High critical current density $YBa_2Cu_3O_{7-\delta}$ coatings on $LaMnO_3$-buffered biaxially textured Cu tapes for coated conductor applications

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High critical current density (high-J_c) $YBa_2Cu_3O_{7-\delta}$ (YBCO) films were obtained by pulsed laser ablation on biaxially textured Cu substrates. To achieve epitaxy of $LaMnO_3$ (LMO) on Cu, thin epitaxial Ni overlayers were deposited on Cu tapes. The structure comprises the layer sequence of YBCO/LMO/Ni/Cu. For 200-nm-thick YBCO, self-field J_c values exceeding 1×10^6 A/cm^2 at 77 K were achieved. Characterization of these short prototype conductors revealed good structural and morphological properties. Magnetic analysis suggested that hysteretic loss due to the ferromagnetic Ni overlayer is minimal.

I. INTRODUCTION

Recently, enormous strides were made toward the applications of high-temperature superconductors (HTS) in power technologies. In such applications, long lengths of superconductors in the form of wire/tape are required. This requires that the substrate material be flexible and have low production cost. Moreover, from the performance point of view, the HTS-coated wire/tape should support high critical current density (high-J_c) in the presence of magnetic fields at liquid-nitrogen temperatures. This can only be achieved if the substrate material has a very sharp, well-developed biaxial texture, so that it can impart sufficient crystalline alignment to the HTS material to ameliorate the pervasive problem of weak-linked, high-angle grain boundaries.[1,2] One of the leading approaches to develop this technology is the so-called rolling-assisted biaxially textured substrates (RABiTS) process.[3–5] In this technique, biaxial cube texture is generated in the base metal substrate by employing the industrially scalable thermomechanical processing. Subsequently, the epitaxial deposition of compatible buffer layer(s) prevents detrimental chemical interactions with the substrate and at the same time yields chemically and structurally compatible surfaces for the epitaxial growth of HTS films. To date, much of the published work using the RABiTS technique has utilized high-purity Ni (99.99%) as the textured material. However, the low mechanical strength and ferromagnetism (FM), with a Curie temperature of 627 K and a saturation magnetization 57.5 G cm^3/g (emu/g), present significant challenges in long-length manufacturing and hinder usage in applications where alternating current (ac) losses are an issue. To overcome these issues, recently Ni-based alloys such as Ni–Cr, Ni–Cu, Ni–Cu–Al, Ni–W, and Ni–V were investigated regarding the development of biaxial texture in these low-magnetic and strengthened materials.[6–15] These dilute alloy substrates are characterized by strong cube texture, significantly reduced Curie temperature and magnetization, and considerably improved tensile strength. On such Ni alloys as well as on pure Ni tapes, development of various buffer layer combinations, including noble metals, such as Pd, Ag, and Pt, and insulating and conducting oxides, such as CeO_2, Y_2O_3, Gd_2O_3, $La_2Zr_2O_7$, YSZ, $La_{0.7}Sr_{0.3}MnO_3$, $LaNiO_3$, and $SrRuO_3$, were investigated by a variety of vacuum-based vapor deposition and/or nonvacuum-based chemical solution techniques.[16–26] Those studies have demonstrated that moderate to high-quality epitaxial $YBa_2Cu_3O_{7-\delta}$ (YBCO) films can be grown on single or multilayer buffer structures with J_c values ranging from 0.1×10^6 to 2×10^6 A/cm^2 at 77 K and self-field.

Another possible alternative for the production of long-length RABiTS-based coated conductors is the use of Cu-based tapes as the substrate material. These materials would offer several advantages over Ni: the absence of FM, substantially lower material cost,[27] and potentially lower electrical resistivity. The latter may be important for practical applications, where the textured Cu substrate could provide the necessary stabilization to the HTS coating in an overcurrent situation. In this case, a conductive buffer layer must be developed to couple the HTS layer to the underlying Cu substrate, thereby mitigating the need for any additional stabilizing metal cap layers.[28] In addition, the attainment of sharp cube texture in Cu by thermomechanical treatment has been reported.[29,30]

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The main drawback to utilizing Cu is its poor oxidation resistance. Since the epitaxial growth of oxide films generally requires elevated temperatures and oxidizing environments, prevention of unfavorably oriented CuO/Cu_2O on the Cu surface presents a major challenge. So far, there have been limited reports on the development of buffer layers and HTS coatings on Cu or Cu alloys for coated conductor applications. Previous results indicated either poor buffer-layer texture, with no attempts to grow HTS films, or YBCO coatings exhibiting low-J_c values.[12,14,30]

Here we present an alternative route for buffer layer fabrication on Cu. First, thin Ni overlayers have been developed on Cu–RABiTS to obtain reproducible epitaxial growth of subsequent oxide films.[31] As the oxide buffer layer, we have chosen $LaMnO_3$ (LMO) since our previous research demonstrated the viability of $LaMnO_3$ as a buffer layer for the fabrication of high-J_c YBCO films on Ni-based and on single-crystal Cu templates.[32–34] In that work, YBCO films deposited on highly textured LMO buffer layers on Ni and Ni-alloy tapes showed high superconducting transition temperatures (T_c = 90–92 K) and high-J_c (77 K, self-field) values ≈ 1.3×10^6 A/cm^2.[32,33] Here, we report the achievement of high-J_c (>1×10^6 A/cm^2 at 77 K) YBCO coatings on biaxially textured Cu substrates. The structural, magnetic (hysteretic loss), electrical, and superconducting properties of these short prototype conductors are reported.

II. EXPERIMENTAL

Biaxially textured 50 μm Cu tapes were prepared by consecutive cold rolling of copper bars to greater than 95% deformation. Details of the rolling and annealing conditions will be reported elsewhere.[35] After the final rolling, the substrates were annealed in vacuum at 800 °C for 1 h in a vacuum furnace with a base pressure of 2×10^{-6} torr, to obtain the desired {100}⟨100⟩ cube texture. Deposition of the Ni overlayers was performed by direct current-magnetron sputtering in a reducing forming-gas (96% Ar + 4% H_2) environment at a substrate temperature of 500 °C. The film thickness was approximately 1.7 μm. Conditions for deposition of Ni were optimized by examining the quality of epitaxy and surface roughness of the Ni films by atomic force microscopy. These results will be published elsewhere.[31] Subsequent growth of LMO layers was accomplished by radio frequency-magnetron sputtering at temperatures ranging from 570 to 670 °C. The sputter target was made from single-phase LMO powder, prepared by solid-state synthesis, and lightly packed into a copper target tray. Typical sputtering conditions consisted of a sputter-gas mixture of forming gas and 2×10^{-5} torr of H_2O at a total pressure of 4 mtorr. Water vapor provided low-level oxygen for the stability of the oxides, and forming gas helped suppress the oxidation of the Ni film surface as well as the Cu tape. The LMO thickness was kept around 300 nm.

The YBCO films were grown by pulsed laser deposition, using a KrF excimer laser system operated at an energy density of ≈2 J/cm^2 and a repetition rate of 15 Hz. During YBCO deposition, the substrates were maintained at 780 °C in 120 mtorr of O_2. After deposition, the samples were first cooled to 500 °C at a rate of 5 °C/min; then the O_2 pressure was increased to 550 torr, and the samples were cooled to room temperature at the same rate. Typical YBCO film thicknesses were 200 nm. Film crystal structures were characterized with a Huber high-resolution x-ray diffractometer (XRD), and microstructural analyses were conducted using a JOEL model, JSM-840 scanning electron microscopy (SEM). Magnetization measurements were conducted in a superconducting quantum interference device (SQUID)-based magnetometer at temperatures 5–100 K, in fields, H, up to 60 kOe. A standard four-probe technique was used to evaluate the electrical transport properties, including T_c and J_c of the YBCO films. For those measurements, electrical contacts of silver were deposited onto the samples with dimensions of 20 mm in length and 3 mm in width using direct current-magnetron sputtering followed by O_2 annealing in 1 atm for 30 min at 500 °C. Values of J_c were assigned at a 1 μV/cm criterion.

III. RESULTS AND DISCUSSION

Figure 1 shows the XRD θ–2θ spectrum of a buffered Ni/Cu substrate with the LMO deposited at 570 °C. It is clear from the figure that both the Ni and LMO layers exhibit a good c-axis-oriented growth on Cu, as evidenced by the strong (00l) reflections. The ratio of integrated intensities for the LMO (110) to (002) rocking

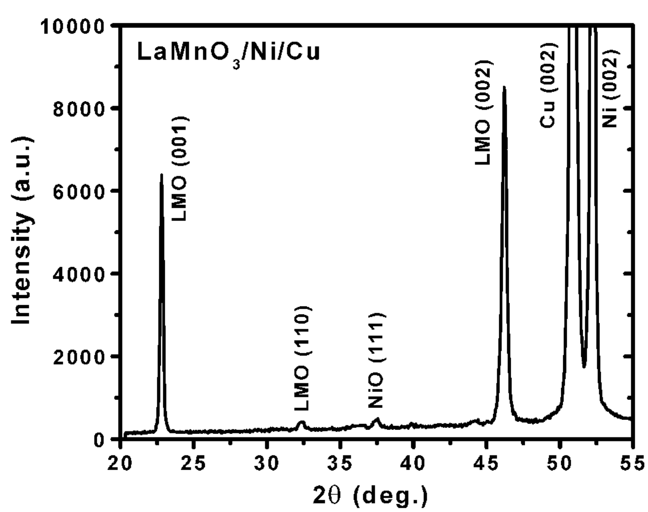

FIG. 1. XRD θ–2θ pattern of LMO film grown on a Ni-capped biaxially textured Cu substrate.

curve peaks is less that 4%, indicating an insignificant volume of the LMO is (110) textured. The presence of this component may be attributed to the presence of a small amount of (111)-oriented NiO at the interface after LMO deposition. Epitaxial orientation of the LMO buffer layers was characterized by XRD pole figure analysis. Typical background-corrected logarithmic (111) pole figures of a 300-nm-thick LMO film, along with that of the underlying Cu substrate, are illustrated in Figs. 2(a) and 2(b), respectively. Both reveal sharp cube texture and cube-on-cube epitaxy with the in-plane LMO[110]// substrate[110]. Quantitative analyses of the pole figure intensities indicate cube volume percentages of 95% and 96%, for the LMO layer and Cu substrate (along with the Ni overlayer), respectively. The ω-scan rocking curve on the (002) and ϕ-scan on the (111) peak reflections for LMO, Ni, and Cu yielded out-of-plane and in-plane peak-width full width half-maximum (FWHM) of $\Delta\omega = 9.2°$, 7.7°, and 6.6° and $\Delta\phi = 8.9°$, 6.8°, and 6.3°, respectively. Clearly, a good epitaxial relationship was obtained among the multilayer stack. The slight increase in both out-of-plane and in-plane texture from the Ni/Cu substrate to the LMO layer probably arises from the relatively low deposition temperature for the growth of LMO layers. Optimization of processing conditions for LMO is underway to develop nearly complete (00*l*) texture with reduced FWHM values.

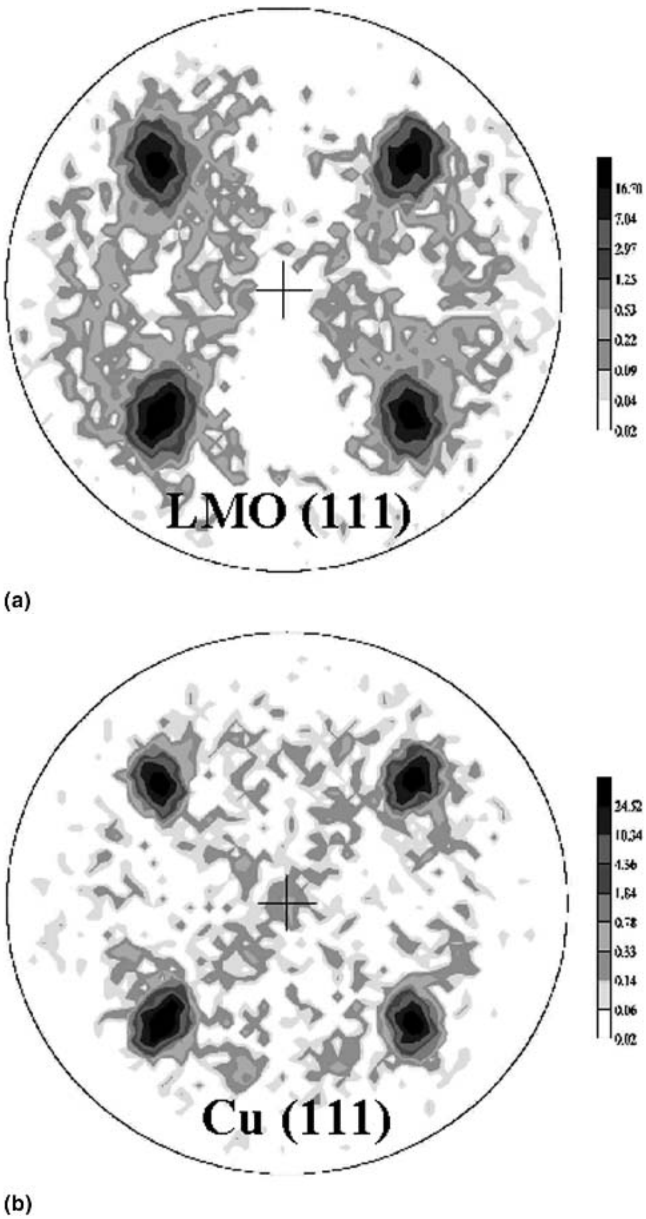

FIG. 2. Background-subtracted, logarithmic scale (111) pole figures of (a) a 300-nm-thick LMO film deposited on Ni/Cu and (b) the underlying biaxially textured Cu substrate.

The YBCO films deposited on this LMO/Ni/Cu substrate were epitaxial with out-of-plane and in-plane FWHM of $\Delta\omega = 9.3°$ and $\Delta\phi = 8.5°$, respectively, and exhibited a smooth, uniform, and dense surface morphology, as illustrated by the SEM micrograph in Fig. 3. This surface microstructure is similar to that observed for the YBCO films on LMO-buffered Ni-based substrates.[33] Electrical transport and superconducting property characterization results are shown in Fig. 4, which displays the magnetic field dependence of transport $J_{c,t}$ at 77 K, with the field applied parallel to the *c* axis. The inset of this figure shows the self-field *I–V* curve of the YBCO/LMO/Ni/Cu, yielding a critical current (I_c) value of 6.75 A (at 1 μV/cm criterion) with the corresponding $J_{c,t}$ of 1.1×10^6 A/cm^2. The high-T_c value approximately 90 K implies that LMO not only blocks Ni diffusion (as shown in the work on Ni-based substrates[33]) but, in conjunction with the nickel layer, also acts as a Cu diffusion barrier. Studies are being made to confirm the overall levels of interdiffusion. The present results represent the first reported achievement of high-J_c YBCO on Cu-based RABiTS.

To examine the current density–temperature characteristics of YBCO/LMO/Ni/Cu conductors, magnetic hysteresis measurements were carried out with a magnetic

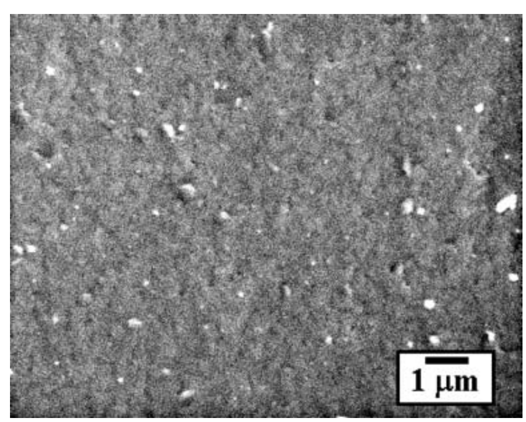

FIG. 3. SEM micrograph of the surface morphology for a YBCO film grown on LMO/Ni/Cu architecture.

field applied normal to the tape. It should be mentioned that these measurements were conducted on another sample having a lower transport $J_{c,t} = 0.6 \times 10^6$ A/cm^2, where the processing parameters of both the Ni and LMO layer depositions were not yet optimized. Nevertheless, the measurements should adequately reflect the relative current density versus temperature behavior of YBCO films on Cu–RABiTS. The magnetic field dependence of persistent current density $J_{c,m}$, as estimated from the magnetic hysteresis loops using the Bean critical state model, $J = 10(M_+ - M_-)/R$, is plotted in Fig. 5. Here, M_+ and M_- are the upper and lower branch magnetization values, respectively. These values were corrected for the small ferromagnetic hysteresis of the substrate, measured at 100 K. The effective circulation radius, R, of the current is estimated using $2R = b(1 - a/3b)$, where a and b are the short and long transverse dimensions of the sample, respectively. We assumed that the critical current in the *ab*-basal plane of the YBCO film is isotropic and the total area covered by the film is used as the relevant loop dimension rather than the grain size. Values in zero-applied field range from $J_{c,m} = 3.5$ MA/cm^2 at 5 K to 0.18 MA/cm^2 at 77 K. The latter value is about one-third that of $J_{c,t}$ obtained by the transport measurements (0.6×10^6 A/cm^2). The discrepancy is most likely due to the differences in electric field criteria in defining J_c. As the temperature increases, $J_{c,m}$ decreases approximately exponentially for a wide range of fields with $J_{c,m}(T) \simeq J_{c,m}(0)\ \exp(-T/T_0)$, with $T_0 \approx 23$ K. These results imply that thermally activated depinning of vortices produces the observed falloff of $J_{c,m}$.

Besides the J_c performance, another important consideration is the overall energy loss in ac applications. It is well known that ac gives rise to hysteretic loss in the superconducting material, even for currents below I_c. Moreover, additional losses can occur in FM substrate material due to hysteretic movement of magnetic domains. In the present case, salient features of the FM Ni layer on the Cu substrate need to be determined. While the Cu substrate is nonmagnetic, the Ni overlayer introduces an additional mechanism for potential energy loss in ac due to the FM hysteresis. Thus, we measured the magnetic response of the substrate at 100 K, just above the superconductive T_c but far below the 627 K Curie temperature of Ni. The results are shown in Fig. 6. With $H \parallel$ tape to reduce demagnetizing effects, the applied fields of ±800 G nearly saturate the Ni; the full saturation moment yields a value of 1.7 μm for the Ni thickness. The coercive field of the coating is about 20 Oe,

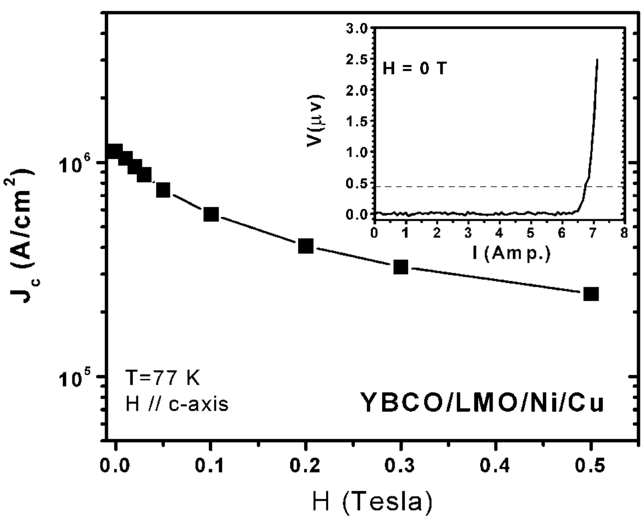

FIG. 4. Magnetic field dependence of the transport J_c, measured at 77 K, for a YBCO film grown on LMO/Ni-buffered Cu substrates. The inset shows the *I–V* curve for the same sample measured in self-field. The dashed line designates the voltage criterion to extract J_c.

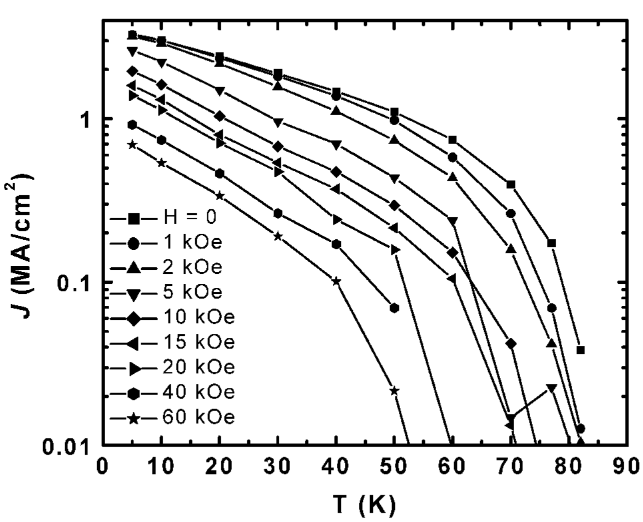

FIG. 5. Persistent current density J versus temperature T, with magnetic field H applied normal to the tape. Values were obtained from a critical state analysis of the magnetization and were corrected for hysteresis from the Ni layer.

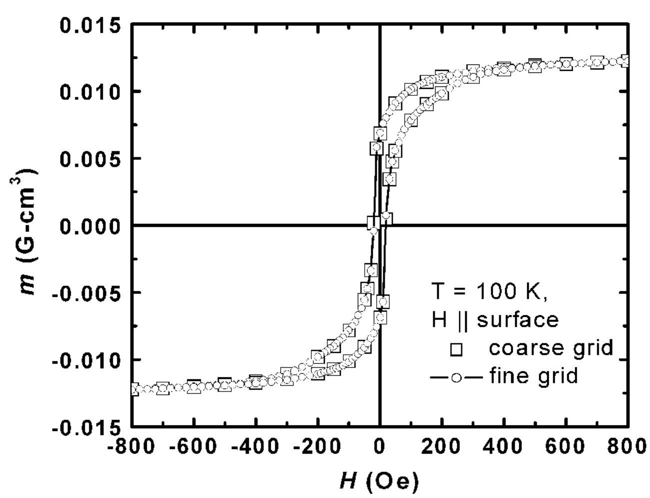

FIG. 6. Ferromagnetic moment hysteresis loop of a YBCO/LMO/Ni/Cu coated conductor at $T = 100$ K, with H in the plane of the tape.

somewhat larger than that of pure RABiTS nickel,[13] apparently due to additional defects in the deposited film, and possibly arising from some level of copper interdiffusion. By integrating the area inside the hysteresis loop, we obtain a FM loss per cycle per unit area of tape = 11.4 erg/cm^2 per cycle. This is the worst case scenario, since the material experiences its largest possible hysteretic loss. Referenced to a unit volume of Ni, this corresponds to 6.8×10^4 erg/cm^3 per cycle, compared with a loss (at 77 K) of 1.8×10^4 erg/cm^3 for fully processed, biaxially textured Ni–RABiTS and 0.2×10^4 erg/cm^3 for similarly processed Ni–7 at.% Cr alloy with reduced FM.[13] However, the present materials contain little actual Ni, as the layer is thin. Consider, for example, 1 m of tape with width of 1 cm and substrate metal thickness of 50 μm, operating at 60 Hz: for the present Cu-based substrate, the *maximum* FM power loss would be 7 mW, compared with 42 mW for the all-Ni case and 6 mW for the case with Ni–7 at.% Cr. These losses of 6–7 mW correspond to modest additions (10–20%) to the losses expected in achievable YBCO-based conductors.[13] In conductor configurations either with reduced Ni overlayer thickness or with lower ac magnetic fields, the FM losses would be still smaller, of course.

IV. SUMMARY

We have demonstrated the successful fabrication of high-quality epitaxial YBCO coatings on nonmagnetic, biaxially textured Cu substrates, employing the layer sequence $LaMnO_3$/Ni/Cu. Property characterization revealed good-quality crystalline structure and surface characteristics for YBCO films on this architecture. For 200-nm-thick YBCO, self-field J_c values exceeding 1×10^6 A/cm^2 were achieved at 77 K. Magnetic studies suggest that the magnetic loss resulting from the Ni overlayer should be small compared to the hysteretic losses arising from the superconducting layer on an implemented coated conductor. These observations demonstrate a potentially promising route toward the fabrication of low-cost, nonmagnetic YBCO-based coated conductors for power applications of superconductivity. Further support requires additional studies that provide optimization of the nickel and LMO layer thicknesses and quality, especially with regard to their stability and function under conditions for the deposition of practical, thick YBCO coatings. Also of interest is the development of conductive buffer layers for a stabilized overall structure.

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