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Vortex correlations in the superconducting transition of $YBa_2(Cu_{1-x}Al_x)_3O_{7-\delta}$ films in high magnetic fields

Dimitrios G. Xenikos, Jin-Tae Kim, and Thomas R. Lemberger
Department of Physics, The Ohio State University, Columbus, Ohio 43210
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We determine the characteristic length scale for vortex correlations in $YBa_2(Cu_{1-x}Al_x)_3O_{7-\delta}$ films [$0 \le x \le 0.03$] by studying their current-voltage characteristics in fields $0.5\ T \le B \le 6\ T$. We find that critical scaling persists over 60% of the resistive transition (which widens from 3 to 30 K as doping concentration and field increase) and breaks down when the correlation length is 500–1250 Å, a value that *does not* depend on the intervortex spacing. This result suggests that pinning in as-made films of $YBa_2Cu_3O_{7-\delta}$ is dominated by extended rather than microscopic defects.

Vortex pinning in disorder high-temperature superconductors has generated a number of different theoretical models, which deal with the static and dynamical characteristics of the mixed state and often introduce phases with qualitatively different correlations among the vortices. There is strong experimental evidence from current-voltage (I-V) curves in $YBa_2Cu_3O_{7-\delta}$ (YBCO) films[1] and twinned crystals[2] supporting the idea of a phase boundary associated with the "irreversibility line"[3] in these materials. The details, though, of the pining mechanism that gives rise to the critical behavior are still a matter of controversy. Fisher and co-workers[4,5] model vortex behavior when vortex pinning is from microscopic quenched disorder, and they find a second-order phase transition separating a vortex-glass phase with vanishing linear resistivity form a vortex-fluid phase with a finite low-voltage resistivity. However, the nature of the phase transition may be quite different from that of the vortex-glass theory, if *correlated disorder* in extended defects is responsible for the flux pinning in these samples.[6] Since both models are consistent with measured exponents and scaling functions for $V(I)$, one must look to nonuniversal parameters to distinguish them.

In this paper, we investigate the type of disorder that pins vortices in the glass state in high-T_c films by examining the length ξ_G associated with vortex correlations above the glass transition. We report that critical scaling persists over temperatures where the spatial extent ξ_G for phase correlations of the superconducting order parameter exceeds a length ξ^* (500–1250 Å) that *does not* depend on the intervortex separation. Such mesoscopic length scale is absent from models that assume pinning only in pointlike disorder, including the vortex-glass model. To investigate the generality of this result, we dope our YBCO films by substituting up to 3% of Cu with Al. This substitution leaves the critical exponents and scaling functions, theoretically universal quantities, unchanged, although it enhances thermal fluctuations and reduces $T_g(B)$. The length ξ^* is hardly affected by doping, and it is independent of the intervortex separation.

The films are made by co-deposition of Y, BaF_2, Cu, and Al, with an *ex situ* postanneal in oxygen.[7] They are 1000-Å thick, single-phase, and highly oriented with the c axis perpendicular to the substrate.[7] Doping suppresses the zero-field transition temperature measured by *ac* susceptibility at only 1.2 K/at. % of Al, a relatively weak suppression compared to other dopants like Ni and Zn.[7,8] The films are patterned with standard photolithographic techniques for four-wire measurements, with the strip between the two voltage leads typically $100 \times 42\ \mu m$. The normal state resistivity $\rho(T)$ is metallic, and $\rho(95\ K)$ increases from 110 $\mu\Omega$ cm for undoped YBCO films to 150 and 200 $\mu\Omega$ cm in the 2% and 3% Al doped films, respectively. Scanning-electron microscopy (SEM) pictures show no evidence of misoriented grains in the surface topography. The most prominent defects we observe in the doped samples are holes about 1000 Å in diameter which occur in groups of 1–5 clustered about 1 μm apart. The clusters are typically 20 μm apart. These holes may arise from the collective release of fluorine gas from the films during postannealing. The similarity of our results with results on laser ablated films[1] without such defects indicates that they are not of fundamental importance here.

The inset in Fig. 1 shows I-V curves for the 2 at. % Al film at $B = 4$ T parallel to c axis at temperatures evenly spaced by 1 K from 54.3 to 73.3 K (all logarithms in this work are at base 10). These I-V's are typical of all films at all fields studied. We define the glass temperature, $T_g = 62 \pm 1$ K, to be the highest temperature at which the I-V fits (to $\pm 30\%$) to a single power law, within our voltage resolution (50 nV–1 mV). *All* I-V's above T_g exhibit systematic upward curvature, thus restricting the uncertainty of T_g to ± 1 K. Figure 1 summarizes $T_g(B)$ [or, $B_g(T)$] for films with 0%, 2%, and 3% Al. Note that T_g for the 2% and 3% Al films is much lower than for undoped YBCO. Surprisingly, doping at this level weakens pinning and enhances flux wandering. It does not change the scaling functions and exponents of the transition. In the inset of Fig. 2, we demonstrate how the I-V's above T_g from different samples collapse on the same scaling curve. The two fitting parameters are the low-voltage linear resistivity $\rho(T)$, and the crossover current density $J_0^+(T)$ separating linear from nonlinear conductivity.

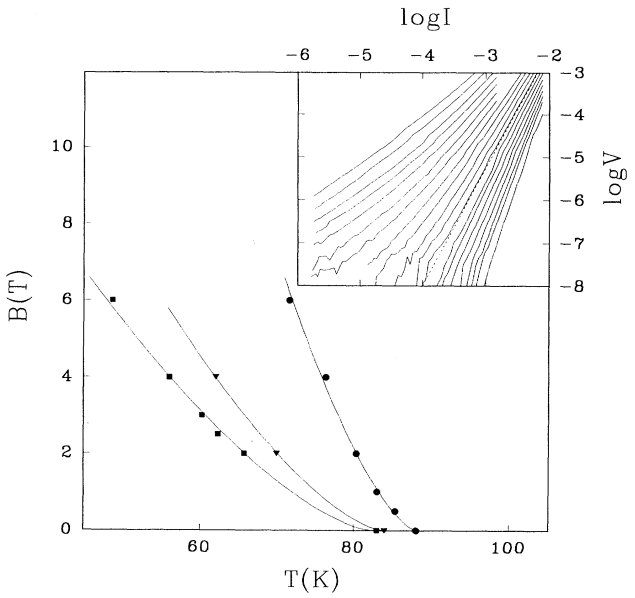

FIG. 1. Transition temperature T_g vs magnetic field, for pure YBCO (circles), 2% Al (triangles) and 3% Al doped films (squares). The solid lines are fits $B \sim (T_c - T_g)^{3/2}$, with T_c where $\rho = 0$ at $B = 0$. Inset: Typical I-V data (in mks units) for a 2% Al film at $B = 4$ T, from 73.3 to 64.3 K. The dashed line indicates the T_g at 62±1 K.

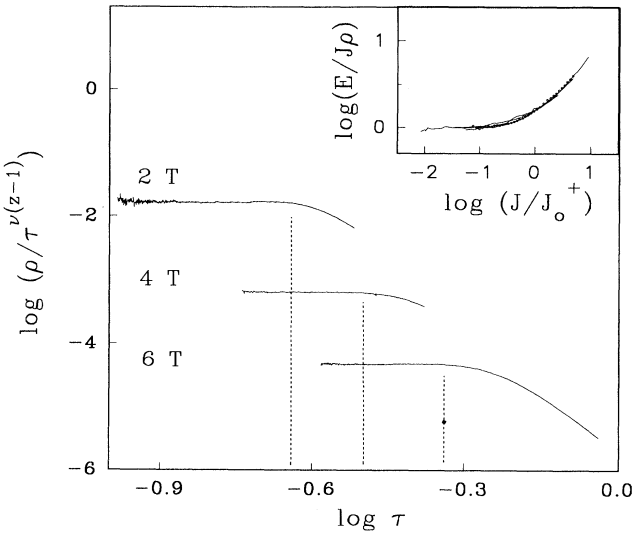

FIG. 2. Normalized resistivity (mks units) from a 3% Al film vs $\tau \equiv (T/T_g - 1)$ for $B = 2$, 4, and 6 T [with $\nu(z-1) = 7.4$, 7.2, and 7.1 respectively]. The dashed lines indicate τ^* which bounds the critical regime at high T. Inset: The scaling functions $E/J\rho$ vs J/J_0^+ for a 0% (lines) and a 3% (circles) film collapse on the same curve.

We extract the critical exponents z and ν of the transition by fitting ρ and J_0^+ to the vortex-glass relations $\rho \propto (T - T_g)^{\nu(z-1)}$ and $J_0^+ \propto (T - T_g)^{2\nu}$. This provides another way to define T_g, that leads to the same value of T_g (within 2 K) with its operational definition from the I-V's. Table I shows values for the exponents $\nu(z-1)$ and 2ν, as well as $(z+1)/2$ from the slope of the I-V at T_g [$V \propto I^{(z+1)/2}$], which are in excellent agreement with values reported for YBCO films[1] and twinned crystals.[2] Theoretical models (other than vortex glass) would predict similar scaling laws with different expressions of the exponents in terms of ν and z.[6] In any case, the fact that scaling occurs with the same exponents and scaling functions, even as the width of the resistive transition increases from 3 to 30 K with increasing doping and field, supports the existence of a phase transition at $T_g(B)$ and our use of appropriate models to interpret our data.

Beyond scaling arguments, it is possible to estimate the *magnitude* of the vortex correlation length $\xi_G(T)$ in a model-independent fashion. In particular, we are interested in estimating ξ_G at the temperature T^* where the scaling breaks down, and associate its value $\xi^* = \xi_G(T^*)$ to some important mesoscopic length scale for vortex correlations. We define T^* as the temperature where $\rho(T)$ decreases 5% below the value predicted by the scaling power law $\rho \propto (T - T_g)^{\nu(z-1)}$. To illustrate, Fig. 2 shows $\rho(T)/(T/T_g - 1)^{\nu(z-1)}$ vs $(T/T_g - 1)$ for the 3% Al doped film at $B = 2$, 4 and 6 T. Dividing out the strongest T dependence of ρ emphasizes the break at T^*. The value of T^* and thus our conclusions are not particularly sensitive to our criterion of 5%, which determines T^* to ± 1 K. Interestingly, the critical scaling persists over 60±5% of the width of the resistive transition, even though the width varies from 3 to 30 K. Here, we take the onset of the resistive transition to be where $\rho(T)$ is about 75% of the extrapolated normal state resistivity. This corresponds to a suppression in the mean-field transition temperature of ~0.6 K/T, about the same as in pure YBCO. The significance of this "universal" behavior for T^* is unknown.

TABLE I. Critical scaling parameters of $YBa_2(Cu_{1-x}Al_x)_3O_{7-\delta}$ films. The uncertainties represent possible systematic errors in the calculations.

% Al	B (T)	$(z+1)/2$ (±0.1)	$\nu(z-1)$ (±0.4)	2ν (±0.6)	ξ^* (±40) Å
3	6	3.0	7.1	2.8	521
3	2	3.0	7.4	3.0	508
2	4	2.8	6.7	3.6	462
2	2	2.8	6.7	3.4	465
0	6	2.8	7.1	3.2	441
0	2	3.0	7.1	3.4	438
0	0.5	3.1	7.2	3.2	439

We now estimate $\xi_G(T)$ using the crossover current density J_0^+, which we define from the scaling forms, as the current density J where $E/\rho J = 1.5$ (inset of Fig. 2). Since all I-V's in the critical regime collapse on the same scaling curve, other choices of this ratio between 1 and 3 do not affect our results. We use the conventional expression[5,6] $J_0^+ \phi_0 \xi_\parallel \xi_\perp \approx kT$ that does not depend on the particular model for the phase transition, to determine $\xi_G^2 \equiv \xi_\parallel \xi_\perp$. Physically, this relation means that nonlinear behavior occurs when the current density is large enough to produce translations of correlated vortex segments of length $\xi_\parallel$ over distance $\xi_\perp$ perpendicular to the current, as readily as thermal fluctuations do. At $J < J_0^+$ translations of correlated segments are thermally induced, while at $J > J_0^+$, Lorenz-force-induced translations dominate. Since $J_0^+ \propto (T - T_g)^{2\nu}$, the size of vortex fluctuations diverges at T_g as $\xi_G^2 \sim (T - T_g)^{-2\nu}$, consistent with an increasing number of vortices becoming correlated and moving coherently as $T \to T_g$. To clarify the notation, we define $\tau \equiv (T/T_g - 1)$ and write $\xi_G = \xi^*(\tau^*/\tau)^\nu$. Then,

$$J_0^+ \simeq \frac{kT}{\phi_0 \xi^{*2}} \left[\frac{\tau}{\tau^*} \right]^{2\nu}.$$

Figure 3 shows $\xi_G^{-2} = J_0^+(T)\phi_0/kT$ vs τ/τ^* for a 3% Al film for $B = 2$ and 6 T. The fact that the two data sets overlap means that both the magnitude and scaling law for $\xi_G(T)$ are independent of B, and hence the average intervortex spacing. The intercept at $\tau/\tau^* = 1$ for this film yields $\xi^* = 515 \pm 40$ Å, which exceeds somewhat the intervortex spacing $a_v \equiv (\phi_0/B)^{1/2} \simeq 190$ Å and 320 Å for $B = 6$ T and 2 T, respectively. Taking into account the uncertainties of our estimation in T_g, T^* and the exponent $\nu(z-1)$, we find $\xi^* \propto B^{0 \pm 0.03}$, i.e., an extremely weak dependence of ξ^* on the applied field. This is the main result of this work and it holds for all the samples we studied, and for the laser ablated films of Ref. 1. Table I records the values of ξ^* obtained for each film. Al doping up to 3 at. % has a small 15% effect on the magnitude of ξ^*. Evidently film morphology affects ξ^*. In a 2500-Å-thick YBCO film with a sharp transition, we found $\xi^* = 1240 \pm 60$ Å. In an unusually resistive 2500-Å-thick YBCO film with a wider superconducting transition but the same scaling exponents we found $\xi^* = 732 \pm 40$ Å. Analyzing the measured $J_0^+(T)$ for $B = 2$, 3, and 4 T from Koch *et al.*[1] on laser-ablated 4000-Å-thick YBCO films, we find $\xi^* = 1100 \pm 200$ Å, independent of the magnetic field.

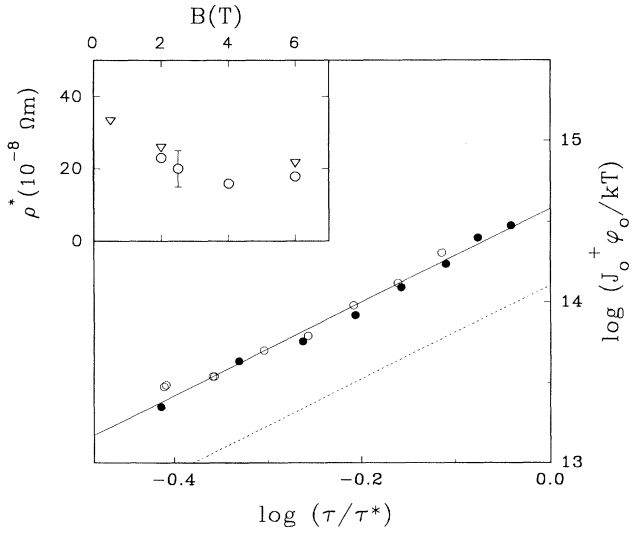

FIG. 3. Plot of $\xi_G^{-2} = (J_0^+ \phi_0/kT)$ of a 3% Al film for $B = 2$ T (filled circles) and $B = 6$ T (open circles) vs τ/τ^*. The fit (solid line) corresponds to $\xi_G^2 = (515\ \text{Å})^2(\tau/\tau^*)^{-3.0}$. If ξ^* were proportional to the intervortex spacing, then the 2 T data would fall on the dashed line. Inset: The resistivity $\rho(T = T^*)$ vs B for the undoped (triangles) and 3%-Al-doped film (circles). The uncertainty in ρ^* is indicated by the error bar in one data point. (All quantities in mks units.)

What is the significance of this result? First we consider the case that ξ^* strongly depends on the behavior at $T \geq T^*$, which is possible if T^* marks the crossover between two regimes with different scaling characteristics. A three-dimensional XY critical scaling model proposed[9] to describe the conductivity around T_c is consistent with our observation that both boundaries $T^*(B)$ and $T_g(B)$ scale with field with the same XY-like exponent $[(T_c - T^*) \propto (T_c - T_g) \propto B^{1/2\nu'}, \ \nu' \approx 0.7]$. However, an attempt to scale our $\rho(T)$ data in the vicinity of T^* to the expected form[9] $\sigma H^{1/2} = s(\xi_{GL} H^{1/2})$[$H \approx B$, σ = ohmic conductivity, $\xi_{GL} \propto (T_c - T)^{-\nu'}$ the coherence length] leads to nonuniversal scaling functions $s(\xi_{GL} H^{1/2})$, strongly dependent on dopant concentration. This is true even if we consider an increase of the magnitude of ξ_{GL} to accommodate the broadening of the transition with doping.

We then consider the magnitude of ξ^* in terms of vortex correlations. In any model for the phase transition at T_g, the critical response must involve correlated motion of multiple vortices.[10] Hence, the correlation length ξ_G (or at least $\xi_\perp$) should be larger than the average intervortex spacing in the critical region. Models, in which pinning is from quenched disorder with an average spacing between pinning centers much smaller than the intervortex spacing, lead to the conclusion that scaling should break down when the correlation length is roughly the intervortex spacing (a field-dependent quantity). In the vortex-glass theory in particular, the spatial extent of phase coherence of the superconducting order parameter ξ_G is assumed to exceed the range of translational order of the flux lattice. Thus, the critical scaling is expected to break down when ξ_G equals the Larkin-Ovchinnikov length,[11] which is at most a few flux-lattice spacings.[5] Again, ξ^* should be quite sensitive to B and T, in sharp contrast to our observations.

A natural interpretation of our result is that ξ^* reflects the underlying pinning structure in the films. Quite generally, models for critical scaling in samples where correlated disorder dominates pinning, require the transverse displacement $\xi_\perp$ of liberated vortex segments to be at least the distance between neighboring macroscopic defects in the sample,[6] and thus lead to a field-independent value for ξ^* at the temperature where critical scaling breaks down. Equally probable sites for flux pinning in our films

are grain boundaries, twin planes perpendicular to the substrate, and columnar defects along the c-crystallographic axis. A typical grain size of 500 Å in YBCO films has been measured by TEM,[12] a value close to our ξ^*. Pinning in twin planes is also shown to dominate the regime above the T_g in twinned YBCO crystals.[13] Finally, assuming that only columnar defects such as screw dislocations in crystallites or edge dislocations in grain boundaries are responsible for the pinning, a Bose-glass[6] calculation leads to a reasonable $d \simeq 200$ Å for the distance between adjacent defects.[14] Although our experiment cannot decide for the nature of the extended defects responsible for the critical scaling, it suggests that their structure is dictated by the morphology of the substrate and/or misfits in the substrate-film interface rather than the dopant concentration, since Al doping has only a 15% effect on ξ^*. Furthermore, the variation of ξ^* with film thickness in undoped films suggests that defects change somewhat when the film thickness exceeds the typical grain size. It remains to be understood how Al doping weakens the pinning properties of the extended defects.

As a final point, we discuss the striking result that the values of $\rho^* \equiv \rho(T=T^*)$ are clustered around 22 $\mu\Omega$ cm for all our samples and at all fields (inset in Fig. 3). Laser-ablated films also show $\rho^* \approx 25$ $\mu\Omega$ cm at fields $B=2-4$ T.[1] We can show that ρ^* and ξ^* set a strong constraint for the magnitude of the relaxation time $t=t^*(\xi_G/\xi^*)^z$ associated with the critical slowing down. We estimate t^* by considering a vortex excitation of area ξ^{*2}, formed when current density J_0^+ passes through the sample at temperature T^*. The energy gained in the system by the generation of the excitation is $J_0^+ \phi_0 \xi^{*2} \approx kT^*$. As this excitation relaxes, it dissipates energy at a rate $\int \mathbf{J}\cdot\mathbf{e}d^2x$, with $\mathbf{J}$ and $\mathbf{e}$ being the local current density and local electric field caused by the excitation, integrated over volume. Assuming that the integral is significant only at the core of the vortex of radius ξ_{GL}, and that $\mathbf{e} \approx \rho^* J_0^+ (a_v/\xi_{GL})$ is the macroscopic electric field $E \approx \rho^* J_0^*$ increased by the geometric factor a_v/ξ_{GL}, we obtain $\int \mathbf{J}\cdot\mathbf{e}d^3x \approx J_0^+ e \xi_{GL}^2 \xi^*$. Letting t^* be the time for the fluctuation to dissipate its energy of kT, we find $t^* \approx (\xi^{*3}\phi_0^2)/(a_v \xi_{GL} kT^* \rho^*)$. Olsson *et al.*[15] find from the complex impedance of a 2500-Å-thick YBCO film at $B=0.55$ T that critical scaling persists for $t \geq 2\times 10^{-8}$ s. Using values for ξ^* and ρ^* from our 2500-Å pure YBCO film, and $\xi_{GL} \approx 100$ Å at $T^*=88$ K, we estimate $t^* \approx 3\times 10^{-8}$ s, in reasonable agreement with the experiment. As B increases and T^* moves away from $T_c(B)$, inflated values of ξ_{GL} are avoided and the system should become slower with $t^* \propto a_v^{-1}$. One should note however, that a different picture for the vortex excitations could lead to a new combination of the relevant length scales (including possibly an anisotropic ξ_G) in the estimation of t^*. The issue may be resolved by dynamical measurements in an extended region of magnetic fields and film thicknesses. These measurements have the potential to estimate directly the relaxation time of the vortex excitations, which reflects the dynamics of the flux lines as they cross and recombine in the sample.

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