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**SUPERCURRENT DENSITY and VORTEX PINNING
in $Y_1Ba_2Cu_3O_{7-\delta}$ MATERIALS, versus OXYGEN CONTENT.**

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ABSTRACT

The superconducting current density and transition temperature T_c vary systematically with the oxygen content in many high- T_c superconductors. In the $Y_1Ba_2Cu_3O_{7-\delta}$ system, decreasing the oxygen content ($7-\delta$) generally reduces the persistent current density. These changes are directly related to changes in the fundamental superconducting parameters. Results of both magnetization and transport studies are discussed.

Introduction

All high temperature superconductors known to date are layered cupric oxides. Generally they allow some variation of the oxygen content and the associated density of charge carriers. For both fundamental insight and potential technological applications, it is important to understand how changes in such a readily varied parameter affect the superconductive properties of the materials. Consequently, this report discusses some recent studies on the impact of varying the oxygen content in the "123" compound $Y_1Ba_2Cu_3O_{7-\delta}$, where $x = (7-\delta)$ is the oxygen content. Results from both magnetization studies on aligned bulk materials and transport investigations on epitaxial thin films are included.¹⁻⁵ For the most part, we consider the compositional range $6.8 \leq x \leq 7.0$, which is the region in which the T_c and current density remain high.

Experimental Features

Most salient details of the materials synthesis and modification have been presented.^{1,6} For the bulk samples used for the magnetization studies, previously prepared $Y_1Ba_2Cu_3O_{7-\delta}$ powder was aligned in a 5 T magnetic field, then sintered to produce a binder-free, porous (76 % dense) array of aligned crystallites. The oxygen content was established in a thermogravimetric apparatus (TGA) under controlled temperature and oxygen partial pressure, and was determined from the mass loss relative

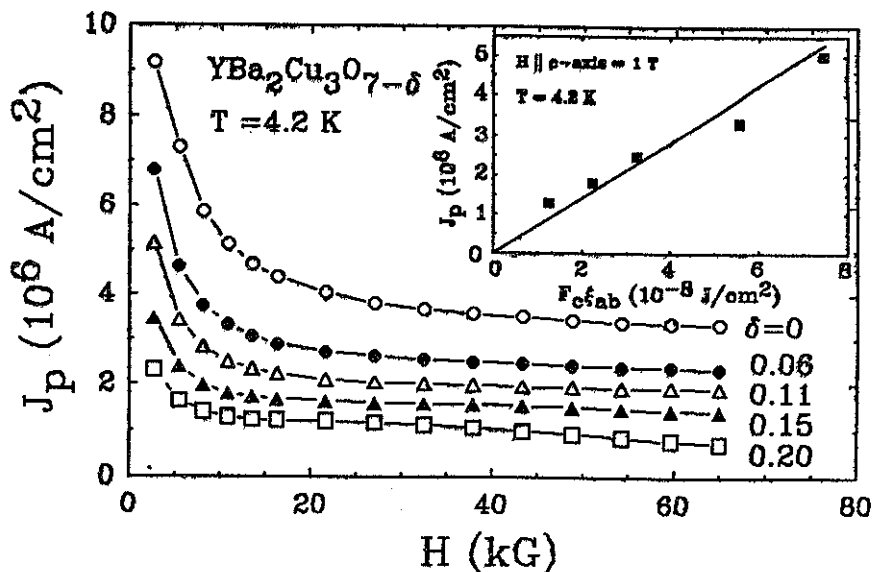


Fig. 1. The persistent current density J_p vs magnetic field H , for $Y_1Ba_2Cu_3O_{7-\delta}$ materials with oxygen deficiency δ . Inset: Correlation of current density J_p with the pinning parameter $F_c^* J_{ab}$; see text.

to full oxygenation. The transport studies used epitaxial films with the Cu-O planes and current flow parallel to the substrate. The oxygen content in the films was established similarly, except that the mass change was too small for direct measurement. As shown below, changes in the electrical conductivity provide a useful and sensitive method for monitoring the changes in the oxygen content of epitaxial films.

The magnetization experiments were conducted using a laboratory constructed vibrating sample magnetometer (VSM) and a commercial SQUID-based unit (Quantum Design MPMS). The magnetization M and hysteresis ΔM were measured as a function of temperature T and applied magnetic field H .⁷ The persistent current density $J_p = 15\Delta M/R$ was calculated from the Bean model,⁸ where R is the transverse grain radius, here $\approx 7 \mu\text{m}$. Further studies of the decay with time of M and supercurrent density J gave insight into the effective pinning energy for vortices, which is intimately related to the realizable current density. The transport studies were conducted using standard techniques with automated data acquisition for current reversal, etc. A standard electric field criterion of $E = 1 \mu\text{V}/\text{cm}$ was used to define the transport J_c . Since the relation between J and E is somewhat rounded and not a step function, one must be aware that the deduced values of current density from different experimental methods can differ greatly, especially at high temperatures in large magnetic fields.⁹ For example, the effective electric field in magnetization experiments is typically orders of magnitude smaller than those in transport studies, so the deduced values of current density are generally smaller, too.

Experimental Results

For internal consistency, magnetization studies were conducted on a single sample of $Y_1Ba_2Cu_3O_{7-\delta}$ with various oxygen contents. Analysis of the equilibrium magnetization (above the irreversibility line), using the theory of Hao-Clem,¹⁰ provided useful information on the superconductive condensation energy $F_c = H_c^2/8\pi$, the ab-plane coherence length ξ_{ab} , and the London penetration depth λ_{ab} corresponding to screening by supercurrents in the ab-plane. These analyses have shown that the F_c decreases sharply with oxygen deficiency $\delta \leq 0.2$, even though the T_c remains fairly high.^{1,11} Concurrently, ξ and λ increase, but the superconducting mass anisotropy $\gamma = 1/\epsilon = (m_c/m_{ab})^{1/2}$ remains near 5 in this compositional range.¹

Below the irreversibility line, magnetization studies have shown a substantial dependence of the persistent current density J_p on oxygen deficiency δ . In particular, J_p systematically *decreased* as δ increased. This effect is illustrated in Fig. 1, a plot of J_p versus magnetic field H at low temperature, for various oxygen compositions. With isolated exceptions, this systematic decrease of J_p extends to higher temperatures, even where secondary maxima ("fishtails") appear. The field at which the peak appears decreases approximately exponentially with δ , as described previously.¹

Let us consider the persistent current density at low temperatures, where the effects of thermally activated flux creep are minimized and the fundamental parameters, e.g., F_c , are temperature independent. A simple single-site pinning model¹ predicts that the critical current density with $H \parallel c$ -axis should scale as $F_c(\delta)\xi_{ab}(\delta)$. A similar dependence arises naturally in the theory of collective pinning by random point-like disorder.¹² In fact, this result is rather general, as the depairing current density $J_0 \propto H_c/\lambda_{ab} \propto H_c^2\xi_{ab}$ scales similarly. Figure 1 (inset) shows an experimental test of this dependence by plotting the measured current density J_p versus the quantity $F_c\xi_{ab}$, for $Y_1Ba_2Cu_3O_{7-\delta}$ with different oxygen contents. The plot is linear within experimental uncertainties, confirming the predicted dependence. This result means that most of the vortex pinning in the YBaCuO specimen came from a fixed number of preexisting, strong pinning sites, such as site-antisite disorder, vacancies, interstitials, etc. Removal of oxygen decreased the effectiveness of the pinning sites by changes in the fundamental parameters, largely reductions in the condensation energy F_c .

Varying the oxygen content has interesting effects on the irreversibility line as well. We determined the irreversibility field $B^*(T)$ as a function of temperature for each composition, using ac methods. The onset of nonlinear response, associated with switching the magnetization between branches of the hysteresis loop, defined B^* . The results have been shown in Fig. 6 of reference 1. As with the persistent current density, the irreversibility line systematically decreases with oxygen deficiency. In fact, $B^*(T)$ scales quite well with the same parameter $F_c\xi_{ab}$ (as seen in Fig. 13 of ref. 1), which collapses all the data onto a single function of reduced temperature T/T_c . This clearly implies that the measured quantity in these experiments is a depinning line. In more oxygen deficient materials, the temperature dependence crosses over from the power law (observed for $\delta \leq 0.2$) to an exponential variation farther below T_c .^{13,14}

Complementary studies of thin films using transport methods are consistent with

most of these findings. For example, the transport J_c of epitaxial, *c*-oriented films systematically decreases upon oxygen removal,⁴ as shown in Fig. 2 for two films grown under reduced oxygen pressure. This figure shows the resistive midpoint T_c and J_c (at three temperatures), versus a measure of oxygen deficiency, $-\Delta\sigma/\sigma_0$. This is the change in electrical conductivity $\Delta\sigma = [\sigma(\delta=0) - \sigma(\delta)]$, relative to its value at full oxygenation $\sigma(\delta=0) \equiv \sigma_0$. This fractional change provides a convenient experimental measure of compositional change in epitaxial films, since mass changes are too small to measure. Even though the films were processed under controlled temperature and oxygen partial pressure to give specific oxygen compositions, one should confirm that the observed decreases in J_c are indeed correlated with oxygen deficiency, i.e., reductions in carrier density. This correlation is demonstrated in Fig. 3, which plots the fractional change in carrier density, as determined from Hall Effect measurements,⁵ versus fractional change in conductivity. The excellent linear proportionality, with slope of unity, shows that the various measures are indeed internally consistent, giving $\delta \approx (1/2)[- \Delta\sigma/\sigma_0]$.

An interesting feature of Fig. 2 is the fact that the maximum J_c does not coincide with the maximum T_c . In general, the best J_c in $Y_1Ba_2Cu_3O_{7-\delta}$ is obtained with full oxygenation. On the other hand, these specific films have the highest T_c when they are slightly substoichiometric in oxygen; thus they are overdoped at full oxygenation. Similar but more pronounced differences in $J_c(\delta)$ versus $T_c(\delta)$ are being found in other high- T_c systems.^{15,16} This means

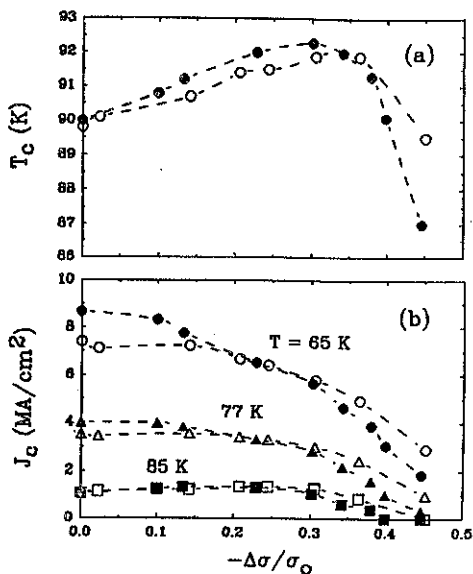


Fig. 2. For two YBaCuO films, the (a) T_c and (b) the transport current density, versus the normalized shift in normal state electrical conductivity $-\Delta\sigma/\sigma_0$. Oxygen content decreases from left to right.

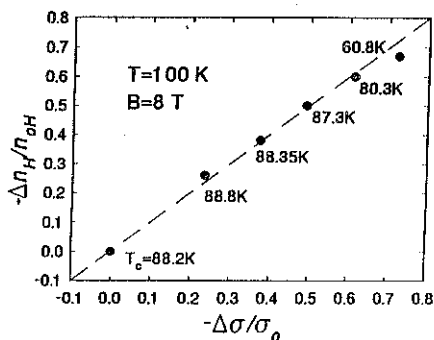


Fig. 3. Correlation of normalized conductivity change $-\Delta\sigma/\sigma_0$ with normalized change in apparent carrier concentration, from Hall Effect measurements.

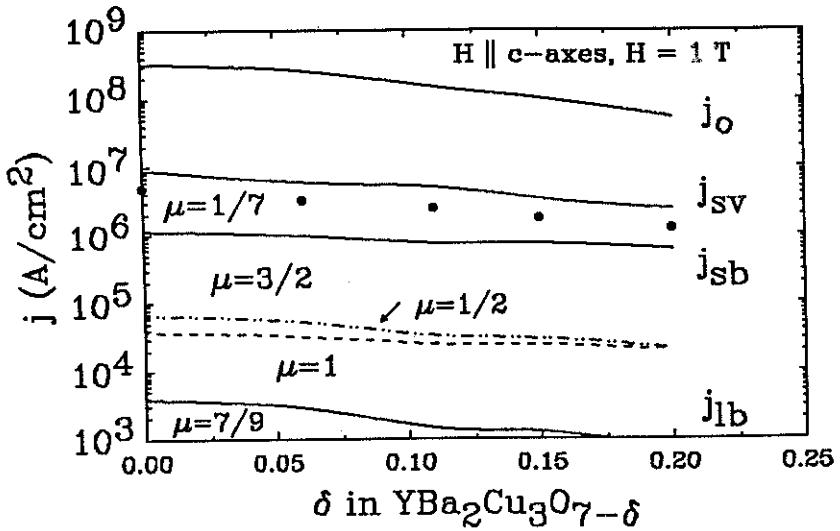


Fig. 4. Current densities vs oxygen deficiency δ for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. Solid lines show calculated values (at $T \ll T_c$) for the departing current density j_0 and crossover current densities j_{sv} , j_{sb} and j_{lb} for predominant pinning of single vortices, small, and large bundles, respectively. The ledge between $j_{sb}\epsilon a_o/\lambda_{ab}$ (---) and $j_{sb}e^2$ (--) has $\mu = 1/2$; here $a_o \approx (\phi_o/H)^{1/2}$ is the intervortex distance. Symbols show experimental values at 4.2 K (●).

that in optimizing materials, one must examine much more than the easily measured transition temperature.

To quantify the effective pinning energy for vortices, we investigated the time decay of supercurrents using dc magnetic methods. The same oxygen-deficient aligned materials were studied. The results of a "Maley-type" analysis¹⁷ support a relation of the form $U_{eff}(j, T) \approx U_o[(j_{co}/j)^\mu - 1]$, as given by vortex glass¹⁸ and collective pinning theories.^{12,19} The current density scale j_{co} decreased significantly as oxygen was removed, while the energy scale $U_o \approx 80$ K and characteristic exponent $\mu \approx 0.9$ varied slightly, but systematically.^{2,3} Our earlier work included a comparison of the measured current density with the predictions of collective creep theory in the *isotropic* case.¹¹ Since then, the theory has been generalized and extended to include the effects of anisotropy.¹⁹ It is therefore important to update the previous analysis and determine whether the inclusion of anisotropy significantly affects the previous conclusions (which remain valid). The results of this comparison are shown in Fig. 4, a plot of experimental and calculated crossover current densities versus δ . This analysis shows that the measured current density at low temperatures lies near the crossover j_{sb} at which collective pinning of small vortex bundles becomes predominate. Also, the experimental value of the characteristic exponent $\mu \approx 0.9$ lies intermediate to the values of $1/7$ (for single vortex pinning) and $3/2$ (for small bundle pinning). This finding is qualitatively consistent with the proximity of the experimental j to j_{sb} and is similar to that of Griessen

et al.²⁰ who concluded that vortices are singly pinned in their thin $Y_1Ba_2Cu_3O_{7-\delta}$ films. Complementary studies to identify various regimes of pinning in a YBaCuO crystal will be presented elsewhere.²¹

Conclusions

Studies of oxygen deficient $Y_1Ba_2Cu_3O_{7-\delta}$ materials have shown that the persistent and transport current densities, as well as the irreversibility line, systematically decrease upon oxygen removal. Many features can be understood in terms of measured changes in the fundamental superconducting parameters.

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