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Effect of oxygen stoichiometry on the vortex-glass phase transition in $YBa_2Cu_3O_{7-\delta}$ thin films

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A series of $YBa_2Cu_3O_{7-\delta}$ epitaxial thin films were prepared with values of δ ranging from 0.05 to 0.65. Electric field as a function of current-density isotherms could be scaled to a universal scaling function over the full δ range and the critical parameters determined were nearly independent of δ. The vortex-glass phase-transition temperature was determined as a function of magnetic field at all of the values of δ studied and the phase boundary was observed to broaden with δ. Based on determinations of the ratio of the zero-temperature coherence length ξ_{c0} to c-axis lattice parameter using normal-state fluctuation conductivity, the anisotropy was shown to increase with δ while ξ_{c0} decreases from 1.6 to 0.8 Å as δ ranges from 0.05 to 0.37.

It has been well established that the superconducting transition temperature T_c of $YBa_2Cu_3O_{7-\delta}$ (YBCO) decreases and the spacing d between pairs of the superconducting CuO_2 planes (CuO_2 bilayers) increases as δ increases. Several recent results on deoxygenated YBCO suggest that as oxygen is removed the "irreversibility line" moves down in temperature and field.[1,2] This is consistent with studies on more anisotropic high-temperature superconductors where the field-induced resistive broadening of the superconducting transition was shown to depend on the interplanar coupling and the width of the insulating layers between the CuO_2 multilayers in these materials.[3] Here we present a determination of the δ dependence of the temperature at which the linear resistivity vanishes (the vortex-glass transition). The critical parameters associated with this continuous transition as well as the anisotropy of the superconducting properties of YBCO are studied over a wide range of δ that includes both the 60 and 90 K plateaus.

Epitaxial YBCO thin films, with the c axis perpendicular to the film plane, were prepared by pulsed laser deposition on heated (001) $LaAlO_3$ substrates as previously described.[4] Each thin-film sample was about 4000 Å thick and initially had a superconducting transition temperature $T_c = 90 \pm 2$ K, with a transition width ΔT_c of about 2 K when measured by ac susceptibility (χ_{ac}) at 1.5 MHz. The oxygen stoichiometry of these films was modified by annealing these films at 400 °C in a controlled flowing mixture of oxygen and argon gases followed by a rapid quench to room temperature. During annealing the oxygen partial pressure was monitored and controlled using an yttria-stabilized zirconia solid-electrolyte oxygen-concentration cell.[5] A separate film was prepared for each oxygen stoichiometry. The c-axis lattice constant **c** was determined using θ-2θ x-ray-diffraction patterns while the values of δ were determined using an idometric titration[6] on polycrystalline YBCO samples that were prepared under identical conditions to those of the films. The values of T_c for a given δ (for both films and bulk samples) corresponded closely with previous studies[7–9] resulting in an estimated error of about 0.05 in the values of δ used here.

The films were patterned using laser ablation into bridges having dimensions of 100 μm wide by 2000 μm long. A standard four-point probe technique was used for the temperature-dependent resistivity measurements $\rho(T)$ and electric field as a function of current density $E(J)$ measurements using procedures described previously.[10] The dc magnetic field (H), ranging from 0 to 50 kOe in steps of 10 kOe, was applied parallel to the crystallographic c axis.

The strong dependence of the physical properties of YBCO on oxygen stoichiometry is evident in Fig. 1, which compares $\rho(T)$ measurements taken in magnetic fields (H) ranging from 0 to 50 kOe for samples with $\delta \sim 0.05$ ($T_c = 87.7$ K) and $\delta \sim 0.37$ ($T_c = 57.0$ K). As δ is increased, T_c is observed to decrease, and there is an increase in the broadening effect associated with the applied magnetic field. For highly anisotropic superconductors, the increase in field broadening has been attributed to the weaker coupling of two-dimensional pancake vortices as the CuO layer spacing increases.[3] The CuO_2 bilayer spacing of YBCO has been found to increase as δ increases,[8] and it has been predicted that the temperature range over which this quasi-two-dimensional behavior occurs increases as the CuO_2 layer spacing increases.[11] The increased broadening effect of a magnetic field on the $\rho(T)$ curves shown in Fig. 1 would then appear to be consistent with YBCO becoming more anisotropic as δ increases.[11,3] The inset of Fig. 1 shows zero-

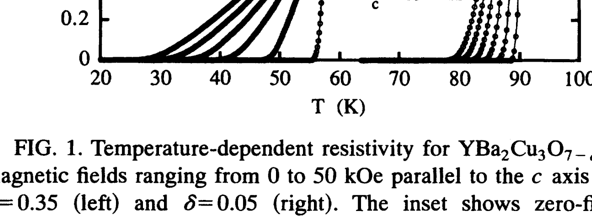

FIG. 1. Temperature-dependent resistivity for $YBa_2Cu_3O_{7-\delta}$ in magnetic fields ranging from 0 to 50 kOe parallel to the c axis for $\delta = 0.35$ (left) and $\delta = 0.05$ (right). The inset shows zero-field $\rho(T)$ curves at $\delta = 0.65$, 0.37, 0.12, 0.05, and 0.05 normalized to their respective resistance values at 100 K.

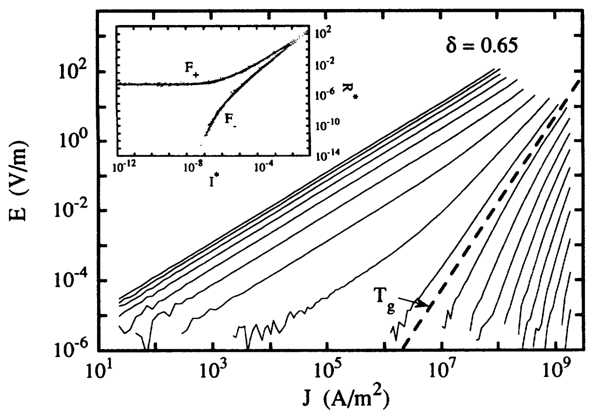

FIG. 2. $E(J)$ isotherms at $H=10$ kOe for a YBCO film with $\delta=0.65$. Temperatures range from 14 K (lower right) to 50 K (upper left) in increments of 2 K. The dashed line labeled T_g is the vortex-glass transition. The inset with axes of $R^*=(V/I)/|T-T_g|^{\nu(z-1)}$ and $I^*=(I/T)/|T-T_g|^{2\nu}$ shows the same $E(J)$ isotherms scaled about T_g.

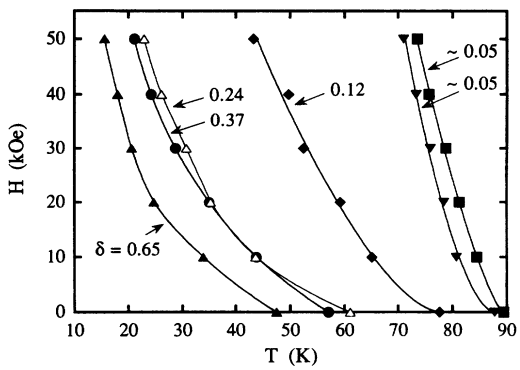

FIG. 3. Vortex-glass phase boundaries determined for YBCO films with $\delta=0.65$, 0.37, 0.12, 0.05, and 0.05.

field $\rho(T)$ curves for samples with δ values ranging from 0.05 to 0.65 that have been normalized to their respective resistances at 100 K (R_{100}). These normalized $\rho(T)$ curves demonstrate that T_c shifts systematically to lower temperature with increasing δ.[8]

YBCO thin films having δ values as large as 0.65 when prepared under the conditions described above have $E(J)$ isotherms in applied magnetic fields that show evidence for a transition into a true superconducting state with zero linear resistance. Figure 2 is a log-log plot of the $E(J)$ isotherms for a film with $\delta\sim0.65$ ($T_c=48.5$ K) in an applied field of 10 kOe. These $E(J)$ isotherms range from 14 K at the lower right to 50 K at the upper left, in steps of 2 K. As observed previously in stoichiometric YBCO thin films,[10,12] below T_c these $E(J)$ isotherms have a characteristic temperature dependence with negative curvature at the lowest temperatures, positive curvature at intermediate temperatures, and ohmic behavior at the highest temperatures. At the line labeled T_g in Fig. 2, a power-law current-density dependence $E\sim J^{2.6}$ was determined using $E(J)$ data sets having higher resolution ($\Delta T=0.25$ K) near the crossover from negative to positive curvature. This power-law isotherm identifies the vortex-glass transition temperature T_g,[12,13] which is characterized by the loss of linear resistivity.

The vortex-glass phase transition evident in the $E(J)$ behavior of these YBCO thin films was analyzed using the scaling relation $E(J)\sim J\xi^{\nu(z-1)}F_\pm(J\xi^{-2\nu})$, where $\xi\sim|T-T_g|^{-\nu}$ is the vortex-glass coherence length, ν and z are the spatial and dynamic critical exponents, and $F_\pm$ is a universal scaling function above (F_+) and below (F_-) T_g.[12,13] The inset of Fig. 2 shows how the $E(J)$ data collapse onto the two universal scaling functions when the $E(J)$ data are plotted as $R^*=(V/I)/|T-T_g|^{\nu(z-1)}$ versus $I^*=(I/T)/|T-T_g|^{2\nu}$ for a unique set of ν and z values.[12–14] The critical exponents were observed to be nearly independent of δ with average values, $\nu=1.8\pm0.1$ and $z=4.5\pm0.6$, which are the same values observed previously in stoichiometric YBCO films[12,15,16] and single crystals with strong disorder.[17] Plots of $[d\ln(\rho)/dT]^{-1}$ versus T were used to provide an independent determination of T_g and the quantity $\nu(z-1)$.[18,19] These were in agreement with the values determined from $E(J)$ scaling. The apparent independence of the critical scaling exponents ν and z on δ implies that these exponents are universal; that is, they depend only on the dimensionality of the superconductor at T_g. This universal scaling behavior is strong evidence that T_g is a true continuous phase transition.[12]

H-T phase diagrams were determined for deoxygenated YBCO thin films having δ values ranging from 0.65 to 0.05. Shown in Fig. 3 are the vortex-glass phase boundaries $H_g(T)$ determined from both $V(I)$ curves at 10 and 50 kOe and plots of $[d\ln(\rho)/dT]^{-1}$ versus T at all other applied fields. The $H=0$ kOe points are the T_c values determined from $\rho(T)$ and χ_{ac} measurements. $H_g(T)$ broadens significantly as δ increases, with the temperature difference $\Delta T=(T_c-T_g)$, where we use T_c at $H=0$ and T_g at $H>0$, is equal to about 10 K for $\delta=0.05$ and $\Delta T\sim40$ K for $\delta=0.65$ when $H=50$ kOe. The temperature dependence of $H_g(T)$ is also observed to change with δ. For $\delta<0.12$, $H_g(T)\sim(1-T/T_c)^x$, with $x=1.3$ to 1.5, which is consistent with previous observations of $H_g(T)$ in anisotropic three-dimensional high-temperature superconductors.[17] In contrast to this, for $\delta>0.12$, $H_g(T)$ can no longer be described by a simple power-law temperature dependence and the behavior in this regime is more consistent with observations of the vortex-glass transition in quasi-two-dimensional superconductors, such as $Tl_2Ba_2CaCu_2O_8$ (Ref. 3) and $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Ref. 19), which show a stronger temperature dependence at low magnetic fields than at high magnetic fields.[13] This is an indication that YBCO crosses over to quasi-two-dimensional superconducting behavior for $\delta>0.12$ ($T_c<77.6$ K) and $H<50$ kOe. A more complete H-T phase diagram for oxygen-deficient YBCO would then include a crossover from two- to three-dimensional behavior at temperatures between $T_c(0)$ and $T_g(H)$, since the vortex-glass phase transition will only occur at nonzero temperature in three dimensions.[13]

The Lawrence-Doniach (LD) model for layered superconductors[20] has been used to describe the zero-field fluctuation conductivity σ_{fl} of YBCO.[21] Within the LD description, the Josephson coupling between the superconducting planes, separated by a distance d, increases as T_c is

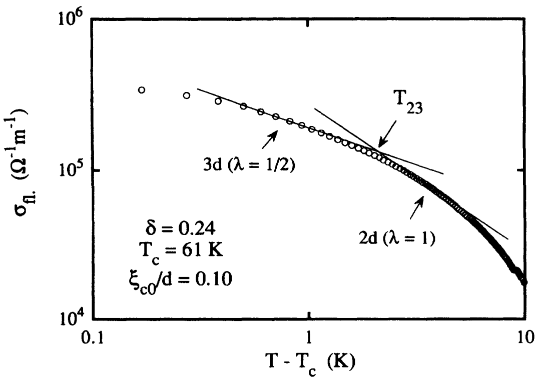

FIG. 4. Excess conductivity versus $(T-T_c)$ on log-log scale. Lines labeled 3d and 2d have slopes of $-\frac{1}{2}$ and -1, respectively, corresponding to the two- and three-dimensional limiting behaviors. T_{23} identifies the crossover between the two- and three-dimensional regimes, determined from the intersection of the 3d and 2d lines. From the value of T_{23} the ratio ξ_{c0}/d can be determined.

approached from above, since the c-axis coherence length $\xi_c(T)=\xi_{c0}|(T-T_c)/T_c|^{-1/2}$, where ξ_{c0} is the c-axis coherence length at $T=0$ K, diverges as T_c is approached. At a temperature near T_c it is expected that a crossover from two-dimensional to three-dimensional fluctuations will occur when $\xi_c(T)\approx d/2$.[21,22] As a result of this crossover, there are two limiting forms for the fluctuation conductivity given by $\sigma_{fl}\sim[(T-T_c)/T_c]^{-\lambda}$; one form is for the three-dimensional limit in which the CuO_2 planes are strongly coupled ($\lambda=\frac{1}{2}$), and the other is the quasi-two-dimensional limit in which the CuO_2 planes are weakly coupled ($\lambda=1$).[21]

The zero-field fluctuation conductivity is experimentally determined from $\sigma_{fl}=[1/\rho(T)-1/\rho_n(T)]$, where $\rho(T)$ is the measured resistivity and $\rho_n(T)$ is determined from extrapolations of the normal-state resistivity to low temperatures.[21] In the present study it was only possible to determine $\rho_n(T)$ for films when the superconducting transition temperature was near either 60 or 90 K (on, but not between, the 60 and 90 K plateaus). For $T_c\sim 90$ K ($\delta<0.12$) the $\rho_n(T)$ curves were determined by extrapolating to low temperatures using a linear fit to $\rho(T)$ in the range of 200 to 280 K,[21,23] while for $T_c\sim 60$ K ($0.37\geqslant\delta\geqslant 0.24$) the $\rho_n(T)$ curves were determined by extrapolation using a power-law fit, $\rho_n\sim T^{2.5}$, to $\rho(T)$ in the range of about 120 to 180 K.[24] Figure 4 is a log-log plot of σ_{fl} versus $(T-T_c)$ for a film with $\delta=0.24$. The lines in Fig. 4 labeled 2d and 3d indicate the regions where σ_{fl} is consistent with the two-dimensional ($\lambda=1$) and three-dimensional ($\lambda=\frac{1}{2}$) limits of the LD model, respectively, while the crossover temperature between two- and three-dimensional fluctuation regimes is labeled T_{23}. T_{23} in this figure occurs at $(T_{23}-T_c)\cong 2$ K. The quantity $(T_{23}-T_c)$ is observed to decrease with increasing δ, ranging from a maximum of 6.7 K at $\delta=0.05$ to about 1.1 K at $\delta=0.65$. The observation that T_{23} occurs unobservably close to T_c in the quasi-two-dimensional high-temperature superconductor $Bi_2Sr_2CaCu_2O_x$,[25] which is much more anisotropic than YBCO, and the observation that $(T_{23}-T_c)$ decreases with increasing δ are both consistent with YBCO becoming more anisotropic as δ increases.

Since the strength of coupling between CuO_2 bilayers in YBCO should increase with ξ_{c0} and decrease with d, the ratio ξ_{c0}/d can be a useful qualitative measure of the anisotropy as a function of δ. In the LD model, this ratio may be expressed in terms of T_{23} as $\xi_{c0}/d=(1/2)[(T_{23}/T_c)-1]^{1/2}$ (Refs. 21 and 25). The value of ξ_{c0}/d is observed to decrease by a factor 2 as δ is varied from 0.05 (on the 90 K plateau) to 0.37 (on the 60 K plateau). The value of **c** is observed to change approximately linearly with δ at a rate of about -0.079 Å per oxygen atom as δ increases. This represents less than a 0.2% change in **c** as δ changes from 0.05 to 0.37. Thus, the change in ξ_{c0}/d is due primarily to changes in ξ_{c0}, which decreases by a factor of 2 from 1.6 Å at $\delta=0.05$ to 0.82 Å at $\delta=0.37$. These values of ξ_{c0} are within 0.5 Å of previously reported values for stoichiometric and oxygen-deficient YBCO crystals.[1,26] For the three ξ_{c0} values that could be determined, there appears to be linear dependence on δ which extrapolates to zero at $\delta=0.69$.

In summary, a series of $YBa_2Cu_3O_{7-\delta}$ thin films with a wide range of δ values were prepared using a method in which samples were annealed at the same temperature under different oxygen partial pressures followed by a rapid quench to room temperature. Studies of the magnetotransport behavior indicate that the anisotropy increases with increasing δ. This is reflected in the increased field broadening of the $\rho(T)$ curves as δ increases which is a result of the vortex-glass transition-phase boundary moving to lower T and H as δ is increased. From the scaling of the $E(J)$ behavior it was possible to identify a vortex-glass transition over the full range of δ studied. The values of the critical exponents ν and z determined were observed to be independent of δ and are the same as those determined in previous studies on fully oxygenated YBCO thin films.[12,15] This observation of universal scaling behavior strongly supports the idea that $H_g(T)$ is a continuous phase transition. Using zero-field $\rho(T)$ measurements, the crossover from two- to three-dimensional fluctuations predicted by the LD model, which occurs at the temperature T_{23} as T_c is approached from above, was also studied as a function of δ. The quantity $(T_{23}-T_c)$ was found to decrease with increasing δ, which is also consistent with YBCO becoming more anisotropic as δ is increased. Finally, ξ_{c0} and ξ_{c0}/d were observed to decrease by a factor of 2 as δ is adjusted from 0.05 (on the 90 K plateau) to 0.37 (on the 60 K plateau).

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