

Au / YBa₂Cu₃O_{7-δ} Thin Film Composites on Various Substrates

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Abstract—We have grown a series of composite films of Au with YBa₂Cu₃O_{7-δ} on various substrates using a bilayer deposition technique to study their suitability for flux flow applications. The T_c 's and J_c 's were measured, and results compared with plain YBCO films for the different substrates. The patterning process resulted in some damage to the composites, but an oxygen plasma treatment was effective in restoring T_c 's and resistance ratios. J_c measurements suggest that Au/YBCO composites behave like SNS-coupled arrays of superconducting grains. Their increased response to small magnetic fields is consistent with Josephson vortices dominating the transport. These findings indicate that Au/YBCO composites are a promising material for flux flow applications.

I. INTRODUCTION

There is considerable interest in developing superconducting flux flow and fluxonic devices [1] - [3]. These devices are based on the motion of either Abrikosov or Josephson vortices and require a material whose properties do not impede the flow of magnetic flux. Our approach to this problem has been to develop a material consisting of a random array of Josephson junctions in which Josephson vortices can move easily throughout the film [4].

We have been studying composite films of YBCO with the noble metals, Au and Ag, and our results suggest that the material behaves as an array of Josephson junctions [5], [6]. The amount of noble metal in the composite can be varied to affect the superconducting properties, but the choice of substrate is also important. Previously, we have shown that substrates with a greater mismatch to the YBCO crystal structure produce films with the greatest response to low magnetic fields [7]. However, these films are also adversely affected by patterning procedures.

In this paper, we report the properties for Au/YBCO films grown on yttria-stabilized zirconia (YSZ), a substrate with a mismatch to YBCO between that of the substrates we have previously studied [7]. In addition, we repeat our studies of strontium titanate (STO) and magnesium oxide (MgO) substrates, because we have made significant improvements in our sputter technique and found a procedure to restore films damaged by patterning.

II. FILM PREPARATION

Substrates of (100)-oriented STO, YSZ, and MgO were cleaned with a series of heated baths of methanol and isopropanol solvents. The MgO substrates, which were coated in mineral oil to prevent hygroscopic damage, required additional pre-scrubbing in trichloroethane (TCA) to remove the oil. The cleaned substrates were then annealed in a flow-

ing oxygen atmosphere at 1000 °C for 12 or more hours. Three substrates (one of each kind) were then silver-pasted to a substrate holder for the deposition.

The Au/YBCO composite films were grown by our bilayer deposition technique which we have modified to be an entirely *in situ* procedure. Briefly, a Au layer was deposited first at ambient temperature. During heating of the substrate for the YBCO layer, the Au film coalesced into "puddles" on the substrate surface, and the YBCO filled in between. The Au layer was grown in a conventional sputter chamber in a 10 mT argon atmosphere. The substrate holder was then transferred under vacuum to our Inverted Cylindrical Magnetron sputtering system where the YBCO layer was grown at 800 °C (measured by a thermocouple inserted into a well in the holder) and in an oxygen/argon atmosphere, 237 and 163 mT, respectively. For these films, our sputter gun was modified to apply a positive bias to the anode, resulting in a more efficient collection of the negative ions that are destructive to optimum YBCO film growth [8]. This change has significantly improved the appearance of our YBCO films on STO and resulted in better reproducibility of their superconducting properties.

A series of eight composite films was grown for each substrate choice. The Au layer thickness was varied between 0 and 1000 Å. The YBCO thickness was 1000 Å for all films, a practical choice for later device fabrication. This is also thin enough to avoid the roughness that occurs for our YBCO films on STO thicker than 1300 Å, yet thick enough to have a complete T_c (transition temperature) of 86.5 K and RR (resistance ratio) of 3.0.

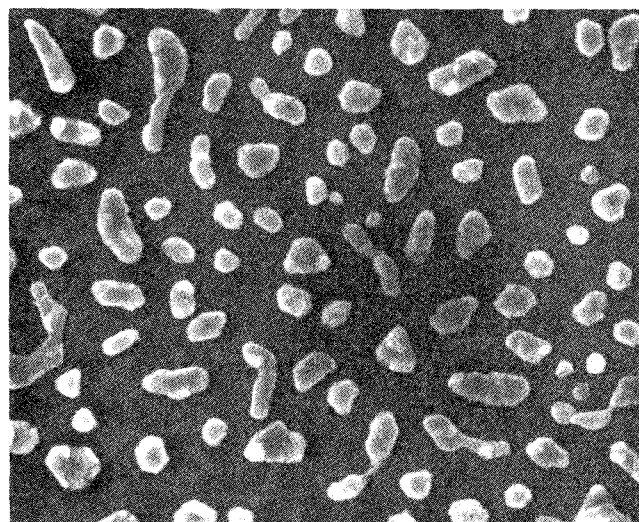


Fig. 1. SEM micrograph of the surface of a Au/YBCO composite with 300Å of Au grown on YSZ. The light regions are the Au "puddles", and the dark background is YBCO.

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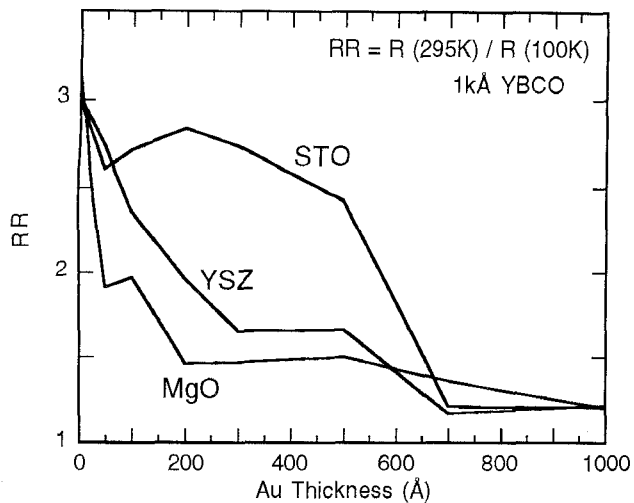


Fig. 2. The resistance ratio as a function of Au thickness. RR falls off fastest for composites grown on MgO, then YSZ, and slowest for STO.

III. MORPHOLOGY

Earlier studies of Au/YBCO composite films showed no evidence for alloying of the Au and YBCO materials for films with Au layers up to about 75% of the YBCO layers' thickness [5]. The Au regions were found to extend from the substrate interface to above the films' surface, looking like plateaus rising above the YBCO background. For thicker Au layers, the distinct regions were not observed.

These composites are similar. SEM micrographs of plain YBCO films on STO, YSZ, and MgO showed c-axis oriented grains on the surface and some scattered voids. STO and MgO films had fewer voids than the YSZ. At 300 Å of Au, the composite films had rounded, bright regions of Au (as identified by Energy Dispersive Spectroscopy) scattered about the darker YBCO background. Higher magnifications showed that the YBCO grains were ordered in-plane, also. These features are seen in Fig. 1, a micrograph of a 300 Å Au composite on YSZ. For films with thicker Au layers, the Au puddles were strung together, in a manner that differed with the substrate. The connected Au puddles on STO and MgO meandered randomly, while on the YSZ they appeared more directional, as we had seen earlier with Ag and YBCO composites on MgO [6]. For the thickest Au layers, between 700 and 1000 Å, the film surfaces had a more homogeneous appearance, and segregated areas of Au and YBCO were no longer seen. In-plane ordering was also not evident.

Since the maximum superconducting coherence length at low temperatures for YBCO is about 10 Å, the micron-size Au clusters seen with the SEM cannot be responsible for the interesting transport properties of our composite films. The grains themselves do not seem to be the source either, as x-ray diffraction and SEM studies indicate that the YBCO grains were as well-oxygenated and ordered as for our plain YBCO films. The YBCO grain boundaries are an obvious candidate, but the resolution of a TEM is needed to study these features. To address these questions, we are pursuing a collaboration to study the grain boundaries with a STEM [9].

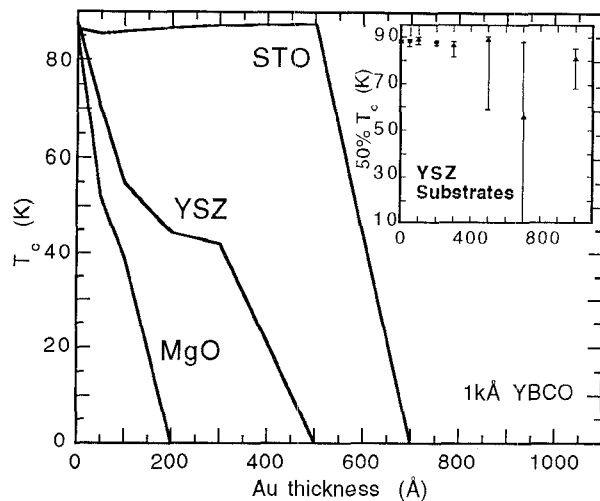


Fig. 3. T_c (the temperature where R goes to zero) as a function of Au thickness. Inset shows mid-point T_c with error bars indicating 10% and 90% points of transition.

IV. TRANSPORT MEASUREMENTS

Ag contact pads were evaporated onto the samples to study the resistive transport properties of the Au/YBCO composites. We cooled the samples in a closed-cycle helium refrigerator and measured the temperature dependence of the resistance to obtain the T_c and the resistance ratio (RR) between room temperature and 100 K. We use T_c here to mean the zero resistance temperature. We also refer to the transition width (ΔT_c) between the 90% and 10% values of the resistance above the transition's onset, and the mid-point of the transition ($50\%-T_c$) which is the temperature at 50% of the onset resistance. Some films were patterned into a bar geometry using standard photolithography and ion milling methods for critical current density measurements (J_c) in ambient and in small applied magnetic fields. We determined the critical current (I_c) as the lowest current producing a non-zero voltage, which corresponds to an electric field criteria of 2×10^{-6} V/cm. To calculate J_c , we divided by the width of our bar and the thickness of the superconducting layer. For some of the films, a 30 minute plasma etch in oxygen was beneficial to restore the samples' superconductivity from damage incurred during the patterning process.

A. Au Thickness Dependence

The resistance ratios of the as-grown composites were measured, and the results are shown in Fig. 2 and listed for some of the samples in Table I. For all the substrates, as the Au thickness was increased the RR fell off, but not at the same rate. For STO, the RR did not change much until the Au layer was 500 Å thick. For YSZ, 100 Å of Au had significantly lowered RR, and for MgO, 50 Å of Au had. Figure 3 is a plot of T_c for the Au composites of all the substrates. For the different substrates, T_c followed the same trends with Au thickness that the RR did. This result is also consistent with increased grain boundary disorder, because

TABLE I
EFFECTS OF PROCESSING ON Au/YBCO COMPOSITES

Au thickness	RR	As-Deposited T_c (K)	ΔT_c (K)	RR	After Patterning T_c (K)	ΔT_c (K)	RR	After Plasma Etch T_c (K)	ΔT_c (K)
STO Substrates									
0 Å	3.0	86.4	0.8	2.7	87.3	0.8			
50 Å	2.6	85.5	0.7	1.9	76.0	4.8			
100 Å	2.7	86.0	0.5	2.5	87.0	1.0	2.7	85.0	2.0
YSZ Substrates									
0 Å	3.0	87.5	0.8	2.8	86.8	0.8			
50 Å	2.7	70.0	2.9	1.4	0.0	>80			
100 Å	2.4	55	3.4	1.8	0.0	15.0	2.0	25	6.9
MgO Substrates									
0 Å	3.1	85.0	0.9	2.8	86.8	1.2			
50 Å	1.9	52	13.4	1.8	30	15.5			
100 Å	2.0	39	14	1.4	0	18.3	2.1	43	12.9

the superconducting length scale is on the order of the lattice parameter for YBCO, and the scattering at grain boundaries affects the superconducting charge carriers. The inset shows the 50%- T_c for the composites on YSZ with the error bars indicating the 10% and 90% points. The T_c 's for MgO followed a similar trend except that the broadening in the transition width and the drop in 50%- T_c began at smaller Au thicknesses. With the STO composites, these effects occurred at larger Au thicknesses.

B. Patterning Effects & Plasma Etching

We patterned some of the samples into bars 1 mm long and 50 μ m wide for the "plain" YBCO films and 150 μ m wide for the composites. Standard photolithography and ion milling techniques were used. T_c 's were re-measured, and the results are shown as the "after patterning" values in Table I. Comparing these numbers with the "as-deposited" figures, shows that the "plain" films on all the substrates were not significantly affected by the processing. The results were substrate dependent for the 50 and 100 Å Au composites, however, with large changes occurring for the MgO and YSZ samples, and insignificant changes for the STO composites.

All of the patterned 100 Å Au composites were treated with a 30-minute oxygen plasma-etch. Interestingly, the properties of all the films improved, with the STO and MgO composite being restored to "as-grown" values of T_c and RR, while the YSZ was partially restored.

C. Critical Current Measurements

Critical current measurements were made on the patterned films, and the results summarized in Table II. The asterisks indicate that the measurement is of a plasma-treated sample. Measurements were made both in ambient field and in perpendicular applied magnetic fields of 100 Gauss or smaller. A solenoid magnet positioned in the refrigerator between the cold stage and the refrigerator's side wall was used to apply the field.

A power law fit to the critical current near T_c can give information about the coupling mechanism between the superconducting grains in the sample. Data from the STO samples were fit to the equation

$$J_c \propto (1 - t)^n$$

where t is the reduced temperature, T/T_c . If the coupling is superconducting/insulating/superconducting (SIS), n is 1 [10]. If the coupling is superconducting/normal-metal/superconducting (SNS), n is 2 [11]. The Ginzburg-Landau theory's prediction for thin films is 1.5, and this is the result found for most YBCO thin films [5]. Figure 4 is a log-log plot of J_c vs. $(1 - t)$ for the "plain" YBCO and the 100 Å Au composite films on STO. A power law dependence will be a straight line whose slope is the value of the exponent. For our "plain" YBCO film, n was close to 1.5, in agreement with the other studies. As Table II shows, n for the "plain" film on MgO was also close to 1.5. The temperature dependence of J_c for the 100 Å Au composite film (shown in Fig. 4) and the 50 Å composite film (Table II) had n 's near 2. This indicates that the behavior of composite films differed from "plain" films and was more like an array of SNS junctions.

To test the response to low magnetic fields of our composite films, we measured the critical current density in ambient and applied field. The "plain" films were also measured to compare with the composites. Typically, the measurements were made at 0.95 T_c and for a 100 Gauss applied field. However, if 100 Gauss quenched the superconductivity of the sample, smaller fields and different temperatures were used. We calculated the percent differ-

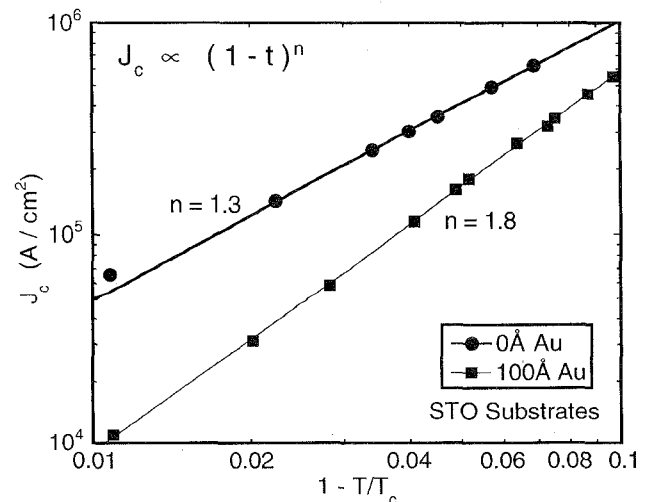


Fig. 4. Temperature dependence of J_c for the plain YBCO film and 100 Å Au composite on STO. Data points are fit to a power law.

TABLE II
J_c MEASUREMENTS FOR Au/YBCO COMPOSITES

Au Thickness	T_c (K)	n	% ΔJ_c at (t, B)	
STO Substrates				
0 Å	87.2	1.3	-10%	(0.95, 100G)
50 Å	78.1	2.1	0%	(0.95, 100G)
100 Å	87.0	1.8	-10%	(0.95, 100G)
100 Å	85.0*		-8%*	(0.95, 100G)
YSZ Substrates				
0 Å	86.8		-25%	(0.95, 100G)
MgO Substrates				
0 Å	86.7	1.7	-9%	(0.95, 100G)
50 Å	30		-33%	(0.65, 2.2G)
100 Å	43*		-48%*	(0.50, 20G)

* indicates after plasma etch

ence of J_c in an applied field B from J_c in ambient field as

$$\% \Delta J_c(t, B) \equiv \frac{J_c(t, B) - J_c(t, 0)}{J_c(t, 0)},$$

and the results are given in Table II. Several observations can be made. One of the more interesting is the non-zero response of the "plain" films. These measurements were all made at 0.95 T_c and 100 Gauss. The STO and MgO films had a 10% difference, but the YSZ was significantly larger at 25%. The composites on STO responded at a similar level to the "plain" film. The MgO composites, however, were more sensitive than the "plain" film, being driven normal by the 100 Gauss field. Large %ΔJ_c's of 30 - 50 percent were found for the 50 and 100 Å Au composites, even at substantially lower fields and reduced temperatures. Figure 5 is a plot for the 50 Å Au composite on MgO showing I-V traces at t = 0.65 in ambient field and also at 2.2 Gauss. (The 2.2 Gauss trace has been shifted 0.2 μV up for clearer presentation.) The table inset in the figure shows the %ΔJ_c's at 2.2 Gauss measured for different temperatures. At lower temperatures, the response increased. This cannot be explained in terms of the unpinning of Abrikosov vortices but is compatible with a situation where Josephson vortices are dominating the transport.

V. CONCLUSIONS

We have used a bilayer deposition technique to grow thin film composites of Au with YBCO on STO, YSZ, and MgO substrates. A series of composites was grown for each substrate with the YBCO layer thickness constant at 1 kÅ and the Au layer thickness varied between 0 and 1 kÅ. As the Au layer thickness was increased, the onset and mid-point of the superconducting transition remained high, and a tail developed at low temperatures. Composites on MgO were affected the most, followed by those on YSZ, and then those on STO. Standard photolithographic procedures to pattern the films for J_c's resulted in degraded T_c's and RR's for the composites, but not the "plain" YBCO films. The amount of damage was greatest for the MgO composites, then YSZ, and then STO. An oxygen plasma etch restored the properties of some of the films to their "as-grown" values. Near T_c, the temperature dependence of the J_c's for our plain YBCO films had a power law dependence with an exponent of 1.5. The composites on STO fit an exponent of 2, as predicted for SNS

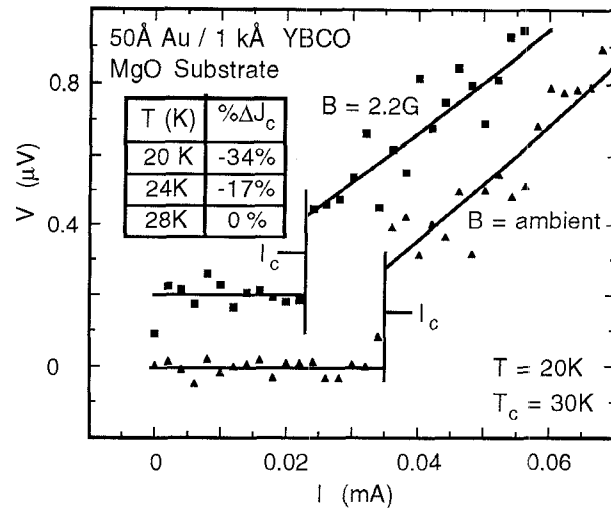


Fig. 5. I-V traces for 50 Å Au composite on MgO in ambient field and for a 2.2 Gauss applied field. For clarity, the 2.2 Gauss data are offset 0.2 μV and lines are drawn to guide the eye. The table inset shows the percent change in J_c for other temperatures.

coupling. We measured the effect of small magnetic fields on the films' I-V curves, and found that "plain" YBCO films were affected (the film on YSZ the most), and that the MgO composites had very large responses. The response of the 50 Å Au composite on MgO increased as temperature was lowered, an observation that can be explained by Josephson vortices dominating the transport. The behavior of these Au/YBCO composite films suggests that they are two-dimensional arrays of SNS junctions and are thus an interesting material system for flux flow device applications.

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