Magnetic field dependence of the critical current anisotropy in normal metal-$YBa_2Cu_3O_{7-\delta}$ thin-film bilayers

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We have measured the transport critical current density (J_c) in epitaxial quality films of $YBa_2Cu_3O_{7-\delta}$ some of which were covered by thin (10 nm) Ag films. The films, both with and without Ag, had J_c values greater than 10^6 A/cm^2 in liquid nitrogen. The effect of the Ag was to greatly reduce the dependence of J_c on external magnetic fields in the case where the field was oriented in the plane of the film, that is, perpendicular to the c axis. It is unlikely that the effect is simply due to altered surface pinning, although qualitative agreement with critical state models is observed.

The critical current density (J_c) of high-temperature superconductors is a crucial parameter which determines the range of applications of the new materials. Nearly epitaxial thin films of $YBa_2Cu_3O_{7-\delta}$ (YBCO) have been prepared by a number of groups using a variety of techniques.[1] These films had transport J_c in excess of 10^7 A/cm^2 at 4.2 K, and greater than 10^6 A/cm^2 at 77 K. These values are more than adequate for superconducting electronics, and are high enough to be interesting for small scale, high current applications. In the latter case, the behavior of the materials in external magnetic fields (H) must also be known. There is strong dependence of J_c on the angle made by the external field with the c axis of oriented films.[2] This letter addresses the question of the magnetic field dependence anisotropy of the critical current density in a unique composite structure of patterned YBCO films with normal-metal overlayers.

We prepared thin films of YBCO *in situ* by pulsed laser ablation (PLA), patterned the films using conventional microlithographic techniques, and measured their transport J_c in applied magnetic fields up to 10 T. Some of the films had thin coatings of Ag evaporated after patterning. The effect of the Ag was startling. The J_c value in zero field was typically unchanged. At 4 K the $J_c(H)$ characteristic was also unaffected. At 76 K, J_c decreased with increasing field. However, the coated films showed a much smaller decrease than did the uncoated films when the field was oriented perpendicular to the c axis of the crystalline YBCO, i.e., in the plane of the film. The critical current densities of the coated films were 50% larger than the J_c of the uncoated samples. When field was applied parallel to the c axis (perpendicular to the film surface), $J_c(H)$ was relatively unaffected by the Ag. We assume that this behavior was due to alteration of the field-angle dependent pinning forces in an anisotropic type II superconductor.

The directions of current flow, field, and crystalline axes are defined in the inset to Fig. 1. Roas *et al.*[2] reported on a large anisotropy in $J_c(H)$ between the cases where the external field is oriented parallel ($\theta = 0°$) or perpendicular ($\theta = 90°$) to the film's c axis. They observed that the angle dependence of J_c varied with temperature; at high-temperature J_c was dissimilar for the 90° and 270° case. This was interpreted as an indication that pinning at the top and bottom surfaces of their film was different. We designed our study of the effect of thin Ag films on $J_c(H)$ in an effort to determine if surface pinning is involved in anisotropies of J_c. YBCO films with Ag overlayers have very different surface characteristics than uncoated films; a Ag layer as thin as 10 nm converts an insulating surface into a metallic one.[3] As discussed by Campbell and Evetts,[4] such a conversion should modify surface pinning barriers. Normal metal overlayers also serve to provide good contacts, reduce the resistance of the composite conductor, and possibly passivate the YBCO surface to environmental damage. The technological advantages of normal-metal/YBCO bilayers and experiments on them are discussed in another paper.[5]

The critical current density of type II superconductors is defined operationally in terms of a measurement criterion such as resistivity or electric field. The microscopic origin of the measured voltage is extremely complex, typically involving the motion of vortices of magnetic flux.[6] High T_c materials often show weak link dominated behavior (a rapid and catastrophic decrease of critical current in even a very small external field, due to Josephson coupling between superconducting grains). The materials used in our experiments were highly oriented (nearly epitaxial) thin films of YBCO which do not show evidence of weak links. Therefore, the transport J_c of our patterned structures should be primarily due to flux creep and flux flow, and be strongly dependent on pinning. Because of the inherently layered nature of YBCO, it is not surprising that single crystal and epitaxial film results show strong anisotropy in the field-angle dependence of $J_c(H)$. The anisotropy may be due in part to "intrinsic" pinning in layered superconductors which applies to the case of interest: vortices lying in the a-b plane of our films. This case is examined in a recent paper by Feinberg and Villard.[7] They calculate a critical angle at which a vortex locks into a plane of the layered material; this angle was calculated to be less than 15° for YBCO under conditions similar to our 76 K experiments. We would expect to see effects due to this lock-in when the J_c is determined as a continuous function of angle.

We grew the YBCO films used in this study by PLA with a frequency-tripled Nd:YAG laser. The target con-

TABLE I. Thin-film sample characteristics.

Sample No.	L2-96B	L2-77.2(Ag)	L3-11A	L3-11A-Ag
Width	100 μm	100 μm	100 μm	100 μm
Thickness	280 nm	200 nm	300 nm	300 nm
Ag anneal	···	500 °C-60 min	···	500 °C-15 min
T_c(K)	90.5	89	91.5	91.2
$J_c(H=0)$ (76 K, 10^6 A/cm^2)	1.42	1.79	2.28	2.42

sisted of a high-density disk (15 mm diameter) of stoichiometric $YBa_2Cu_3O_x$. The 10 ns, 355 nm laser pulse was fired at 10 Hz and focused to a 5 mm^2 spot (for an energy density of 1–2 J/cm^2). The substrate material used for these experiments was single-crystal (100)$LaAlO_3$. The film growth conditions were nominally 700–750 °C substrate temperature, 27–33 Pa (200–250 mTorr) oxygen pressure, and a film growth rate of 1–2 nm/s. Final YBCO thicknesses ranged from 220 to 400 nm. As grown, these films had zero resistance temperatures (T_c) of 87–91 K. The films were patterned into 100-μm-wide strips with current and voltage taps ending in large pads. The processing used standard photolithography[8] and wet etching in a saturated solution of ethylenediaminetetraacetic acid (EDTA).[9] We have used this process to make undamaged structures as small as 3 μm wide; the strips used in these experiments were deliberately made as large as possible to eliminate the influence of patterning damage. After wet etching and photoresist removal (in acetone), Ag pads were deposited on the current and voltage pads through a metal stencil. The samples were then annealed in flowing O_2 to produce low contact resistances; the anneal temperature and time was nominally 500 °C for 15–30 min. The films were processed further for the thin Ag layers. A metal stencil was used to deposit Ag in the pattern shown schematically in Fig. 1(b). The samples were annealed again in flowing O_2 as listed in Table I.

The samples were mounted on a "flip table" which allowed the 0° and 90° measurements. Both soldered (In-2% Ag) and pressure contacts were used. Critical currents in excess of 10 A were easily measured with soldered contacts. Pressure contacts were adequate for currents less than 2 A. Four-terminal, dc current-voltage characteristics were acquired and J_c extracted using an electric field criterion of 1.0 μV/cm. The computer-automated data acquisition system is described in another paper.[10] Experiments were conducted with the samples immersed in liquid cryogen (helium or nitrogen) to facilitate cooling. The current was perpendicular to the applied field to within 4°. The precision of θ alignment was 4° for $\theta = 0$ ° and 1° for $\theta = 90°$. This amount of misalignment was determined to have little effect on J_c in repeat determinations under the conditions reported. These errors are also smaller than the "critical" angle calculated for YBCO in Ref. 7.

Relevant parameters of the J_c samples are summarized in Table I. The upper portion of Fig. 1 shows $J_c(H)$ for two 100 μm strips at 4 K in an external field up to 10 T, the perpendicular and parallel orientations of Fig. 1. The samples were made in different deposition runs and processed separately; L2-77.2 had a 10 nm coating of Ag. The results were qualitatively similar to other data reported. The perpendicular field J_c was nearly independent of field, supporting the idea of intrinsic flux pinning between CuO planes. These data are representative of more than six samples: the Ag has no consistent effect at 4 K. It is important to note that there is no evident degradation due to the Ag layer.

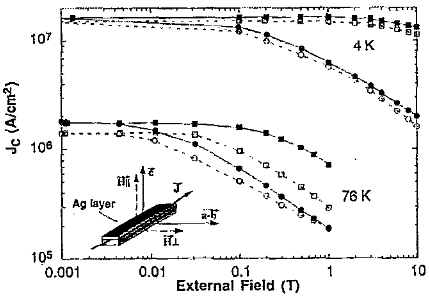

FIG. 1. Critical current density, vs external field for 3 and 76 K in parallel (circles) and perpendicular (squares) field, and for a coated (L2-77.2, solid lines) and uncoated (L2-96B, dashed lines) samples.

At 76 K (0–1 T), the field characteristics were markedly different, as seen in the lower part of Fig. 1. The effect of the Ag layer is evident from the difference between the solid lines (representing the sample with 10 nm of Ag) and the dashed lines (uncoated sample). In parallel field the two samples have different curvatures but are nearly identical at 1 T. There is a much greater difference for perpendicular field: at 1 T the coated sample had a 50% greater J_c [normalized to $J_c(H=0)$]. Again, this is typical of all of our samples to date. The effect also holds for a film that was tested and then coated. In Fig. 2, $J_c(H)$ is plotted for the same sample before and after Ag deposition and annealing. The curves for parallel field approach each other at 1 T while the normalized J_c at perpendicular field is 44% greater after the Ag treatment.

If surface pinning were to account for the difference in J_c at 1 T, changing the direction of the force on the vortices should change the measured J_c. Simply reversing the direction of current flow reverses the direction of the force on the vortex as diagrammed in the inset to Fig. 3. If there were a barrier to flux penetration which was larger at one interface, there should be an anisotropy between the field dependence of "forward" and "reverse" current density.

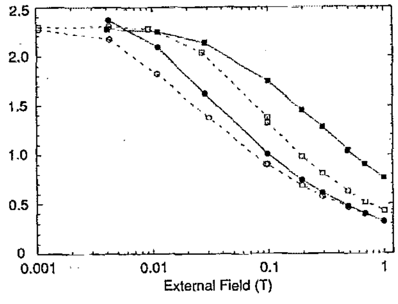

FIG. 2. J_c vs H for a sample L3-11A before and after silver coating. The solid lines and symbols correspond to the sample *with* Ag. Squares correspond to $H \perp c$, circles correspond to $H \parallel c$.

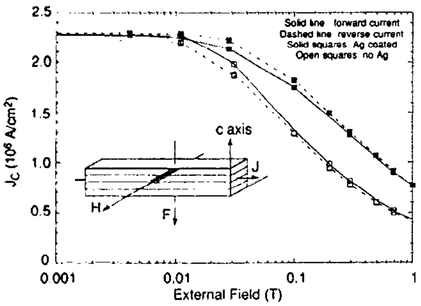

FIG. 3. J_c vs H for L3-11A before and after coating (open and solid squares) for forward and reverse current (solid and dashed lines). The inset shows the Loretnz force on a vortex for field pointing out of the page, current flowing from left to right.

We would thus expect that J_c of Ag-coated samples should depend on the direction of current flow differently than uncoated samples. In Fig. 3, we plot $J_c(H)$ in parallel field for forward and reverse current. Before the overlayer, J_c was slightly higher when the force was down: the vortices entered the top surface of the film and left at the substrate. This effect reversed after the Ag was deposited and annealed. But the difference is small, much smaller than the total difference between the coated and uncoated cases. Moreover, another sample, made at the same time as L3-11A and coated with Ag, showed the same dependence on current directing as did the uncoated L3-11A. The differences all diminished as the field approached 1 T.

An important factor in understanding the observed effect is the uniformity and distribution of the Ag in the annealed samples. Scanning electron microscope studies revealed that the Ag was not at all homogeneous on the surface of the YBCO. After annealing at 500 °C for 10 min, the surface of the Ag-YBCO bilayer appeared to have thin patches of Ag (or Ag oxide) scattered a few micrometers apart. There may be surface pinning changes due the patchiness of the Ag coverage; this can be distinct from the simple order parameter gradient effect of interfaces.

We now examine these various field-dependent data within the context of the standard flux motion picture. Critical state models[11] have been used to explain the $J_c(H)$ behavior of practical type II superconductors. In such formulations, the "critical" current of a superconductor is the current at which enough flux is depinned (by the Lorentz force due to the current) to allow a measurable voltage to appear. Most models predict the relationship $J_c \propto (1/H)^n$, without the explicit temperature dependence needed to explain the difference between our 4 and 76 K data. Some calculations of temperature-dependent behavior have been carried out using flux creep models, the simplest of which is the Anderson–Kim model.[12] The Anderson–Kim model of flux creep predicts a functional form $J_c \propto 1/H$, for $H > H_{C1}$, the critical field for flux entry into a type II superconductor. Our $J_c(H)$ data can be described[13] by the functional form $J_0/(1 + H/H_0)^n$ where n typically lies between 0.5 and 1.0.

The experimental evidence is clear. The Ag treatment reduced the effect of external field on J_c, without decreasing the already very high J_c values. The reduction of contact resistance and increased thermal stability commensurate with a normal-metal layer can be achieved without deleterious effect.[5] While interesting from a technological standpoint, this also opens up an area of investigation in the physics of highly oriented high T_c superconductor films.

We are clearly probing fundamental differences in pinning forces in these layered compounds; the dramatic effect in perpendicular field becomes only a small and inconsistent one in parallel field. The inability of the simplest phenomenological models to explain the experimental observations is not surprising. The J_c increase is not expected to be a simple surface pinning effect because of the observed lack of current reversal anisotropy. The simplest Anderson–Kim flux creep model is not adequate to describe the behavior of either coated or uncoated films, as seen in the functional fit of $J_c(H)$. However, the general form $J_c \propto 1/(1 + H/H_0)^n$ is seen and probably an indication that the physics of flux creep in the bilayers we studied is similar to that observed in conventional low T_c type II materials. Added pinning sites may have resulted from Ag doping in the bulk of the film. This explains neither the nearly identical response in the parallel field case nor the lack of an effect at 4 K. The Ag thus appears to strengthen existing anisotropic pinning forces rather than create new ones.

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