Sudden Irreversibility Collapse in YBaCuO Crystals: Possible Evidence for Thermal Softening of the Core Pinning

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We report a new transition in the magnetic (H-T) phase diagram of the high-temperature superconductor $YBa_2Cu_3O_7$. A sharp ($\Delta T \simeq 0.020$ K) step in the irreversibility line is observed within about 1 K of T_c at fields considerably above the lower critical field. A contour map probing the nonlinear region of the H-T plane reveals a "backflow" and reentrant behavior below the irreversibility line, reflecting the collapse step. These observations are consistent with the existence of a thermal softening boundary which crosses the irreversibility line in the H-T plane, and at which vortex cores are delocalized on the scale of the coherence length ξ.

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The complexity of the magnetic-field–temperature (H-T) phase diagram in high-temperature superconductors has been recognized now for some time and there has been extensive discussion of various boundaries in the H-T plane [1]. In addition to the classic mean-field upper and lower critical fields H_{c2} and H_{c1}, attention has focused on an "irreversibility line," typically revealed by ac or dc susceptibility measurements [2,3], or by mechanical oscillator measurements [4]. This phenomenon has been variously interpreted in terms of a vortex thermal activation model [2,3], or a vortex-lattice melting model [4–7], or a vortex-glass freezing [8–10]. All of these models involve thermal excitation of vortices, and all lead to predictions of a transition boundary in the H-T plane which *decreases* in temperature with increasing field as a power law, $H_{irr} \propto (1-t)^\alpha$, where t is the reduced temperature T/T_c and α is between $\frac{4}{3}$ and 2.

A new crossover in vortex behavior was suggested recently by Feigel'man and Vinokur [11], and elaborated by others [12,13]. Their effect while also involving vortices under the influence of thermal excitation is qualitatively different; it has to do with the microscopics of the pinning mechanism, which is not directly addressed in other models. They point out that if the pinning involves a highly localized core pinning mechanism on the scale of the coherence length ξ, then when the mean-square thermal displacement $\langle u^2\rangle^{1/2}$ of the vortices exceeds ξ, the strength of the pinning will be strongly reduced.

This is distinct from Lindemann melting [4], in which melting of the vortex lattice is expected when $\langle u^2\rangle^{1/2}$ becomes larger than some fraction of the distance between vortices, and it leads to a qualitatively different prediction, namely, of a *thermal softening boundary* which *crosses* the usual irreversibility line. Indeed, the prediction of the Feigel'man and Vinokur theory [11] is that this new boundary *increases* in temperature with increasing field. It is important to recognize that this effect coexists with the other line or lines, that is, there can be an irreversibility line above and below the thermal softening boundary. If, however, the position of the irreversibility line depends on the pinning strength (as it does in the thermal activation and vortex-glass models but not in the lattice melting model), then one might expect the irreversibility line to be shifted to lower fields below thermal softening boundary; i.e., an anomaly where the two lines cross.

In this Letter, we report the discovery of a novel feature in the irreversibility line [2] of YBaCuO single crystals near T_c: The irreversibility line suddenly shifts to lower fields. This phenomenon appears to be the first experimental evidence for a crossover in pinning behavior proposed by Feigel'man and Vinokur [11], namely, a *thermal softening of the core pinning.*

The technique we use to define the irreversibility line $H_{irr}(T)$ is the usual tracking of the peak position in χ'', the out-of-phase component of the ac susceptibility. This technique was described elsewhere [14], but for the clarity of this discussion let us first comment on why indeed it is a probe of $H_{irr}(T)$. To be sure, χ'' exhibits a maximum even in a normal metal or in a superconductor without pinning when the skin depth $\delta_s = (c^2\rho/2\pi\omega)^{1/2}$ is equal to the sample thickness [15]. Here ρ is the normal or flux-flow resistivity and ω is the excitation frequency. This resistive absorption is a *linear* effect and so δ_s (and χ) does not depend on the amplitude of the ac field H_{ac}. In a superconductor with pinning, I-V curves are nonlinear in some range of fields and temperatures. In the nonlinear regime, the maximum in χ'' will still occur approximately when the characteristic penetration distance δ of the ac field equals the sample thickness. However, δ will now depend on H_{ac}, and the location of the maximum in χ'' (and, in general, the whole shape of χ' and χ'') will be amplitude dependent. The maximum in χ'' may thus occur [16] in the linear or nonlinear portion of the I-V curves depending on H_{ac}, ω, and sample dimensions. We define the irreversibility line at the *onset of nonlinearity*, which is a result of *pinning.* Figure 1 shows χ'' for several values of H_{ac}. For our 30-μm-thick crystal and with H_{ac} in our experiments typically 0.05–0.1 Oe, we *observe* that the position of the maximum roughly separates regions of linear and nonlinear amplitude response. This is true at all dc fields. Figure 1 shows that the maximum in χ'' approximately coincides with half screening as determined from χ', shown in the inset. This condition corresponds to a current density of $\simeq 20$ A/cm^2 (Ref. [17]). From the estimate in the linear regime, at $\omega = 1$ MHz, the peak in

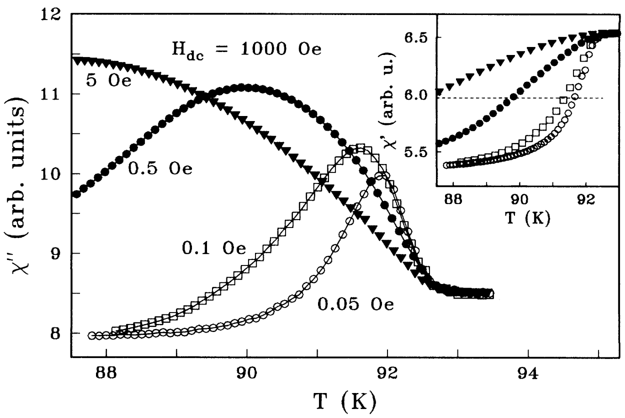

FIG. 1. Out-of-phase component of the ac susceptibility χ'' measured in a single crystal of $YBa_2Cu_3O_7$ at 1 MHz in 1000-Oe dc field for several amplitudes of H_{ac}. Inset: Corresponding χ' curves. Half screening is indicated by the dashed line.

χ'' occurs near resistivity of 3.6×10^{-7} Ω cm. Transport measurements on a crystal [9] similar to those used in this experiment confirm that under these conditions, and for this crystal, the maximum in χ'' will occur very near the temperature where nonlinear behavior is first noted in the *I-V* curves.

Irreversibility lines for the $YBa_2Cu_3O_7$ crystal [18] measured at 1 MHz with $H_{ac}=0.1$ Oe and in dc fields up to 6 T are shown in Fig. 2(a) for the two extreme alignments of the crystal with respect to the applied dc field. This result is similar to the ones reported previously [14]. In Fig. 2(b) we show in detail the data below 0.6 T and near T_c for a dc field parallel to the *c* axis ($H^{\parallel}_{dc}$), the shaded region in Fig. 2(a). A sharp (and reproducible on cycling) step in $H_{irr}(T)$ initiates at $H^{\parallel}_{dc}\simeq0.1$ T and completes, within $\Delta T\sim0.020$ K, at $H^{\parallel}_{dc}\simeq200$ Oe. The collapse is even more dramatic when displayed in a log-log plot of Fig. 3. The high-field power law is described here by the exponent $\alpha=1.33\pm0.05$ $(\sim\frac{4}{3})$, while below the step (or edge) the power-law exponent is 1.48 ± 0.08 $(\sim\frac{3}{2})$. The power laws were determined from least-squares fits in the field ranges from 0.35 to 6 T and from 0.001 to 0.02 T, respectively, where $T_c=92.69$ K was taken as the location of the maximum in χ'' in zero applied field; it was not a fitting parameter. The uncertainty we quote above is the standard deviation of the fit. We observe this new transition in the same field range in both twinned and fully untwinned crystals; six crystals were measured, all with T_c between 92.5 and 93.5 K, ΔT_c of ≤400 mK, and 100% shielding. The α values are consistently 1.2 to 1.4 at high fields and somewhat larger, between 1.5 and 2, at lower fields. Experimentally, the key difficulty to overcome is the maintenance of 20-mK stability at about 90 K; without it the collapse edge is smeared.

We argue now that the irreversibility collapse is a *symptom* of the thermal softening of the core pinning of

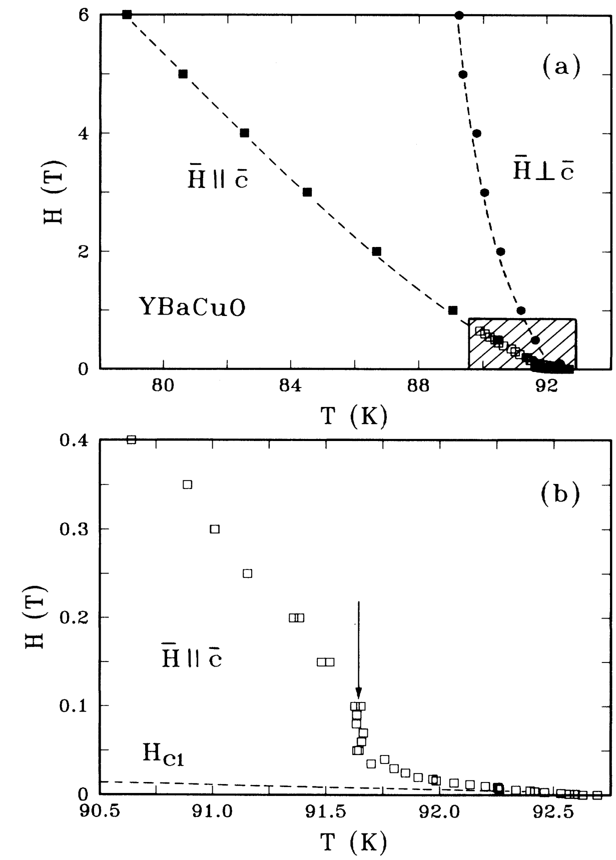

FIG. 2. (a) Irreversibility lines for the YBaCuO crystal of Fig. 1 for H_{dc} $\perp$ and $\parallel$ to the *c* axis measured at 1 MHz with $H_{ac}=0.1$ Oe. (b) Details of the irreversibility line at low fields for the $H^{\parallel}_{dc}$ orientation [the shaded region in (a)]. $H^{\parallel}_{c1}$ is shown for comparison.

the vortices [11,12]. The idea of Feigel'man and Vinokur [11] is, essentially, that if the pinning centers are of the atomic origin [19], vortex cores become too large (delocalized on the scale of ξ) for the pinning wells to be fully effective. Within the framework of collective pinning theory, the critical current J_c is strongly reduced [11,12] when the thermal displacement of the vortex core $\langle u^2\rangle \geq (1.4\xi)^2$. In an anisotropic superconductor, the harmonic thermal fluctuation of the vortex line at high temperatures is given by [12]

$$\langle u^2\rangle=\frac{4\pi^2k_BT\lambda^2\sqrt{\Gamma}}{\Phi_0^{3/2}\sqrt{B}\,[\ln(\sqrt{\Gamma}L_{c0}/\xi^{\parallel})]^{2/3}}\,. \qquad (1)$$

Here Γ is the anisotropy ($\sqrt{\Gamma}=\lambda^{\perp}/\lambda^{\parallel}$, the ratio of the penetration depths perpendicular and parallel to the *a-b* plane), $L_{c0}\simeq\xi^{\parallel}/\sqrt{\Gamma}(j_s/j_{c0})^{1/2}$ is the pinning correlation length in the single-vortex pinning limit, j_{c0} is the critical current in the same limit, and $j_s\simeq cH_c/\lambda^{\parallel}$ is the depairing current. Equating the right-hand side of Eq. (1) to $(1.4\xi)^2$, a parabolic $(\sim T^2)$ thermal softening boundary

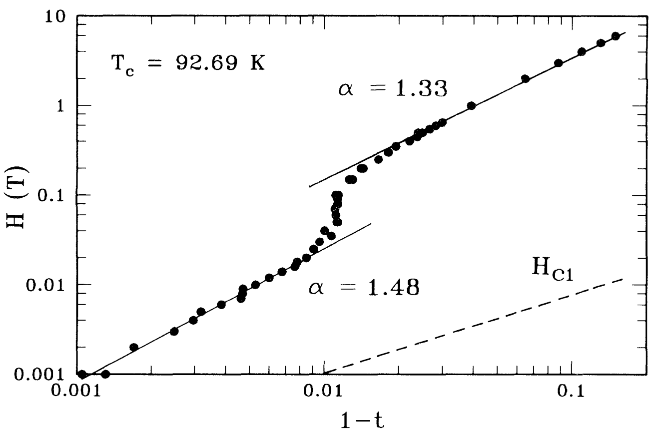

FIG. 3. Same as Fig. 2(a) for $H^{\parallel}_{dc}$ on a log-log scale. The lines represent fits of the data by the power laws below and above the collapse edge. The high-field exponent α is $\simeq \frac{4}{3}$ and at low fields $\alpha \simeq \frac{3}{2}$. $H^{\parallel}_{c1}$ is also shown.

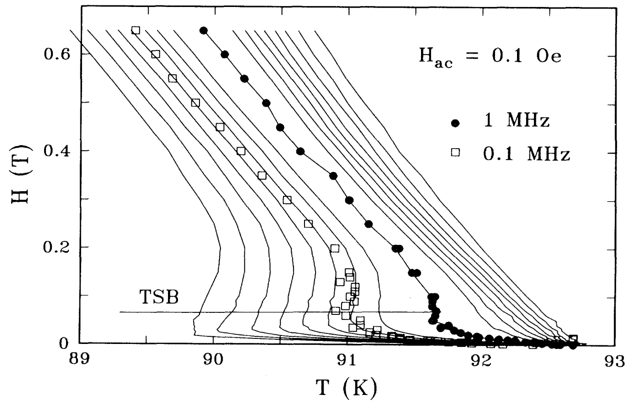

FIG. 4. Lines obtained from the maximum in $\chi''(T,H)$ at 1 MHz (irreversibility line shown as solid dots) and 0.1 MHz (open squares). The low-field anomaly is present in both and is consistent with the thermal softening boundary (TSB) $B \propto T^2$ [see Eq. (2)], indicated as a solid line; TSB crosses the irreversibility line. The contours probing the nonlinear regime below $H_{irr}(T)$ also show the anomaly, suggesting a change in the pinning behavior.

("depinning line" of Refs. [11,12]) is obtained:

$$B \simeq \frac{16\pi^4\kappa^4\Gamma}{1.96\Phi_0^3\{\ln[(j_s/j_{c0})^{1/2}]\}^{4/3}}(k_BT)^2. \quad (2)$$

This boundary *crosses* the usual upward-curving irreversibility line. We propose that the collapse edge articulates where the irreversibility and thermal softening lines meet. From Eq. (2) we estimate the magnetic field at which it takes place; the experimental anomaly is at $T \sim 92$ K and with $\Gamma \sim 25$, $\kappa \sim 57$ [20], and the ratio $j_s/j_{c0} \simeq 100$, the crossing should occur at $B \simeq 700$ Oe, in reasonable agreement with the middle of the observed anomaly. Of course, we recognize the extreme sensitivity of this estimate to κ (it comes in the fourth power).

As we argued earlier, the position of the maximum in χ'', $\chi''_{max}(T,H)$, is a reasonable description of $H_{irr}(T)$ line for our crystal at 1 MHz. At lower frequencies we are probing further into the nonlinear regime, and, if our argument holds, the anomaly we observe in $H_{irr}(T)$ should also appear there. This is clearly evident in Fig. 4, where we also show the line obtained from χ''_{max} measured at 0.1 MHz. Another way to examine the nonlinear regime is by projecting the *entire* peak onto the temperature axis (i.e., the temperature locations of, for example, 90%, 80%, 70%, etc., of the peak height on both sides of the maximum) and tracking it as a function of H_{dc}. The contours in the H-T plane obtained by such a procedure are distinctly different above and below the irreversibility line (Fig. 4). Above, in the linear regime, they can be interpreted as lines of constant linear resistivity. Our anomaly appears at the irreversibility line and propagates into the nonlinear regime towards lower temperatures, convincingly indicating the thermal softening boundary (TSB) which crosses the usual irreversibility line. The 0.1-MHz line, of course, coincides with one of the contours in the nonlinear regime, which all show a reentrant behavior below TSB. Similar analysis of χ' shows an identical contour behavior. For a more complete mapping at low temperatures of J_c = const lines in the H-T plane, transport and $M(H)$ hysteresis loop measurements are being pursued.

An alternative explanation is suggested by a possible superfluid entangled vortex-liquid ground state [1,7]; indeed, a reentrant boundary between entangled and lattice phases is predicted [1] near H_{c1}. In YBaCuO, the estimate of the field range above H_{c1} where the entropic effects due to flux line wondering are dominant is $(H - H_{c1})/H_{c1} \sim 1/\ln\kappa \sim 0.25$, 2 orders of magnitude smaller than we observe. Thus, unless pinning (which is not included in this theory) modifies such an estimate, we argue against it.

Finally, we note that the applied dc field at which the collapse occurs is 1 to 2 orders of magnitude above the lower critical field $H^{\parallel}_{c1}$. In our crystals, H_{c1} shows a simple linear behavior [with the slope ~ 10 Oe/K (Ref. [21])] extrapolating to T_c, as shown in Figs. 2(b) and 3. At these fields the flux-lattice spacing is smaller than magnetic penetration depth λ and, hence, it is unlikely that this new effect is due to the change in the nature of the vortex-vortex interaction (i.e., from logarithmic at higher fields to exponential near H_{c1}). There have been a few reports of the anomalies in H_{c1}; one such anomaly, namely, a step at T_c in some (particularly untwinned) crystals, was attributed to the presence of surface barriers [22]. Another kind of anomaly was observed by Safar and co-workers [23], who report a drop in H_{c1} a few degrees below T_c, independent of the orientation of the field. They also report an anomaly in the relaxation of the remanent moment at the same temperature in fields up to 350 Oe, suggesting an essentially *vertical* boundary in the H-T plane related to thermal decoupling of CuO_2 planes. In contrast to our results, these authors report no anomaly in the irreversibility line. Our observation is quite different: We see a shift in irreversibility line at about 1 K below T_c when H_{dc} is along the c axis in all our crystals, and it depends sensitively on field orientation [24]. Our experimental results and interpretation suggest a roughly *horizontal* (see Fig. 4) rather than *vertical* line. So, we conclude that there is no obvious relation of our irreversibility line anomaly to the occasional anomalies in H_{c1}.

In summary, we have observed a sudden collapse of the irreversibility line at low fields. We argue that this sharp transition provides the first evidence for the existence of a new boundary in the H-T plane at which thermal fluctuations on the scale of the coherence length ξ reduce the importance of the pinning energy which varies on the same length scale. A contour map in the nonlinear region of the H-T plane reveals a pronounced "backflow" (and reentrant) pattern and is consistent with this idea. The abruptness of the collapse calls for a more detailed explanation; presumably, it is controlled by the temperature dependence of J_c, which is suppressed strongly by the thermal fluctuations of the vortex lines and shows a fast power-law or exponential decay with increasing temperatures [11,12]. This thermal softening is quite different from the dislocation melting in the Lindemann sense [4,5,12], which occurs when the thermal displacement of the vortices is about 10% of the lattice spacing; at low enough fields the vortex lattice will first depin by thermal agitation and then possibly melt at higher temperatures [12]. Our interpretation also suggests that the irreversibility line depends on pinning.

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