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Frequency dependence of the ac susceptibility in a Y-Ba-Cu-O crystal: A reinterpretation of H_{c2}

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We report a small roughly logarithmic increase with frequency of the apparent transition temperature in ac susceptibility experiments on a Y-Ba-Cu-O superconducting crystal. Magnetic field enhances the effect. A flux creep model predicts the correct order of magnitude and implies that the transitions previously used to determine the upper-critical field H_{c2} actually represent an "irreversibility" line.

We report new data on a Y-Ba-Cu-O single crystal showing a small logarithmic dependence of the apparent transition temperature on frequency in an ac susceptibility measurement. While small, this effect is of importance, we believe, because it points strongly to a new interpretation of the apparent transition temperature. This interpretation, based on the concept of giant flux creep, points in turn to the need for a reevaluation of other data on the upper-critical field H_{c2} and of the fundamental parameters like coherence lengths deduced from that data.

A variety of techniques have previously been used to determine H_{c2}, including ac susceptibility,[1,2] the onset of resistance in transport measurements,[3] disappearance of irreversible magnetization and the corresponding critical current or pinning force determination,[4] and dc reversible magnetization measurements.[5] The results present a confusing picture. The first three techniques tend to show an upward curvature in the apparent $H_{c2}(T)$ as illustrated by the points labeled F_p (pinning force), χ (ac susceptibility), and R (resistance onset) in Fig. 1 for data on a single batch of Y-Ba-Cu-O crystals prepared at ALMOS (Amsterdam Lieden Cooperation for Materials Research in Amsterdam).[4] This curvature can be fitted to a dependence[2,3]

$$1-t=aH^{q}, \tag{1}$$

where t is the reduced temperature T/T_c relative to the true zero-field superconducting transition temperature T_c and where the exponent q ranges from about $\frac{2}{3}$ to $\frac{3}{4}$ in different measurements. Fits with $q=\frac{2}{3}$ are shown as dashed lines in Fig. 1.

This curvature has been variously attributed to dimensionality effects or non-mean-field critical fluctuations.[2,3] The latter interpretation is undermined by specific heat measurements[6] which indicate a critical region only a degree or so wide, while the $\frac{2}{3}$ law persists for more than 20 K below T_c. However, the curvature is remarkably similar to the so-called irreversibility line or "quasi-de Almeida–Thouless" line deduced from comparisons of filed-cooled and zero-field-cooled dc magnetic measurements in ceramics[7,8] and single crystals.[9] This is no coincidence, as will be argued below.

Another disturbing feature of Fig. 1 is the fact that data from different techniques on the same group of samples do not agree. There have also been reports of dc magnetic measurements,[5] on aligned granular Y-Ba-Cu-O collections of particles, which show $H_{c2}(T)$ to be linear or even concave downward. These data, adjusted for transition temperature T_c and labeled M_{dc}, are shown for comparison in Fig. 1. They lie well above data from the other three techniques. For comparison, we also show the transition determined from resistance measurements on the ALMOS Y-Ba-Cu-O crystals, but now, rather than taking the point of zero resistance, we extrapolate the roughly linear $R(T)$ data in the dropoff region upwards to where it intersects the high-temperature normal resistance. The resulting temperatures, plotted as squares in

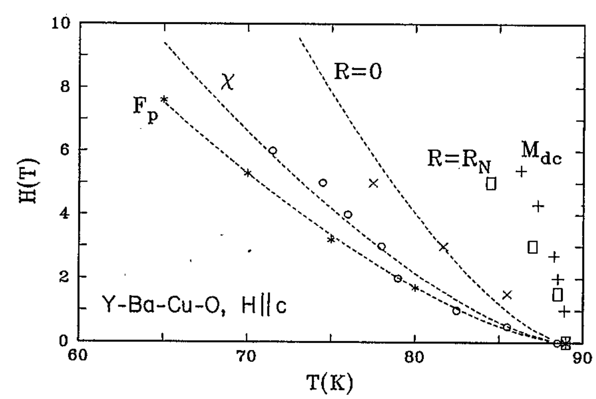

FIG. 1. Supposed determinations of H_{c2} for Y-Ba-Cu-O crystals and aligned granular material with $H \parallel c$. Stars, circles, and crosses represent measurements of zero-pinning force, ac susceptibility (100 Hz) anomalies, and zero resistance for crystals prepared from the same batch at ALMOS, Amsterdam (Ref. 4). Dashed lines represent fits to these data using a $1-t \propto H^{2/3}$ law. Squares represent resistance vs field data for the Amsterdam crystals, but with the transition determined from the onset of the resistance anomaly, as described in the text. Crosses represent the data of Fang *et al.* (Ref. 5) using reversible magnetization measurements on aligned granular material. The first three measurements are argued in the text to represent an irreversibility line rather than H_{c2}.

Fig. 1, also show the concave downward curvature.

To gain some insight into these measurements, we have studied the in-phase and out-of-phase ac susceptibility χ' and χ'' on the same Yorktown Y-Ba-Cu-O crystal as in Ref. 2, but now as a function of measurement frequency, and at applied dc fields from 0 to 1.4 T oriented parallel to the c axis. The ac field is also along this axis. Details of the measurement technique are given elsewhere.[2] Typical behavior is shown for $H=0.6$ T in Fig. 2. Temperatures (henceforth called $T_{\rm irr}$) determined from the maximum in χ'' and from the maximum slope of χ' are shown in Fig. 2 as squares and circles, respectively. Two experimental procedures, reducing temperature at fixed frequency or increasing frequency at fixed temperature, give equivalent results (as will be seen below, both procedures correspond to a change from the reversible to the irreversible regime).

At the lowest frequencies, near 10 kHz, the signal to noise is marginal, making it difficult to pick out the transition accurately. At the highest frequencies, near 100 MHz, cable resonances also introduce a systematic scatter in the data (e.g., the point at 60 MHz). Nevertheless, the trend of the data over the entire frequency range shows a roughly *logarithmic* increase of the apparent transition temperature with frequency. The slope $dT_{\rm irr}/d\ln(f)$ is determined from the region between 100 kHz and 10 MHz and is plotted as a function of field in Fig. 3. Because of the remarkable sharpness of the transition (0.2 K for 10–90% of the excursion), the positions can be measured to an accuracy approaching 0.01 K, and a weak effect can also be detected at zero-dc field, with $dT_{\rm irr}/d\ln(f)\simeq 0.019$ K.

While small, these frequency dependences give an essential clue to the origin of the discrepancies in Fig. 1, namely the possible differences in effective frequency of the different measurements. The logarithmic nature of the dependence appears to exclude interpretations based on phenomena like the frequency-dependent skin depth or temperature and field-dependent gap frequencies. A natural possibility is offered by a recent interpretation of the

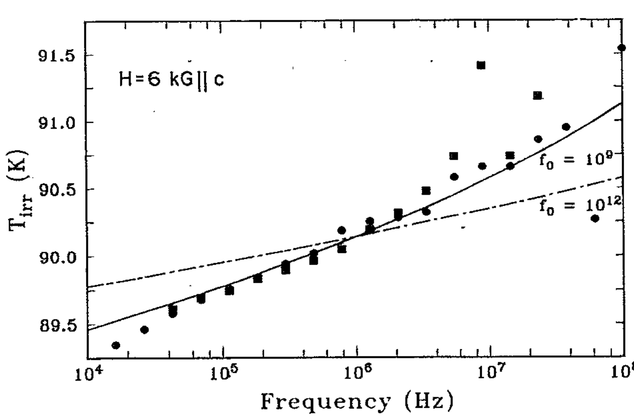

FIG. 2. ac susceptibility measurments on a Yorktown Y-Ba-Cu-O crystal with $H \parallel c$ as a function of measurement frequency. Squares represent the maximum in χ'' while circles represent the maximum slope in χ'. Solid and dashed lines represent theoretical predictions from the analog of Eq. (3) with the $\frac{3}{4}$-power and with $f_0 \simeq 10^9$ and 10^{12} Hz, respectively.

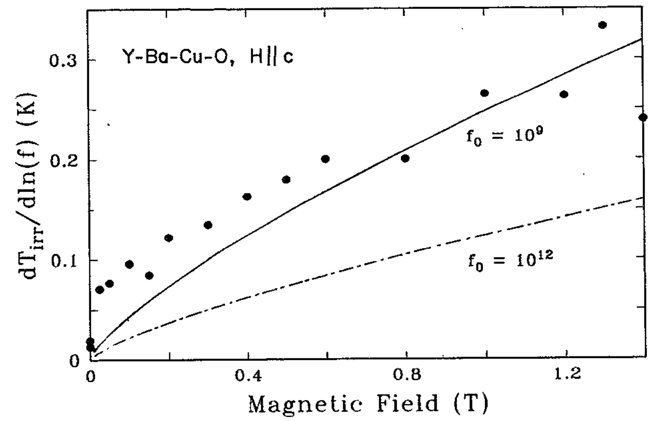

FIG. 3. The slope $dT_{\rm irr}/d\ln(f)$ as a function of applied field determined from data such as in Fig. 2 for the Yorktown Y-Ba-Cu-O crystal with $H \parallel c$, using the region between 0.1 and 10 MHz. The solid and dashed lines are the predictions of Eq. (5) with $f_0 \simeq 10^9$ and 10^{12} Hz, respectively.

time-logarithmic magnetic relaxation in Y-Ba-Cu-O in terms of flux creep.[9]

The basic idea is that the loss peak in the susceptibility is dominated by the onset of irreversible behavior in the magnetization, rather than by the onset of reversible magnetization at H_{c2}. It occurs when vortex lines are thermally activating very rapidly across the pinning barriers, corresponding to a point where irreversible magnetization, and the critical current which depends on it, drop below measurable levels given the experimental measurement times. The first three measurement techniques mentioned above all depend on such irreversible phenomena, and thus the values of H_{c2} extracted from such measurements might be interpreted as a determination of the irreversibility line. This, or course, would explain the apparent coincidence between the q-exponent close to $\frac{2}{3}$ in the ac susceptibility and in the original field-cooled and zero-field-cooled dc measurements.[7,8] The smearing in the resistivity drop as a function of field can now be understood as a flux-flow resistance effect, suggesting that the *onset* of the resistance drop might in fact be a better measure of the true H_{c2}.

To develop a semiquantitative theory of this effect, we follow the flux creep theory of Anderson and Kim[10] as developed by Campbell and Evetts[11] and more recently by Yeshurun and Malozemoff.[9] In this theory, thermal activation of vortices over an energy barrier U_0 is modulated by a Lorentz force on the vortices proportional to the critical current J_c and leads to a reduction of the measured critical current below its value J_{c0} in the absence of thermal activation[11]

$$J_c = J_{c0}[1-(kT/U_0)\ln(f_0/f)] , \tag{2}$$

where f_0 is a characteristic attempt frequency and f is the measurement frequency. We estimate f_0 from a phonon frequency at a wavelength $\xi \simeq 2$ nm, considering that the pinning is by the vortex cores and the pinning centers are attached to the crystal lattice. With a sound velocity of order 10^5 cm/sec, we have $f_0 \simeq 10^{12}$ Hz. A larger wavelength would give a smaller f_0, and, as will be seen below, $f_0 \simeq 10^9$ Hz gives the best agreement with experiment. Effects of backwards hopping modify Eq. (2) and the effective f_0, but do not change the behavior qualitatively; this will be discussed in more detail elsewhere.

Conventionally the thermal activation term in Eq. (2) is a negligible correction[11] to J_c, and even though the activation energy U_0 drops to zero at T_c, the temperature difference over which one might be able to observe an effect on J_c near T_c is exceedingly small in low-temperature superconductors. This is no longer true in the high-temperature superconductors, where the temperature T is an order of magnitude larger than in conventional superconductors and where the activation energy U_0 is generally lower.[9] This suggests an explanation of the irreversibility line because now the temperature difference over which J_c is measurably affected by flux creep is much larger. In particular $J_c = 0$ in Eq. (2) corresponds roughly to the temperature T_{irr} where the critical current becomes immeasurably small and where magnetic irreversibility, which depends on a finite J_c, disappears.

The field and temperature dependence of this line is determined by the scaling of U_0 with field and temperature. Many models of pinning in superconductors have been proposed.[11] However, at this point, the pinning mechanisms in the new high-temperature superconductors are just beginning to be studied,[4,12] and so we confine ourselves here, as in the earlier work,[9] to a very simple scaling argument which gives only one of many possible scaling forms. The basic idea is that the activation energy U_0 scales as the condensation energy per volume $H_c^2/8\pi$ times a characteristic excitation volume.[10,13] For sufficiently large fields, that volume is limited laterally by the area which a single-flux quantum occupies in the flux-line lattice. This area is approximately a_0^2, where $a_0 = 1.075\sqrt{\phi_0/B}$ is the flux-line spacing in field B (ϕ_0 is the flux quantum).

In the third direction, along the applied field, the minimum possible extent of the activation volume[13] is ξ, giving $U_0 \propto (H_c^2/8\pi)\, a_0^2 \xi$. [A second possibility, depending on the pinning mechanism, is for that third dimension to be a_0, which would give $U_0 \propto (H_c^2/8\pi) a_0^3$.] Now, according to the Ginsburg-Landau theory, H_c scales as $1-t$ near H_{c2} for large κ, and ξ scales as $(1-t)^{-1/2}$, so U_0 scales as $(1-t)^{3/2}/B$ [or, in the second case, $(1-t)^2/B^{3/2}$]. Substituting in Eq. (2), approximating T by T_c, and solving for $1-t$ with $t = T_{irr}/T_c$, one finds expressions for the irreversibility line valid with $f \ll f_0$ for the two cases

$$1-t = p[BkT_c \ln(f_0/f)]^{2/3}, \tag{3a}$$

$$1-t = p' B^{3/4}[kT_c \ln(f_0/f)]^{1/2}, \tag{3b}$$

where p and p' are constants to be determined experimentally. The powers $B^{3/4}$ and $B^{2/3}$ are in fact observed[2] for the directions $H \parallel c$ and $H \perp c$, respectively, in our crystal (applied field H and induction B are interchangeable because the magnetization is small). Least-square fits[2] to the actual data give $H(T) = 0.195(T_c - T)^{1.35}$ for $H \parallel c$ and $H(T) = 1.455(T_c - T)^{1.48}$ for $H \perp c$. Since the exact power may depend on the pinning mechanism, it is possible that the ALMOS crystals show a $\frac{2}{3}$ law while the Yorktown crystal shows a $\frac{3}{4}$ for $H \parallel c$.

To derive the frequency dependence, we differentiate with respect to $\ln f$ to obtain

$$dT_{irr}/d\ln(f) = (2p/3)(BkT_c)^{2/3}[\ln(f_0/f)]^{-1/3}, \tag{4a}$$

$$dT_{irr}/d\ln(f) = (p'/2)T_c B^{3/4}(kT_c)^{1/2}[\ln(f_0/f)]^{-1/2}. \tag{4b}$$

Equations (3) and (4) show that the apparent transition temperature *increases* with increasing frequency. This is physically plausible because at longer measurement times there is more time for flux lines to relax to an equilibrium configuration, and thus reversibility can be achieved at lower temperatures. The effect is close to logarithmic in frequency, although the $-\frac{1}{3}$ or $-\frac{1}{2}$ powers cause a small upward curvature.

It was earlier shown[9] that Eq. (3a) gives a semiquantitative description of the dc irreversibility line in an Y-Ba-Cu-O crystal. The explicit frequency dependence in Eq. (3) now offers an explanation of the different upward curving lines in Fig. 1 provided we interpret these lines as irreversibility lines rather than as the thermodynamic H_{c2}, which, of course, should not be dependent on frequency. Fits to the data of Fig. 1 give $1-t = 1.6$, 1.3, and $0.8 \times 10^{-4} B^{2/3}$ for the pinning, ac susceptibility, and resistive measurements, respectively (with B in gauss). Using the approximate measurement frequencies[14] of 0.3, 100, and 22000 Hz, respectively, and taking $f_0 \simeq 10^9$ Hz, we predict from Eq. (4a) 1.6, 1.3, and $1.0 \times 10^{-4} B^{2/3}$ (predictions relative to the susceptibility value, which was fixed from experiment). This crude estimate of the relative shifts already shows that they can be understood to within a factor of two from the different measurement time scales.

Direct measurements of frequency dependence reported in Figs. 2 and 3 provide a more stringent test of the theory. Using the earlier ac susceptibility data[2] and interpreting it now as the irreversibility line, we predict from Eqs. (3)-(4b) T_{irr} versus frequency and $dT_{irr}/d\ln(f)$ versus field (an $H^{3/4}$ law). These are shown in Figs. 2 and 3 as solid and dashed lines for $f_0 \simeq 10^9$ and 10^{12} Hz, respectively. While we have no *a priori* justification for the particular choice $f_0 = 10^9$ Hz, the theory is in good agreement with the rather substantial amount of data.

What do these results imply for the true H_{c2}'s and the fundamental parameters like the coherence lengths deduced from them? One cannot simply extrapolate the irreversibility line to higher frequencies to determine H_{c2}, although surely the true H_{c2}'s lie above the irreversibility lines, and so coherence lengths are smaller than those previously quoted.[1,2] The data of Fang *et al.*[5] shown in Fig. 1 are based on the reversible magnetization to determine H_{c2}, although this is a difficult measurement because of poor signal-to-noise ratio in a regime where the magnetization is exceedingly small. Such reversible measurements are certainly a more plausible way of determining the true thermodynamic H_{c2}, but the results are surprising and suggest some complications due to twin boundaries.[5] Similar results come from measurements of the *onset* of the resistivity drop as a function of field, as shown by the squares in Fig. 1. (Oh *et al.*[3] report onsets giving linear behavior.) Unfortunately, until the twin boundary effects or possible fluctuation effects are understood, the coherence lengths are not easily extracted form these measurements.

In summary, our results indicate that "giant flux creep" can explain the unexpected frequency dependence observed in ac susceptibility measurements. This implies that many measurements previously interpreted as H_{c2} should rather be interpreted as an irreversibility line. For example, the puzzling broadening in the resistance drop with field is likely to be a flux-flow effect. There is clearly more work to be done before a meaningful determination of fundamental parameters like the coherence lengths can be made.

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