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Metalorganic chemical vapor deposition of double-sided $YBa_2Cu_3O_7$ thin films

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Thin films of $YBa_2Cu_3O_{7-\delta}$ were deposited on both sides of (100) $LaAlO_3$ substrates by metalorganic chemical vapor deposition. Using a gold-coated wafer carrier, improvements in both the transition temperature and critical current density of the film exposed to the carrier (first side) were demonstrated. T_c's of 86–88 K and J_c's $> 10^6$ A/cm^2 at 77 K were achieved. The microwave surface resistance of both sides of double-sided samples measured at 77 K was approximately 8 mΩ at 36 GHz, scaling to less than 700 $\mu\Omega$ at 10 GHz.

For continued improvements in low-loss high-temperature superconductor (HTS) microwave components, substrates coated on both sides with HTS films (to be referred to as "double-sided HTS films") are required to eliminate losses in the ground plane. In many physical deposition processes, a thermal adhesive such as silver paste provides thermal contact between the substrate and substrate heater, and thus maintains substrate temperature during film growth. The thermal adhesive and its removal make double-sided HTS film processing as well as circuit fabrication difficult. Takemoto *et al.*[1] have demonstrated double-sided $YBa_2Cu_3O_{7-\delta}$ film growth by metalorganic chemical vapor deposition (MOCVD) at atmospheric pressure without the use of a thermal adhesive, however their film properties were somewhat degraded. In our previous work, we demonstrated that low microwave surface resistance $YBa_2Cu_3O_{7-\delta}$ films can be processed at reduced pressure by MOCVD without using silver paste to thermally bond the substrate to the substrate heater.[2] In this work, we show a simplified process for fabricating high-quality double-sided HTS films utilizing MOCVD processing at growth pressures of 5 Torr.

The superconducting films were grown in a commercial-scale vertical stainless-steel MOCVD reactor (Emcore Corp.).[2] Thoroughly degreased $LaAlO_3$ (100) substrates, $15\times15\times0.5$ mm, were resistively heated on a high-speed rotating disk susceptor. No thermal adhesive was used; the substrates were simply placed in a recess in the susceptor. Yttrium, barium, and copper derivatives of 2,2,6,6-tetramethyl-3,5-heptanedionate (Strem Chemicals) were used as metalorganic sources for transport of the metals into the growth chamber. Source temperatures were 118, 215–230, and 123 °C for the Y, Ba, and Cu complexes. Argon flow rates through the Y, Ba, and Cu bubblers were 35, 135, and 35 sccm, respectively. Growth pressure was 4–5 Torr with 300 sccm of flowing nitrous oxide. At the conclusion of film growth, the chamber was backfilled with oxygen, and slowly cooled to room temperature.

Growth of double-sided films involved a modification of the susceptor. A thin layer of gold (0.6 μm) was evaporated onto the NiCrAlY alloy susceptor and served a two-fold purpose. First, it acted as a diffusion barrier, eliminating contamination from the susceptor onto the first-deposited high T_c film. Second, during first-side deposition, contamination from the susceptor onto the bare substrate was minimized, thus eliminating the need for extensive cleaning and etching of the backside before subsequent deposition. With the presence of the gold barrier layer on the susceptor, the deposition temperature was optimized at 750 °C. The resulting double-sided HTS films were 4000 Å thick on both sides.

Films grown in this study were slightly yttrium-rich with respect to barium, as analyzed by elastic backscattering. A recent study[3] indicated yttrium-rich MOCVD films contained finely dispersed Y_2O_3 precipitates which were self-limited in size and epitaxially oriented with respect to the *c*-axis oriented YBCO matrix. It is our experience that yttrium-rich films are readily grown with good electrical properties. The effect of these precipitates on microwave surface resistance at 77 K is considered negligible in relation to the many other defects present in high T_c thin films.

Superconducting transition temperatures (T_c) and critical current densities (J_c) were measured by an inductive method.[4] Transition temperatures for both sides of a double-sided sample grown on the susceptor without the gold barrier coating are shown in Fig. 1. The as-grown electrical properties of the first-side coatings measured before second-side deposition exhibited T_c's of 88 K and J_c's $> 10^6$ A/cm^2. The electrical properties of the first-side film measured after being exposed to the susceptor during second side growth were severely degraded. The transition temperature was both lowered and broadened and the critical current density dropped two orders of magnitude to 5×10^4 A/cm^2 at 77 K. These properties are typically observed in granular films. Such films are unacceptable for passive microwave device application.

Without the gold diffusion barrier, an apparent surface reaction took place with the previously deposited yttrium, barium, and copper oxides on the susceptor. The surface morphologies of these "contaminated" films were extremely rough with particulates larger than 1 μm on the surface. In addition, reproducibility from run to run was poor, depending upon where the substrate was placed on the susceptor. However, the second-side film was unaffected by the presence of a YBCO coating on the other side of the substrate, resulting in T_c's and J_c's exceeding 87 K and 10^6 A/cm^2, respectively.

Transition temperatures for double-sided coatings pro-

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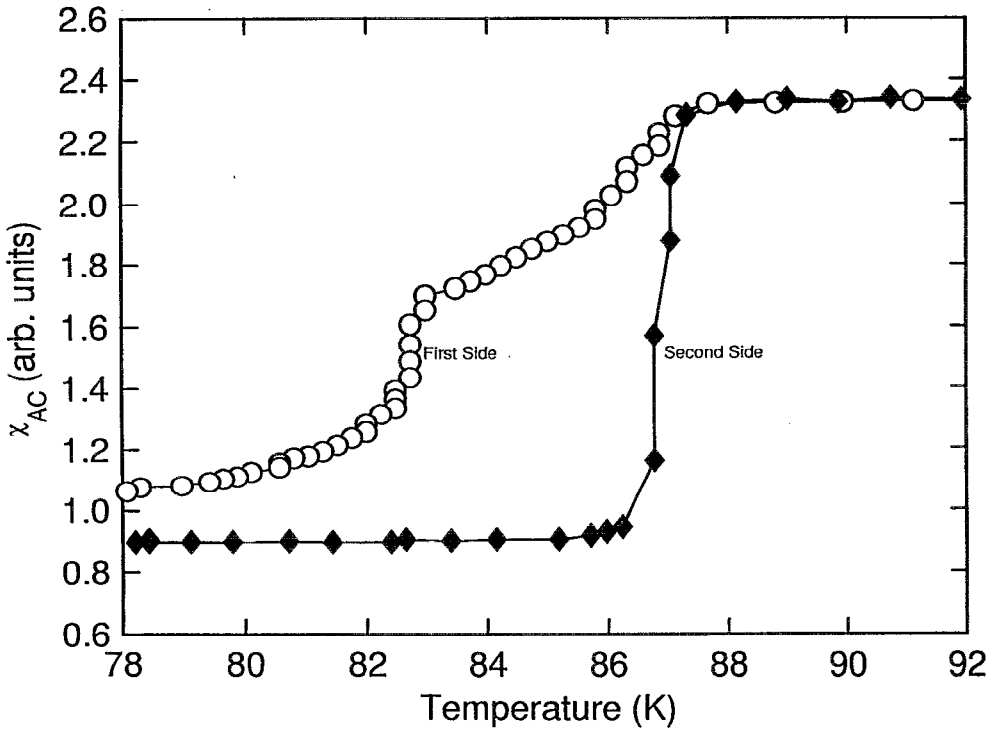

FIG. 1. Transition temperatures for both sides of a double-sided HTS substrate processed on a NiCrAlY alloy susceptor.

cessed using the gold-coated susceptor are shown in Fig. 2. The transition temperature and critical current density of the first-side coating remained high, i.e., $T_c > 87$ K and $J_c > 10^6$ A/cm^2 at 77 K, indicating the effectiveness of the gold as a diffusion barrier. In addition, examination of the surface morphologies of first-side films indicated no change, unlike films processed without the gold-coated wafer carrier. The transition temperature of the second-deposited film was measured to be 86 K, and was slightly depressed from normal values. The transition was sharp,

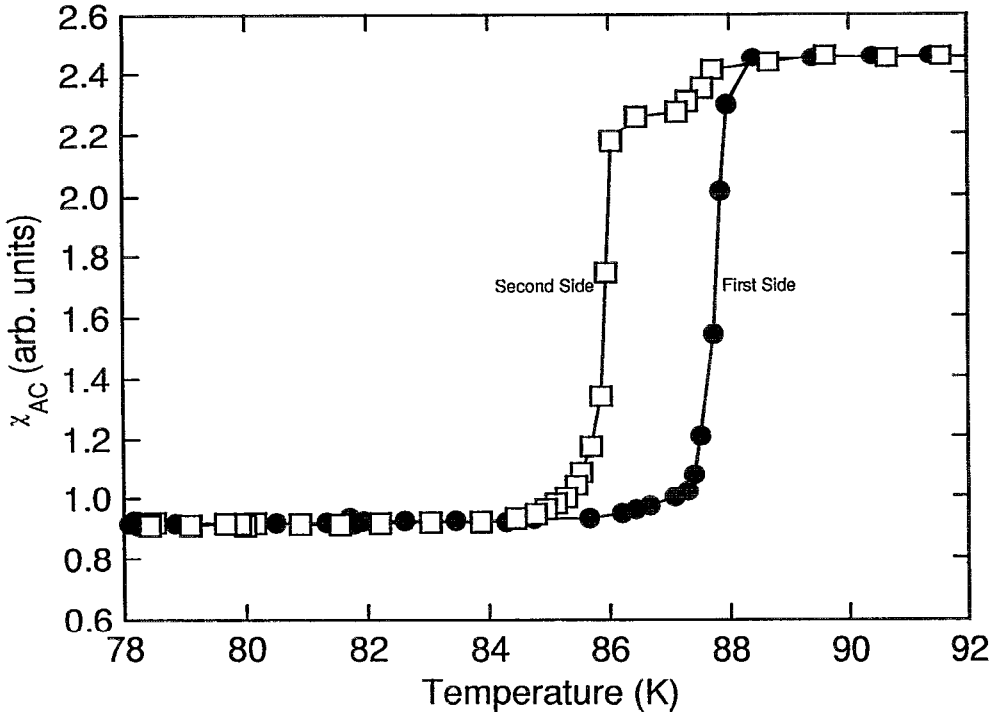

FIG. 2. Transition temperatures for both sides of a double-sided HTS substrate processed on a gold-coated NiCrAlY alloy susceptor.

however, and the critical current density measured at 77 K exceeded 10^6 A/cm^2. The apparent double-transition seen in the figure arose from the induction coil picking up a weak signal due to the transition of the backside film during measurement. The slight depression in T_c of the second-side film is reproducible and remains unresolved at this time.

The microwave surface resistances (R_s) of both sides of double-sided samples were measured in a copper cavity at 36.4 GHz using an endwall replacement technique,[5] having a sensitivity limit of about 1 mΩ. For double-sided films deposited at substrate temperatures of 750 °C, the measured R_s at 77 K for both sides was approximately 8 mΩ, indicative of high-quality material. In double-sided film growth, producing high-quality films on both sides of the substrate requires protecting the previously deposited (first-side) film exposed to the substrate carrier as well as maintaining proper deposition conditions for the second-side film. As the processing temperature is increased, the chances of first-side film degradation also increases. However, at lower substrate temperatures the rf loss of the film being deposited may also be degraded. At substrate temperatures less than 750 °C we observed a slight increase in R_s measured at 77 K, of the as-deposited film. At substrate temperatures greater than 750 °C we also observed increases in R_s measured at 77 K in films on both sides of the substrate.

In conclusion, we have demonstrated an improved two-step process for double-sided HTS film growth by conventional multisource MOCVD in which the films on both sides of the substrate were determined to have state-of-the-art electrical properties. The $YBa_2Cu_3O_{7-\delta}$ films were deposited on $LaAlO_3$ substrates using a gold coated wafer carrier which prevented chemical interaction, and thus subsequent film degradation. Films with high critical current densities, $J_c > 10^6$ A/cm^2 at 77 K, and low microwave surface resistance, R_s approximately 8 mΩ (36 GHz, 77 K) were achieved on both sides of the substrate.

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