3D *XY* Scaling of the Irreversibility Line of $YBa_2Cu_3O_7$ Crystals

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We find experimentally that the irreversibility line, $H_{\rm irr}(T)$, and the reversible magnetization of twinned $YBa_2Cu_3O_7$ crystals both show 3D *XY* scaling, which suggests a common origin for both quantities. Analysis of this result and of a correlation between the condensation energy and $H_{\rm irr}(T)$ of oxygen deficient $YBa_2Cu_3O_{7-\delta}$ for $0 < \delta < 0.6$, implies that $H_{\rm irr}(T)$ is strongly influenced by critical fluctuations. It is possible that these values of $H_{\rm irr}(T)$ and the recently discovered vortex melting line, $H_m(T)$, both correspond to an upper critical field line which has been suppressed by thermodynamic fluctuations. [S0031-9007(97)03779-4]

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High temperature oxide superconductors (HTSC) are unusual in that their superconducting magnetization curves $M(H, T)$ often appear to be perfectly reversible [1] for a wide range of fields (H) and temperatures (T) above a certain irreversibility line $H_{\rm irr}(T)$. This line is of great technical importance because it marks the onset of significant resistivity, i.e., power dissipation, in a magnetic field. It is strongly dependent on the strength of the interplane coupling [2], and $YBa_2Cu_3O_7$ is particularly interesting because it has the largest value of $H_{\rm irr}(T/T_c)$. The present paper deals exclusively with $H_{\rm irr}(T)$ of typical twinned $YBa_2Cu_3O_{7-\delta}$ crystals. However, for both untwinned [3] and twinned [4] $YBa_2Cu_3O_7$ there is evidence for latent heat at a phase transition line $H_m(T)$, widely referred to as the vortex lattice melting line which for $YBa_2Cu_3O_7$ is often very close to $H_{\rm irr}(T)$ [5–8]. Neutron scattering studies of $YBa_2Cu_3O_7$ show that the flux line lattice (FLL) disappears at $H_{\rm irr}(T)$ [9] but give no indications of line vortices in the liquid state. Therefore other possibilities such as a transition from a FLL to a "gas" of uncorrelated vortex pancakes [10,11] or as discussed here to a state with large (critical) superconducting fluctuations, remain open.

Thermodynamic fluctuations are much larger in HTSCs than in classical superconductors because the free energy difference between normal and superconducting states ($F_n - F_s$), multiplied by the coherence volume $\Omega(T)$ is comparable to the thermal energy k_BT over a significant temperature range. For $YBa_2Cu_3O_7$, the reduced Ginzburg temperature τ_G [12], which characterizes the width of this critical region, is 0.0023 at $H = 0$ [13] but is expected to increase in a field [12] to 0.011 at 20 kOe. For superfluid He^4 the width of the critical region is about 50 times larger than the value of τ_G, so it is not surprising that in a magnetic field the electronic specific heat [14,15] magnetization [16,17] and electrical resistivity [18] of $YBa_2Cu_3O_7$ crystals do obey the 3D *XY* scaling laws expected for critical fluctuations in a type II superconductor with a 2-component order parameter.

Here we show that for our $YBa_2Cu_3O_7$ crystals, 3D *XY* scaling holds over a very wide range of H and T and that $H_{\rm irr}$ occurs at a fixed point on the 3D *XY* plots. We discuss these results in terms of critical fluctuations and predict a correlation between $H_{\rm irr}$ and the condensation energy, which agrees with experiments on oxygen depleted crystals.

$YBa_2Cu_3O_7$ crystals were grown from flux in an Y_2O_3 crucible (with 1% BaO as binder) using 99.999% purity chemicals [19]. The duration of the final treatment in flowing O_2 gas at 420 °C was adequate because identical results were obtained after annealing for 7 and 11.5 days. Data are reported for a 5.86 mg, $1.6 \times 1.5 \times 0.38$ mm^3 twinned crystal with H along the shortest dimension, the crystallographic c axis. Similar results were obtained for another crystal.

Figure 1(a) shows $M(H)$ data taken with a Quantum Design SQUID magnetometer using a scan length of 4 cm and 30 sec pause at each field. The normal state paramagnetism has been subtracted by fitting the susceptibility at 50 kOe to the law $A + C/T$ from 100 to 300 K. The Curie term, $C = 0.016$ emu K/mole, corresponds to 1.4% spin $\frac{1}{2}$ defects per Cu atom while $A = 3.9 \pm 0.1$ 10^{-4} emu/mole agrees with the value of 3.7 ± 0.15 10^{-4} emu/mole expected for $YBa_2Cu_3O_7$ crystals with $H \parallel c$ [20]. The remaining magnetization is referred to as $M_{\rm fluc}$. The value of T_c ($H = 0$) for the 3D *XY* analysis was determined from slow T sweeps in low fields in an Oxford Instruments vibrating sample magnetometer (VSM), the SQUID magnetometer, and a Lake Shore ac susceptometer. The latter gave a 90%–10% width of 0.4 K, which is ascribed to a distribution of T_c values in the crystal arising from defects or strains, and we therefore use the midpoint, $T_c = 91.3 \pm 0.1$ K.

Figure 1(b) shows our SQUID data on the usual 3D *XY* scaling plot [16,17], namely $M_{\rm fluc}/H^{0.5}T$ vs

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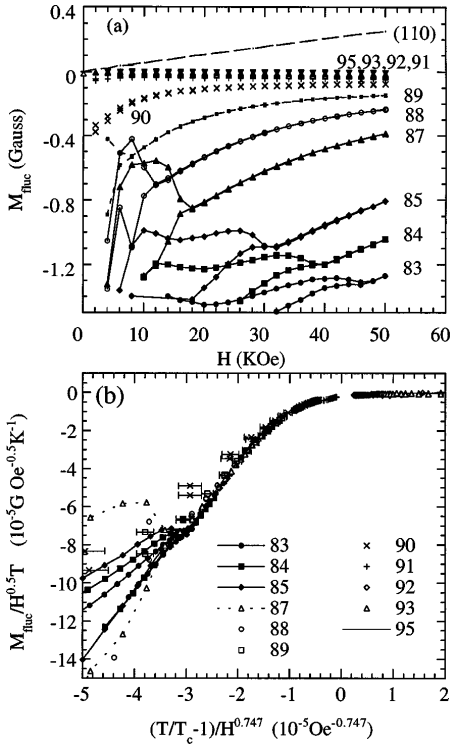

FIG. 1. (a) Magnetization (gauss, i.e., emu/cm^3) for a 5.86 mg $YBa_2Cu_3O_7$ crystal vs magnetic field (kOe) applied $\| c$ axis at the temperatures indicated. The normal state paramagnetism has been subtracted except for the data at 110 K, shown by the dashed line. (b) 3D XY scaling plot of data in Fig. 1(a) for the temperatures indicated, taking $T_c = 91.3 \pm 0.1$ K.

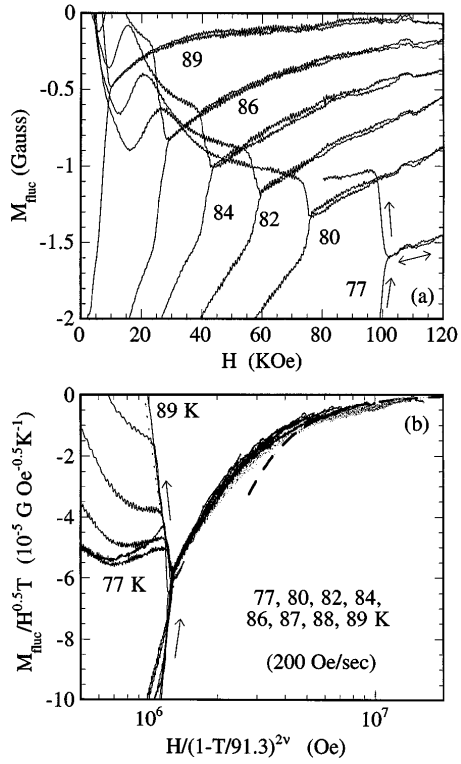

FIG. 2. (a) VSM data for the same crystal as in Fig. 1(a), with $H \| c$, after subtracting the normal state paramagnetism. (b) 3D XY plots of VSM data at the temperatures indicated with the same values of T_c and ν as in Fig. 1. The dashed line shows the $1/y^2$ extrapolation used in the integration of $M_{\rm fluc}$ (see text).

$-t/H^{1/2\nu}$ where $\nu = 0.669$ and $t \equiv 1 - T/T_c$ $(H = 0)$. In the reversible region there is excellent agreement with published work [16,17]. *An important difference is that here the irreversibility fields scale to the same point on the 3D XY plot.* The measurements were extended up to 120 kOe using the VSM, taking $M(H)$ data at fixed T [e.g., Fig. 2(a)] as well as FC and ZFC (field and zero-field cooled) $M(T)$ data (not shown). There is good 3D XY scaling of both M and $H_{\rm irr}$ over a wider range of H and T than previous studies, as shown in Fig. 2(b) on an $H/t^{2\nu}$ scale to emphasise the high field region.

Combined plots of $H_{\rm irr}(t)$ are shown in the main part of Fig. 3. The values from $M(H)$ sweeps at 20–200 Oe/sec are identical and fit the relation $H_{\rm irr} = H^* t^{2\nu}$, with $2\nu \approx \frac{4}{3}$ as expected for 3D XY scaling. The inset of Fig. 3 shows good agreement between $H_{\rm irr}(T)$ from these sweeps and the $H_m(T)$ line at which a latent heat is observed [3]. Both quantities agree well with $H_{\rm irr}$ determined from resistivity data [5] (not shown). The slower SQUID $M(H)$ and VSM $M(T)$ sweeps give 15% lower values for $H_{\rm irr}(T)$—this is probably because there is sufficient flux creep just below $H_m(T)$ for the FLL to respond reversibly on these longer time scales.

The data in Figs. 1(b) and 2(b) show that above $H_{\rm irr}(T)$, $M_{\rm fluc}$ scales as

$$\frac{M_{\rm fluc}(T,H)}{H^{1/2}} = -\frac{Ck_BT}{\Phi_0^{3/2}}\,\frac{\xi_{ab}^0}{\xi_c^0}\,\frac{G'(y)}{y^{1/2}} \qquad (1)$$

as expected in the 3D XY model [16,17,21]. Here Φ_0 is the flux quantum for pairs, ξ_{ab}^0 and ξ_c^0 are the coherence lengths in the ab and c directions extrapolated to $T = 0$ [22], C is a number of order unity, and $G'(y) \equiv dG(y)/dy$, where $G(y)$ is the (unknown) scaling function for the free energy. The parameter $y \equiv H(\xi_{ab}^0)^2/\Phi_0 t^{2\nu}$ can either be viewed as the ratio of the two area scales, $\xi_{ab}^2(t)$ and Φ_0/H, which determines the strength of superconducting fluctuations in a field H applied parallel to the c axis, or as the ratio of H to $H_{c2}(t)$, where $H_{c2}(t) = \Phi_0 t^{2\nu}/2\pi(\xi_{ab}^0)^2$ is the appropriate form of the upper critical field in the critical region [23]. *Thus the fact that H_{irr} occurs at the same value of y implies that H_{irr} is a fixed fraction of $H_{c2}(t)$ [conclusion A].*

The normal state paramagnetism does not affect $F_n - F_s$ significantly because $4\pi M \ll H$ [Fig. 2(a)] so we can take $F_n(T,H) = F_n(T,\infty) \equiv F_s(T,\infty)$. Using the thermodynamic relation $M(T,H) = -\partial F_s(T,H)/\partial H$, Eq. (1) can then be integrated to give [24]

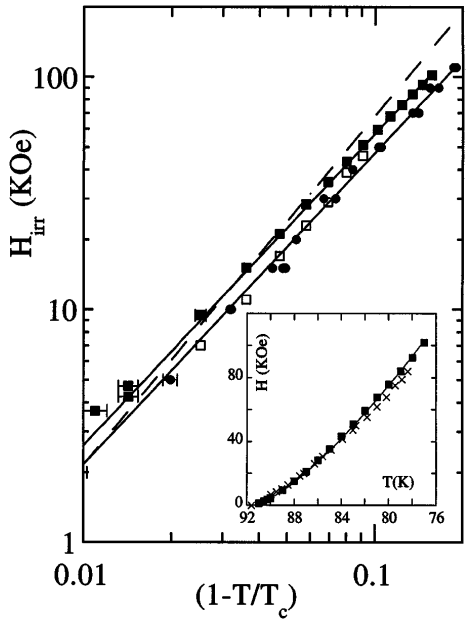

FIG. 3. Log-log plots of the irreversibility fields $H_{\rm irr}$ (in kOe) vs $1 - T/T_c$ for $T_c = 91.3 \pm 0.1$. Solid squares, $H_{\rm irr}$ from VSM $M(H)$ loops at 20–200 Oe/sec. Open squares, from SQUID $M(H)$ data. Closed circles from FC and ZFC $M(T)$ curves taken with the VSM when sweeping T at ± 0.67 K/min. The solid lines are fits to a $\frac{4}{3}$ power law with a $\frac{3}{2}$ law shown by the dashed line. The inset shows $H_{\rm irr}(T)$ from the VSM $M(H)$ data (squares), compared with $H_m(T)$ from calorimetry [3] (crosses).

$$F_n(T,H) - F_s(T,H) = -\int_H^\infty M_{\rm fluc}\, dH = Ck_BTG(y)/\{\xi_{ab}(t)^2\xi_c(t)\}. \quad (2)$$

This relation is a general feature of scaling and renormalization group theories of phase transitions [21,25]. *From Eq. (2), the fact that $H_{irr}(T)$ occurs at a fixed value of y also means that at $H_{irr}(T)$ the free energy in a correlation volume is a fixed multiple of k_BT [conclusion B].* We estimate the actual value of $(F_n - F_s)\Omega(T)/k_BT$ at $H_{\rm irr}(T)$ by integrating our $M(H)$ data, numerically from $H = H_{\rm irr}$ up to ca. 80 kOe, and then analytically at higher fields by making an empirical fit, $G'(y)/\sqrt{y} \propto 1/y^2$ [Fig. 2(b)]. The small contribution above T_c [26] was neglected. For example, at 87 K, $F_n(H_{\rm irr}) - F_s(H_{\rm irr}) = 5.1 \pm 0.6$ mJ/cm^3 or 30% of the total condensation energy at 87 K [26]. If we take $\Omega(0) = 400$ Å^3 from a Gaussian fluctuation analysis of the zero field specific heat above T_c [13] for well-oxygenated $YBa_2Cu_3O_{7-\delta}$, then at $H_{\rm irr}(T)$ the free energy in a coherence volume is $0.8k_BT$. A smaller value, $0.3k_BT$, is obtained if $\Omega(0) = 140$ Å^3 as deduced from a 3D *XY* analysis of the specific heat of a single crystal [14] using the critical amplitude $A^+ = 59$ (mJ/K)/g and the universal constant $R_\xi = 0.3$ [21]. This discrepancy may arise from small differences in oxygen content, which affect $\Omega(0)$ but not $\xi_{ab}(0)$ [13] or because in mean field (MF) and 3D *XY* expressions, prefactors such as $\xi_{ab}(0)$ are not the same [22]. In either case our numerical analysis of conclusion B shows that $H_{\rm irr}(T)$ corresponds to $(F_n - F_s)\Omega(T) \approx k_BT$. This implies that *thermodynamic fluctuations determine $H_{irr}(T)$ of our crystals and also, from the inset to Fig. 3, the melting line of $YBa_2Cu_3O_7$.*

For $0 < \delta < 0.1$, the MF value of $\xi_{ab}(0)^2 = 120$ Å^2 [13], so the MF value of $H_{c2}(0) = 2750$ kOe, is only a factor 2.2 larger than $H^* = 1225$ kOe obtained from the fit, $H_{\rm irr}(t) = H^*t^{4/3}$, in Fig. 3. This numerical analysis of conclusion A suggests that $H_{\rm irr}(t) \approx H_{c2}(t)$, i.e., both $H_{\rm irr}(t)$ and $H_m(t)$ correspond to the formation of a FLL that is suppressed to lower temperatures by critical fluctuations. In support of this we note that the first theoretical prediction [27] of a first order phase transition at $H_{c2}(T)$ was also based on critical fluctuations and that modern scaling treatments apply to both first order and continuous phase transitions [25].

We have also applied this analysis to underdoped $YBa_2Cu_3O_{7-\delta}$. The condensation energy $H_c(0)^2$ obtained by integrating specific heat data [28] is plotted vs δ in Fig. 4. The drastic fall with δ is ascribed to the opening of the normal state gap [29] above T_c which reduces the spectral weight available for superconductivity. The variation of H^* with δ, derived from fits to $H_{\rm irr} = H^*t^{4/3}$ for $0 < t < 0.2$, using resistivity data for crystalline films [30] and magnetic data for single crystals [31], is also shown in Fig. 4. *It can be seen that $H_c(0)^2$ and H^* change by more than 100 but have the same variation with δ.* Although this result could be interpreted in terms of pinning, it is consistent with a critical fluctuation picture. Namely, in the critical region the product $H_c(t)^2\xi_{ab}(t)^2\xi_c(t)$ is independent of t [21,23], and from Eq. (2) at $H = 0$, we have [22]

$$H_c(0)^2(\xi_{ab}^0)^2\xi_c^0 = Dk_BT_c\,, \quad (3)$$

where D is a constant of order 4. Since experimentally $H^* \propto H_{c2}(0) \propto (\xi_{ab}^0)^{-2}$ (conclusion A), Eq. (3) yields $H^* \propto H_c(0)^2$, provided the weaker doping dependence of T_c/ξ_c^0 can be ignored.

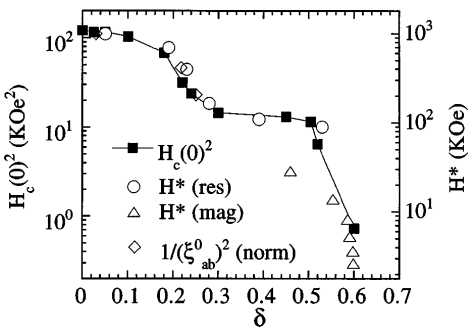

FIG. 4. Left hand scale: plot of condensation energy, i.e., $H_c(0)^2$ vs δ for $YBa_2Cu_3O_{7-\delta}$ obtained by integrating specific heat data from [28]. Right hand scale: values of H^* used in fits of $H_{\rm irr}(T) = H^*t^{4/3}$, vs δ. Circles: H^* from resistivity data [30]; triangles: from magnetic data [31]. Open diamonds show that the normalized values of $1/(\xi_{ab}^0)^2$ from a 3D *XY* scaling analysis [16] vary as H^*.

At lower T, outside the critical region, $H_{\rm irr}$ and H_{c2} will increase faster than $t^{4/3}$ as observed experimentally [31] but thermodynamic fluctuations can still have significant effects [32]. Figure 4 also shows that $(\xi^0_{ab})^{-2}$ obtained from 3D XY analysis of $M(H)$ curves [16] and H^* from resistivity [30] vary in the same way with δ, implying that $H_{\rm irr}$ also occurs at the same value of y for $0 < \delta < 0.25$.

For optimally doped compounds $H_{\rm irr}$ falls drastically as the interlayer coupling is reduced [2]. This agrees qualitatively with Eq. (3), because if $H_c(0)$ and T_c are fixed, then $H_{\rm irr}$ [i.e., $H^*\alpha(\xi^0_{ab})^{-2}$] falls as the anisotropy $\gamma \cong \xi^0_{ab}/\xi^0_c$ increases. The recent observation of a broad 3D XY scaling region for highly anisotropic $Bi_2Sr_2CaCu_2O_8$ crystals [33] below T_c seems to support this viewpoint. But for $Bi_2Sr_2CaCu_2O_8$ further work is needed to clarify the physical meaning of the large value of ξ^0_{ab} obtained using the 3D XY approach, the fact that $H_{\rm m}$ and $H_{\rm irr}$ can be very different [34] and the level of quantiative agreement with Eq. (3).

We conclude that the irreversibility line of our $YBa_2Cu_3O_7$ crystals, and (by association) the first order melting transition, are determined by thermodynamic fluctuations, because they correspond to the line where the free energy in a coherence volume is a fixed fraction of k_BT (of order unity). This suggests an intrinsic upper limit to $H_{\rm irr}(T)$ irrespective of the amount of disorder. The observation that $H_{\rm irr}(T)$ and $H_m(T)$ are proportional to $H_{c2}(T)$ (conclusion A) may indicate that $H_m(T)$ is actually the $H_{c2}(T)$ line which has been suppressed by critical fluctuations.

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