field of 300 Oe.

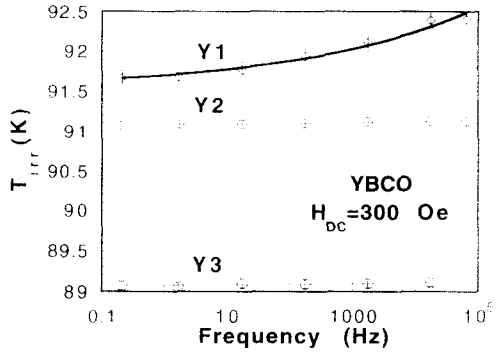

Fig. 4. Frequency dependence of the irreversibility point T_{irr} for three different crystals of YBCO, all measured in DC field of 300 Oe. The solid line for sample Y1 is a fit to Eq. (2) with $\nu(z-1)=6.5$.

(1) The third-harmonic signal rises abruptly as the sample is cooled from above T_c.

(2) The signal is strongly frequency dependent; as the frequency increases the signal decreases and its peak position is shifted towards higher temperatures.

(3) The onset of V_3 is weakly frequency dependent. The abrupt rise of V_3 at T_{irr} reflects a sharp transition from a reversible to an irreversible state. The possibility of a phase transition will be discussed in detail in the next section. The decrease of V_3 with increasing frequency can be attributed to the decrease in the penetration depth of the AC field as the frequency increases [35]. The peak is obtained when the alternating field fully penetrates the sample, and as the frequency increases this occurs at higher temperatures. A full analysis of the frequency dependence of the third-harmonic signal will be given elsewhere [36]. In the present paper we concentrate on the frequency dependence of the onset temperature of this signal.

Fig. 4 shows the frequency dependence of the irreversibility point T_{irr} as determined from the onset of V_3 for the three different crystals of YBCO, all measured in a DC field of 300 Oe. Apparently, the frequency dependence of T_{irr} is sample dependent; however, it is rather weak, with a maximum average variation of approximately 0.1 K/decade for sample Y1. A much stronger frequency dependence of T_{irr} is observed in both the BSCCO and TBCCO crystals as shown in Figs. 5 and 6, respectively, for various fields. In low fields the average variation of T_{irr} over five decades of frequency is approximately 2.5 K/decade for BSCCO and 6 K/decade for TBCCO. On increas-

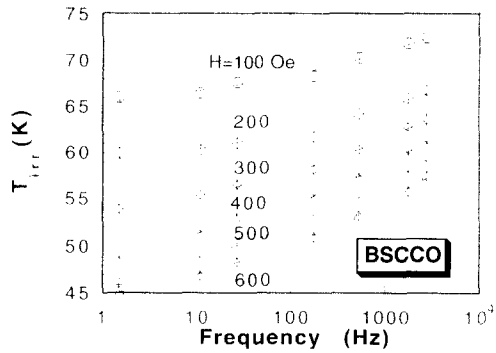

Fig. 5. Frequency dependence of the irreversibility point T_{irr} for BSCCO crystal measured at the indicated fields. The solid lines are fits to Eq. (2) with $\nu(z-1)=6.5$.

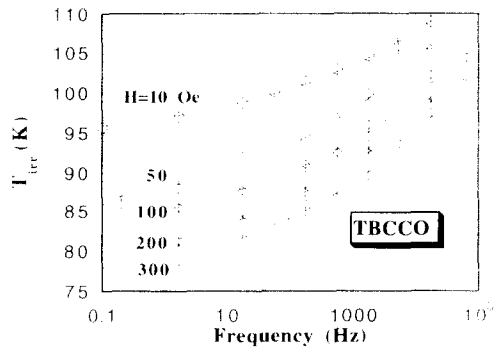

Fig. 6. Frequency dependence of the irreversibility point T_{irr} for TBCCO crystal measured at the indicated fields. The solid lines are fits to Eq. (2) with $\nu(z-1)=6.5$.

ing the DC field, the frequency dependence of T_{irr} in the BSCCO and TBCCO crystals becomes even stronger, while in the YBCO crystals it remains weak (of order 0.1 K/decade for Y1 and less than 0.02 K/decade for Y2 and Y3, in a field of 4 kOe). It is important to note from Figs. 4–6 that for all three systems (YBCO, BSCCO and TBCCO) T_{irr} *converges to a finite temperature as the frequency approaches zero.* The field dependence of T_{irr} for $f \to 0$ is described below for the various systems.

3. Discussion

The two competing models described in section 1 give different predictions for the frequency dependence of T_{irr}. In the gradual-freezing scenario the irreversible phase is a non-equilibrium state which slowly decays towards equilibrium via thermal-activation processes. The relaxation time τ is related [3] to the pinning energy U and temperature T according to the Arrhenius law $\tau = \tau_0 \exp(U/k_B T)$. If the time window of the measurement is much larger than τ, the system will appear reversible. Thus, for a given frequency, f, the onset of irreversibility is expected to be at a temperature T_{irr} for which τ is of the same order as $1/f$. Therefore, $1/T_{irr}$ should be linear with $\ln(f)$:

$$\ln(f) = \ln(f_0) - \left(\frac{U}{k_B}\right)\left(\frac{1}{T_{irr}}\right). \qquad (1)$$

Fig. 7 describes the logarithm of the driving frequency as a function of the inverse of the irreversibility temperature for the three HTSC systems in a field of 300 Oe. The data for YBCO show the linear relationship predicted by Eq. (1); however, the fit yields unphysically large values for the activation energy (~ 20 eV) and for f_0 (10^{50} Hz). For BSCCO and TBCCO the data deviate from linearity and cannot be fitted to Eq. (1) with a constant U. We thus fit the data in the lower temperature range where presumably U depends only weakly on the temperature (solid lines in Fig. 7); The fits yield unphysically large values for U and f_0 similar to those obtained for YBCO. One may attempt to fit the data over the whole temperature range by taking into account the temperature dependence of the activation energy U in Eq. (1). Note that in this process we add additional parameters to the fit, hence decreasing its reliability. Nevertheless, for any reasonable assumption [37] on $U(T)$ (e.g. $U = U_0(1-(T/T_c)^m)$, $m = 1$–4) we obtain for U_0 values which are orders of magnitude higher than those reported from relaxation measurements [4]. We thus conclude that gradual freezing of fluxons cannot describe the measured frequency dependence of T_{irr} in these crystals.

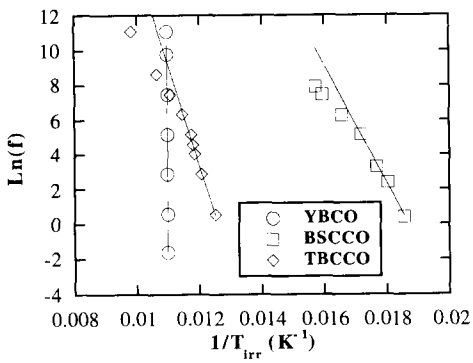

Fig. 7. Logarithm of the driving frequency as a function of the inverse of the irreversibility temperature for three HTSC systems in a field of 300 Oe. Solid lines are attempts to fit the data to Eq. (1).

The frequency dependence of T_{irr} may be better interpreted in the framework of the vortex-glass phase-transition model. For a second-order phase transition at T_g, one expects a power-law divergence of the correlation length $\xi \propto |T - T_g|^{-\nu}$, characterized by a critical exponent ν. Critical slowing down of the dynamics is also expected near T_g with a power-law divergence of the relaxation time: $\tau \propto |T - T_g|^{-\nu z}$, where z is the dynamical critical exponent. From the latter relationship one can deduce the frequency dependence of the irreversible temperature [1,2]:

$$T_{irr}(H, f) = T_g(H) + A(H) f^{1/\nu(z-1)}, \qquad (2)$$

where $T_{irr}(H, 0)$ is identified with $T_g(H)$. The solid lines in Figs. 5 and 6 show fits of this relationship to the experimental data obtained in BSCCO and TBCCO. In these fits we fixed $\nu(z-1) = 6.5$, to be consistent with the values determined in transport measurements [9,10,17,38]. From these fits we deduce the field dependence of the transition temperature T_g for BSCCO and TBCCO as shown in Fig. 8. For YBCO, a reasonable fit to Eq. (2) is obtained only for one sample (Y1, solid line in Fig. 4). For the other two samples which show essentially no fre-

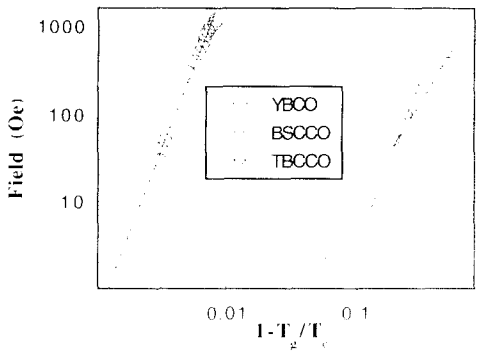

Fig. 8. Transition temperature $T_g(H)$ for YBCO, BSCCO and TBCCO deduced from Figs. 4–6. Solid lines are fits to $H \propto (1 - T_g/T_c)^p$.

quency dependence of T_{irr}, no attempt was made to fit the data, and T_g was identified with the frequency-independent value of T_{irr}. It should be noted that in the scale of Fig. 8 the T_g values for the three YBCO crystals are indistinguishable. The three curves in Fig. 8 describe a fit to a power law $H \propto (1 - T_g/T_c)^p$, as predicted by the vortex-glass phase-transition model [1,2]. The derived exponents $p = 3.8$, 2.6, and 3.8 for YBCO, BSCCO and TBCCO, respectively, deviate markedly from the predicted values [1,2] of $\frac{4}{3}$ or 2. However, one should keep in mind that because of the multiple fitting process and the number of parameters involved, the accuracy of the derived exponents is very limited and the data for BSCCO may be consistent with $p = 2$. Nevertheless, the outstandingly large value of p obtained for YBCO and TBCCO requires a further explanation. We note that because of the large slope of T_g versus H in YBCO, all the $T_g(H)$ data are concentrated in a limited range of temperature close to T_c. Most of this range is within the fluctuation region [39,40] of the zero-field superconducting transition T_c. (The width of this region is approximately 0.5 K for YBCO [40].) This is in contrast with the bulk of other experimental data where much higher fields were used and therefore their derived values of T_g are far below the fluctuation region. These experiments [10,16] yield p values which are consistent with the theoretical predictions. However, it should be noted that so far, no theoretical description of the vortex-glass transition in a magnetic field within the fluctuation region has been developed. Therefore, our results in YBCO may indicate a qualitatively different $T_g(H)$ behavior within and outside the fluctuation region of the zero-field transition. It is difficult to adopt this approach in explaining the large value of p obtained for TBCCO, since the range of $T_g(H)$ data for this crystal is extended over approximately 40 K below T_c. More measurements on different TBCCO crystals are required to resolve this problem.

Another issue which requires an explanation concerns the large variations in the rate of change of T_{irr} with frequency exhibited by the different HTSC systems. This can be attributed to the prefactor A in Eq. (2) which may depend on the system as well as on the quality of the sample. A thorough understanding of these issues requires further investigation. Nevertheless, our experimental observations of diverging time-scales for the onset of irreversibility and the power-law dependence, depicted in Fig. 8, point to a phase-transition rather than a gradual-freezing process.

In the previous paragraph we attributed the difference in the frequency dependence of $T_g(H)$ among the various HTSC systems to the prefactor A in Eq. (2). The exceptionally weak frequency dependence of T_{irr} in the YBCO crystals calls for further discussion. The data for one sample (Y1) show some frequency dependence of T_{irr}, which was analyzed above in the framework of a second-order phase-transition model. However, the other two samples (Y2 and Y3) show practically no frequency dependence (see Fig. 4), suggesting the possibility of a first-order phase transition. Such a transition is expected in the clean limit of an unpinned Abrikosov lattice, as was recently shown by Monte Carlo simulations [41]. Experimental evidence for a first-order phase transition was observed in transport measurements in untwinned YBCO single crystals [15,42]. Similar measurements in twinned crystals indicate a second-order phase transition [10]. Thus, one may conclude that the level of defects in the crystals influences the nature of the transition. This conclusion is supported by recent magnetic measurements of T_{irr} in thin films [17] which show a much stronger dependence on frequency than that exhibited by our crystals. Following this conclusion one may attempt to attribute the differences in the frequency dependence of T_{irr} in our three YBCO samples (Fig. 4) to the different levels of defects in these samples. However, it is difficult to accept this explanation in the light of the fact that

sample Y3 was electron irradiated and, therefore, there is no reason to believe that this sample is cleaner than the unirradiated sample Y1.

Finally, we comment on our AC magnetic technique for determining $T_g(H)$. This technique consists of two steps. First we measure the onset of a nonlinear magnetic response as a function of frequency and identify it with the onset T_{irr} of magnetic irreversibility. Second, we extrapolate the data to zero frequency and identify $T_g(H)$ with $T_{irr}(H,f)$ for $f \to 0$. Since $T_{irr}(H,f) \geq T_g(H)$, it follows that the onset of a nonlinear magnetic response may be in the *liquid* phase at temperatures well above $T_g(H)$. This is consistent with the experimental information obtained from the conventional I–V (current versus voltage) measurements of T_g. In these measurements, T_g is identified by the isotherm for which V is proportional to I^{α}, with $\alpha > 2.5$ for all current scales [9,40]. For a constant I, as the temperature is increased above T_g, α decreases gradually towards $\alpha = 1$ characterizing the ohmic regime. A nonlinear magnetic response may already be expected for $\alpha > 1$, i.e., in the liquid phase [43,44].

In conclusion we have measured the frequency dependence of the onset temperature, T_{irr}, for the magnetic irreversibility in HTSC's. This dependence cannot be described by an Arrhenius law as predicted by the conventional theories of thermally activated vortex motion. In contrast, the experimental data for YBCO, BSCCO and TBCCO suggest that T_{irr} for $f \to 0$ may be interpreted as a transition temperature separating the reversible vortex-liquid phase from the irreversible vortex-glass phase. The kinetics of the transition is consistent with a second-order phase transition. Two YBCO crystals exhibit practically no frequency dependence of T_{irr} suggestive of a first-order phase transition. More work is required to understand the physical reasons for the strong variation of $T_{irr}(f)$ among the different HTSC crystals.

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