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NEW APPROACH TO DEPOSITING YTTRIA-STABILIZED ZIRCONIA BUFFER LAYERS FOR COATED CONDUCTORS (POSTPRINT)

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14. ABSTRACT
A new approach for the production of yttria-stabilized zirconia (YSZ) oxide buffer layers directly on metal rolling-assisted biaxially textured substrates (RABiTS) is described in this paper. This represents a significant advance over existing techniques and avoids the need for complicated steps to avoid substrate oxidation during direct deposition of oxides. Current densities of about 1 MA/cm^2 have been achieved for $YBa_2Cu_3O_{7-\delta}$ layers on the YSZ buffer, with an intermediate CeO_2 layer. The process consists of reactive sputtering of a $Y_xZr_{1-x}N$ film directly on the RABiTS, which adopts its biaxial texture. This nitride film is then converted to YSZ via a thermal oxidation step. The YSZ films retain the texture of the nitride film (and of the RABiTS) through local syntaxy. In many cases, YSZ films exhibit improved biaxial texture over that of the RABiTS substrate. Nitrides can be sputter deposited at much higher rates relative to oxides, making the approach industrially scalable and economical.

15. SUBJECT TERMS
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New approach to depositing yttria-stabilized zirconia buffer layers for coated conductors

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A new approach for the production of yttria-stabilized zirconia (YSZ) oxide buffer layers directly on metal rolling-assisted biaxially textured substrates (RABiTS) is described in this paper. This represents a significant advance over existing techniques and avoids the need for complicated steps to avoid substrate oxidation during direct deposition of oxides. Current densities of about 1 MA/cm^2 have been achieved for $YBa_2Cu_3O_{7-\delta}$ layers on the YSZ buffer, with an intermediate CeO_2 layer. The process consists of reactive sputtering of a $Y_xZr_{1-x}N$ film directly on the RABiTS, which adopts its biaxial texture. This nitride film is then converted to YSZ via a thermal oxidation step. The YSZ films retain the texture of the nitride film (and of the RABiTS) through local syntaxy. In many cases, YSZ films exhibit improved biaxial texture over that of the RABiTS substrate. Nitrides can be sputter deposited at much higher rates relative to oxides, making the approach industrially scalable and economical.

I. INTRODUCTION

High-temperature superconductor (HTS)-coated conductors represent the next generation of HTS wire technology, with primary application in power generation, power transmission, and compact motors for both commercial and military use. HTS tapes, formed by HTS deposition onto buffered, biaxially textured metal substrate tapes, are intended to replace copper as a conductor in these applications. Cold-rolled and annealed nickel can yield tapes with strong biaxial texture, called rolling-assisted biaxially textured substrates (RABiTS).[1–4] $YBa_2Cu_3O_{7-\delta}$ (YBCO) cannot be deposited directly onto the tapes due to tape oxidation and chemical interdiffusion issues,[5] so buffer layers must be used. The buffers act as a chemical diffusion barrier, adopt the biaxial texture of the tape, and provide a surface upon which (001) biaxially textured YBCO can be deposited. Control of the biaxial texture of the final YBCO superconducting layer is critical to the success of the YBCO-coated conductor. Misorientation at YBCO grain boundaries, both [100]-tilt and [100]-twist, have a significant impact on critical current density (J_c); nearly an order-of-magnitude drop in intergrain critical current densities is observed between 5° and 10° of [100] misorientation.[6–8] Theoretical calculations show that the maximum reduction in J_c in going from a short sample to a kilometer length is only 10–20% for the typical texture attained in most RABiTS tapes, thereby making this a very attractive process for fabrication of long length high-J_c conductors.[9]

The current approach being pursued by several research groups worldwide makes use of a three-layer buffer architecture: a few tens of nanometers of CeO_2/Y_2O_3 seed layer, several hundreds of nanometers of yttria-stabilized zirconia (YSZ) barrier layer, and a few tens of nanometers CeO_2 cap layer. From 1 to 3 μm YBCO is deposited onto the buffer layer to produce a coated conductor.[1–3] Short-length samples with J_c over 1 MA/cm^2 have been demonstrated reproducibly.[1,10] However, kilometer lengths are needed, and the technical challenges to develop an economical continuous-production process for this architecture are significant.

A primary concern with the current architecture is due to the difficulties inherent in oxide growth on elemental (metal) substrates, most importantly the issue of substrate

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oxidation. Precise techniques are required to deposit the initial oxide seed layer. Any oxidation of the underlying substrate during growth of the seed layer results in loss of texture. In the case of pure nickel substrates and the current architecture, NiO forms quite readily and interferes with the epitaxial growth of the CeO_2 layer, although some recent work has focused on attempts to use the native NiO layer for subsequent epitaxy.[11] For CeO_2 growth, a precise H_2O/H_2 balance must be maintained[12,13] to avoid nickel oxidation. New alloys such as Ni–Cr or Ni–W that have suppressed secondary recrystallization and greater yield strengths than pure Ni have been developed.[4,14] Ni–Cr (13%) is also nonmagnetic at 77 K. The concern for substrate oxidation is exaggerated by the presence of Cr or W, which oxidize more readily than Ni. One solution is to deposit an additional layer in the buffer layer stack—a transient nickel layer deposited directly on the substrate. Regardless of the substrate and protective layers used, the rates at which oxides can be deposited are low, and this must be addressed for economical, continuous production. For transitioning the current approach to an industrially scalable process, these issues present significant technical and engineering challenges and, more importantly, impose a high cost, which impedes deployment of HTS-coated conductors as competitive products.

These problems are circumvented by using a novel process that avoids substrate oxidation issues. Nitrides have been under investigation as potential buffer layers for use with textured nickel substrates for the past five years.[15] The observation that biaxially textured nitride films on Ni RABiTS maintain the same texture after being oxidized led to the development of a process to exploit this phenomenon, with the goal of bringing next-generation coated conductors a step closer to economical production. The process, termed epitaxial conversion to oxide via nitride oxidation (ECONO),[16] consists of the deposition of a biaxially textured nitride layer directly on a substrate, which is then converted to an oxide via a thermal oxidation step. In the case of Ni RABiTS, a $(Y_{0.2}Zr_{0.8})N$ precursor (YZN) is deposited and subsequently converted to YSZ. The oxidation proceeds in a highly cooperative manner whereby the texture both in- and out-of plane is maintained. A CeO_2 cap layer can be deposited by the conventional means to provide a template layer for the growth of high-quality YBCO by the two-step BaF_2 process[17] or other methods. High J_c values (about 1 MA/cm^2) on short samples have been demonstrated using nickel RABiTS substrates. Films are fully dense and smooth and free of the types of defects common in oxides deposited directly by radio frequency (rf) sputtering. The high deposition rate at which nitrides can be grown is expected to be industrially advantageous, for example in a high-rate reel-to-reel deposition system.

Nitride films are much tougher than oxide films, and as such, RABiTS tapes can be spooled and stored between the nitride deposition and thermal oxidation steps. The nitride films are robust and can survive deformation to tight radii of curvature without visible cracking (e.g., 0.7 cm), that oxide films cannot.[18]

In this article, we describe the process and present data showing the degree to which the biaxial texture of the RABiTS substrate is adopted by the various layers, all the way to YBCO layers deposited on the buffer layer stacks. Electrical measurement results of the same film are also presented. The morphology of the films, determined by scanning electron microscopy (SEM) and transmission electron microscopy (TEM), are described. The nitride-to-oxide conversion mechanism is also discussed.

II. EXPERIMENTAL

Nitride films were grown in a direct current (dc) reactive magnetron sputtering system with an all-metal sealed chamber with a sample-insertion load-lock system, with typical base pressures of about 10^{-8} torr. Magnetron sputtering sources (US GUN I, US Inc., San Jose, CA) utilized 5-cm-diameter, 3.2-mm-thick sputtering targets of $Y_{0.2}Zr_{0.8}$ (99.95% purity). Background pressures of 15–20 mtorr of Ar-N_2 (>99.999%) were maintained during growth. Annealed cube-textured substrates of Ni and Ni-alloy RABiTS from Oak Ridge National Laboratory (ORNL), Oak Ridge, TN, were precleaned in organic solvents (acetone, ethanol, chloroform) using ultrasonic agitation before nitride deposition by a two-temperature technique, at temperatures from 500 to 900 °C. Initial growth (first about 30 nm) at a low temperature favors planar growth, and the high temperatures used for the remainder promote a narrow texture.

YZN buffer layers deposited onto Ni and Ni–Cr (13%) RABiTS substrates were converted to YSZ by an *ex situ* thermal oxidation step in an atmospheric-pressure chamber, using hydrogen-water vapor-argon gas mixtures and temperatures from 500 to 800 °C. Oxidation times ranged from 0.5 to 20 min. Standard pulsed laser deposition (PLD) was used to deposit CeO_2 layers approximately 20 nm thick and YBCO layers approximately 250 nm thick on selected converted-YSZ/RABiTS substrates at ORNL.

Crystalline quality of films was evaluated using θ-2θ and ϕ-scan x-ray diffraction (XRD) analyses. Computer-optimized Gaussian fitting was used to compute the full-width at half maximum (FWHM) of XRD peaks as a measure of texture quality. Pole figures were collected for selected specimens. Morphology and microstructure of the surface was characterized by SEM, and of the bulk in cross-section by TEM and high-resolution TEM (HRTEM), with the aid of a Gatan imaging filter (GIF)

(Gatan Inc., Pleasanton, CA). Critical current (I_c) and J_c were determined by a standard four-point configuration with a 1 μV/cm criterion at 77 K.

III. ECONO PROCESS

A. Process description

The ECONO process allows the production of high-quality epitaxial oxide layers on nonoxide substrates, achieved in two steps. A nitride film is deposited onto a substrate such that it adopts the biaxial texture of the substrate. The second step consists of conversion of the nitride to an oxide through a thermal oxidation step. The oxide maintains the same orientation as the nitride, so an epitaxial nitride will produce an epitaxial oxide.

The entire procedure for YSZ buffer layers on RABiTS using the process is shown schematically in Fig. 1. A precursor nitride layer of composition $Y_{0.2}Zr_{0.8}N$ is first deposited on a Ni, Ni–Cr, or other composition RABiTS substrate. The film adopts the same biaxial texture as the RABiTS substrate, believed to occur through local epitaxy with individual Ni-alloy grains. This texture is retained after oxidation to yield a biaxially textured YSZ film. After this, ceria cap and YBCO superconductor layers are deposited in the conventional manner.

The oxidation step can be conducted either *in situ* directly after nitride deposition, or *ex situ* in a conventional furnace with a controlled atmosphere. As described in more detail below, the conversion from nitride to oxide is very fast and proceeds from the surface of the nitride layer, which then serves as the template for subsequent oxide conversion inward. The oxidation proceeds in a highly cooperative manner whereby grain orientations both in and out of plane are maintained.

B. Process advantages

The key advantages of the process include: (i) deposition of a smooth, high-quality nitride layer, relatively free of defects, is straightforward (defects are prevalent in oxide deposition by reactive or rf sputtering), (ii) buffer layer structure is simplified, and (iii) the nitride can be deposited at relatively high rates compared to oxides.

It is important to note that even though the above approach can yield a compositionally complex nitride stack, the deposition of all nitride layer(s) can be performed in a single step, in the same chamber, without compromising the process efficiency as compared to a single-layer YZN deposition.

One of the key competitive advantages offered by the process is the ability to perform deposition at high rates. To meet the cost target of the final YBCO conductor, costs associated with fabrication of each component must be minimized without compromising film quality, and a critical parameter affecting the cost of buffer layer production is the deposition rate. Direct deposition of nitride layers by reactive sputtering is fast relative to oxide deposition techniques. Sputtered nitride layers are dense and uniform, with high-quality texture.

Nitride precursor films offer several advantages. Cubic nitrides can be epitaxially deposited on reactive metals such as Ni in a straightforward manner. The nitride deposition on Ni is done in a nitrogen-containing ambient, and because Ni does not form a nitride, the strict process control required for the current oxide/Ni deposition techniques is not needed. No separate partial pressure of H_2O is required, so the process is easy to control and to scale up. Nitrides have been used in the tool industry for many years, and their manufacturability in various configurations has been demonstrated. High-rate deposition (up to 1 μm/min) of nitride coatings is being developed and used in many industrial applications.

Most metal nitrides exhibit a cubic symmetry with a range of lattice parameters suitable for the coated conductor application. The extensive solid solubility of transition-metal nitrides and rare-earth nitrides can be used to tailor lattice parameters. The compositional flexibility and chemical compatibility among these nitrides potentially can be used to tailor other properties as well.

C. Broad process applicability

Broader significance beyond the use for coated conductors or HTS devices is also apparent and deserves much attention. The ECONO process should be considered a generic epitaxial template technology suitable for

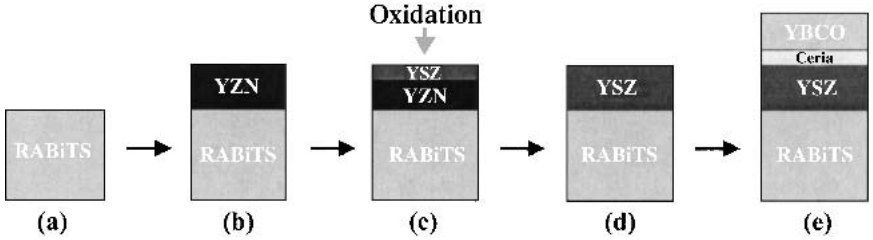

FIG. 1. Schematic diagram of YBCO growth on a RABiTS using the ECONO process. (a) A bare RABiTS is prepared. (b) $Y_{0.2}Zr_{0.8}N$ is deposited. (c,d) The nitride is converted to the oxide (YSZ) by a thermal oxidation step to form an oxide film on the alloy RABiTS. (e) CeO_2 and YBCO are then deposited by PLD, TFA, or other suitable technique.

a broad range of commercial applications. In addition to RABiTS, the process has been demonstrated for depositing high-quality biaxially textured YSZ layers on silicon and sapphire. YSZ, with cubic symmetry, is an excellent template layer for subsequent growth of oxide device layers and has been used for the deposition of many oxides, among them $Pb(Zr,Ti)O_3$, $SrTiO_3$, $(Ba,Sr)TiO_3$, YBCO, and RuO_2.[19,20] YBCO films on silicon, using YSZ as the buffer layer, are being pursued by industry for fabricating magnetic resonance imaging systems, bolometers, high-speed computing devices, high efficiency radio frequency filters for telecommunication systems, and others.[21,22] Reactive sputtering is a well-established thin film deposition process routinely used in the semiconductor industry and can be easily integrated into a manufacturing process. Nitrides also exhibit a wide range of solid solubility and incorporate many transition and rare-earth metals such that properties can be tailored through graded compositions. For example, a more oxidation-resistant nitride could be deposited directly on the substrate with subsequent deposition of YZN. The wide solid solubility range of nitrides will also allow for tailoring of lattice parameters to fit a wide variety of substrates for better epitaxial quality.

IV. RESULTS AND DISCUSSION

A. YSZ buffer layer growth on Ni RABiTS

YZN films grown on Ni RABiTS substrates by high-rate reactive sputtering are single phase and show excellent orientation retention from RABiTS to converted YSZ. The θ-2θ XRD scan of a YZN/Ni-RABiTS film is shown in Fig. 2(a), and the θ-2θ scan of the film after conversion to YSZ is shown in Fig. 2(b). Both scans show only the desired phases. Proper control of the nitride deposition parameters is required to achieve high-quality biaxial texture. For example, if deposition temperatures are too high, excessive amounts of (111)-oriented YZN may form. Note that the (111) content is virtually absent (less than 1%) in Fig. 2(a). Rocking curve scans of the Ni RABiTS, YZN, and YSZ, all from the same sample, are shown in Fig. 2(c). The out-of-plane texture, measured by the FWHM in the rolling direction, of the YZN (3.4°) is improved over that of the Ni (5.5°), likely due to competitive grain growth in the nitride film. This tight out-of-plane orientation is maintained upon conversion of the YZN to YSZ (4.8°), with an overall improvement in texture of 0.7°. In some cases, the rocking curve narrows even further upon conversion to YSZ. This is undoubtedly controlled by some aspect of the conversion mechanics and is not yet fully understood. These values for out-of-plane orientation compare favorably with those of YSZ buffers grown by other techniques.

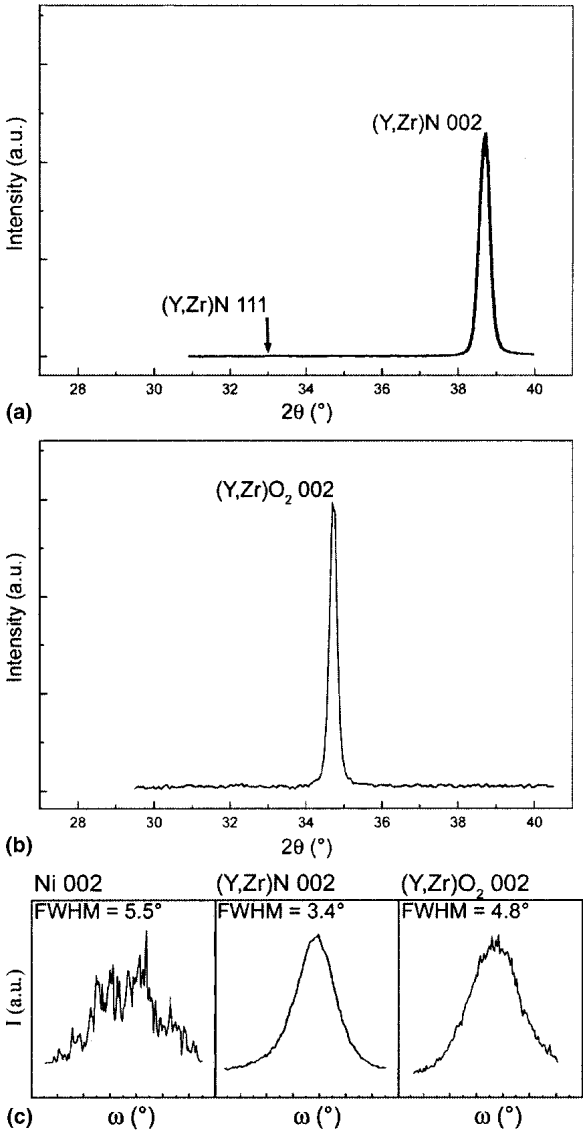

FIG. 2. Rolling-direction θ-2θ XRD scans of (a) (Y,Zr)N and (b) $(Y,Zr)O_2$ films on Ni RABiTS. The scans show only 002-oriented films, with no 111-oriented material, which is sometimes observed in films deposited under sub-optimal conditions. (c) Rocking curve XRD scans of the 002 peaks of Ni-RABiTS substrate, as-deposited (Y,Zr)N film, and converted $(Y,Zr)O_2$ film, all from the same sample. The rocking curves indicate that the out-of-plane texture of the $(Y,Zr)O_2$ is improved by 0.7° over that of the Ni-RABiTS substrate.

Biaxial texture of the RABiTS about a surface-normal vector is retained in addition to surface-plane orientation, with cube-on-cube epitaxy. ϕ-scans of the Ni, YZN, and YSZ from the same stack described above are shown in Figs. 3(a)–3(c), with FHWMs of 8.2°, 10.9°, and 11.0°, respectively. A slight increase in rotation about the surface normal, corresponding to degree of [001] tilt in subsequently deposited YBCO films, typically occurs. The increase in FWHM of ϕ-scan YBCO 111 peaks can ranges from about 0.3° to 3°, depending upon deposition methods and parameters. The texture obtained for a given

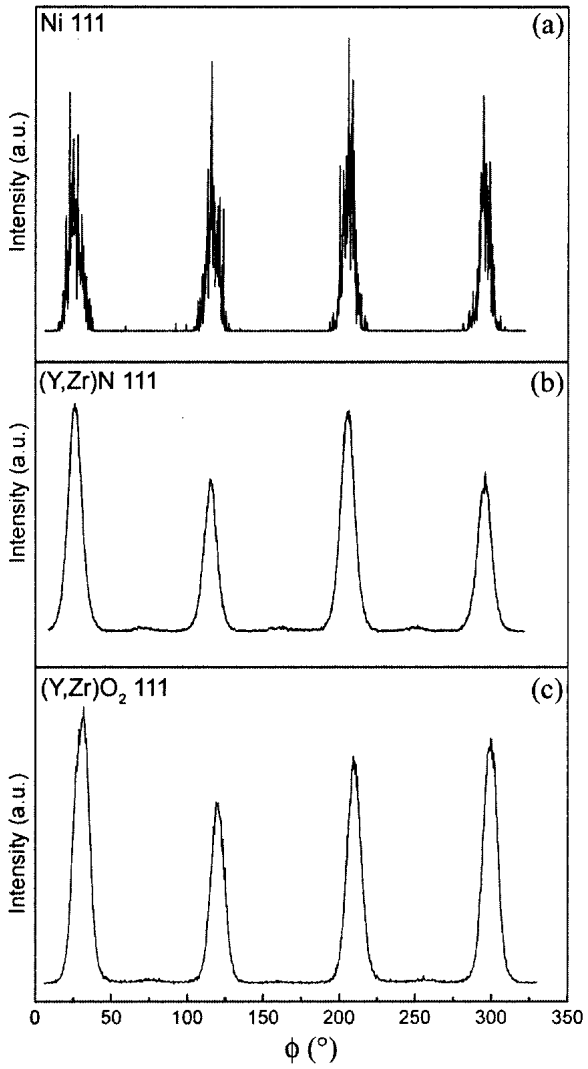

FIG. 3. ϕ XRD scans of the same sample described in Fig. 2, indicating that the biaxial texture of (a) the Ni-RABiTS was imparted to (b) the as-grown nitride film, which was maintained through conversion to (c) $(Y,Zr)O_2$. Only a single cube-on-cube surface normal orientation is observed. FWHM values of the peaks are 8.2°, 10.5°, and 11.0°, respectively.

set of conditions is reproducible, but is also dependent upon the texture quality of the RABiTS substrates. The texture retention occurs despite the large lattice parameter mismatch between YSZ and YZN (13%).

B. YSZ buffer layer growth on Ni–Cr RABiTS

The field of coated conductors has been moving toward alloy compositions such as Ni–Cr (13%) RABiTS to improve substrate strength as well as to render the substrate nonmagnetic at liquid-nitrogen temperatures for minimizing alternating current (ac) losses. Therefore, growths on these substrates have also been pursued. A nickel cap layer is typically deposited on these substrates to eliminate chromium oxidation issues. None is required using this process. Magnetron-sputtered YZN films grown on Ni–Cr (13%) RABiTS showed good in-plane and out-of-plane alignment. A θ-2θ XRD scan of a YZN film on Ni–Cr is shown in Fig. 4(a), and that of the YSZ converted from this film is shown in Fig. 4(b). The only peaks observed in the θ-2θ scans are the 002s, indicating a single surface-normal plane orientation. The out-of-plane orientation is good, with FWHM of the rolling-direction rocking curves of the Ni–Cr (13%), YZN, and converted YSZ of 5.8°, 4.2°, and 4.8°, respectively, an overall decrease of 1.0°. FWHM values measured about

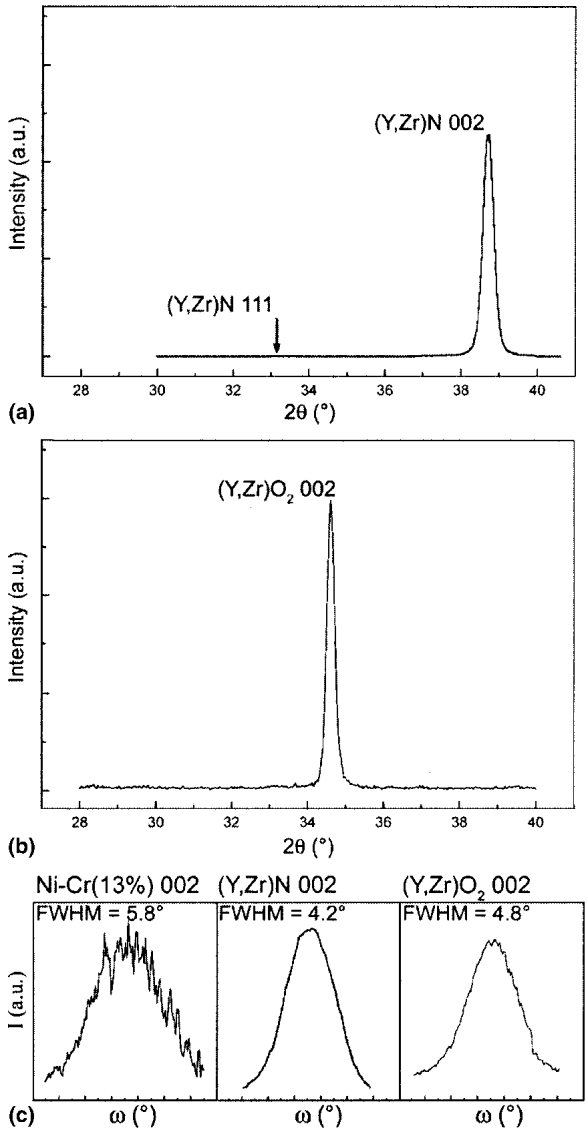

FIG. 4. Rolling-direction θ-2θ XRD scans of (a) (Y,Zr)N and (b) $(Y,Zr)O_2$ films on Ni–Cr (13%) RABiTS. The scans show only 002-oriented films, with no 111-oriented material, which is sometimes observed in films deposited under sub-optimal conditions. (c) Rocking curve XRD scans of the 002 peaks of Ni–Cr (13%) RABiTS substrate, as-deposited (Y,Zr)N film, and converted $(Y,Zr)O_2$ film, all from the same sample. The rocking curves indicate that the texture of the $(Y,Zr)O_2$ is improved by 1.0° over that of the Ni–Cr (13%) RABiTS substrate.

the rolling direction are similar, 7.5° for Ni–Cr (13%) and 6.4° for YZN (not shown), again showing a substantial improvement. 111 ϕ-scans of Ni–Cr (13%), YZN, and converted YSZ of the same stack [Figs. 5(a)–5(c), respectively] show a single in-plane orientation. The existence of only four peaks demonstrates a cube-on-cube epitaxial orientation. FWHM values are 7.2°, 10.6°, and 11.8°.

Thus, the process allows for direct formation of epitaxial YSZ on a Ni–Cr alloy substrate without predeposition of Ni prior to the oxide buffer. This is possible because, unlike oxide deposition, oxidation in this case proceeds from the film surface, in the substrate direction. Any potential oxidation of the substrate would be due to diffusion of oxygen through the already-converted YSZ film, the rate of which is negligible at the expected use temperatures of coated conductors. Oxygen diffusion in the YSZ at the YBCO deposition temperature might lead to some oxidation of the substrate, but this is not expected to affect the texture of the YSZ and therefore is not expected to result in a degradation of YBCO properties. High-J_c films have been grown (Sec. IV. C). Studies of the mechanical stability of these interfaces are underway, and strategies to prevent oxygen diffusion to the substrate during subsequent processing are under investigation. The ease of nitride growth and subsequent oxidation without Ni predeposition on Ni–Cr alloy substrates demonstrates a substantial advantage of this technique over conventional buffer processing and is additionally more amenable to industrial scale-up.

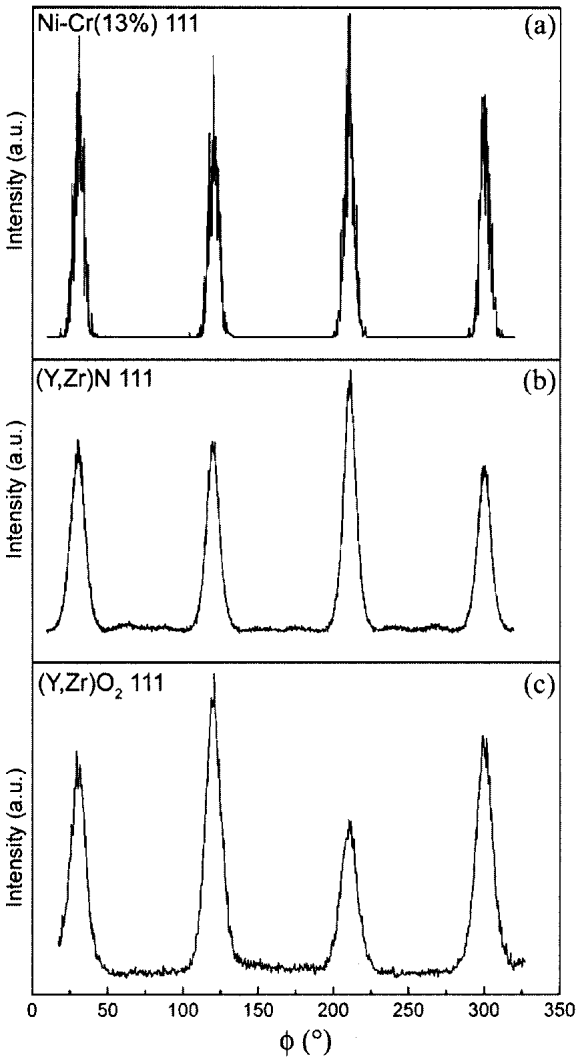

FIG. 5. ϕ XRD scans of the same sample described in Fig. 4, indicating that the biaxial texture of (a) the Ni–Cr (13%) RABiTS was imparted to (b) the as-grown nitride film which was maintained through conversion to (c) $(Y,Zr)O_2$. Only a single cube-on-cube surface normal orientation is observed. FWHM values of the peaks are 7.2°, 10.6°, and 11.8°, respectively.

C. Superconductor tape stack fabrication

Complete superconducting tape stacks, YBCO/CeO_2/YSZ / Ni-RABiTS, were fabricated by standard PLD at ORNL to deposit about 20 nm of CeO_2 and about 250 nm of YBCO on YSZ converted from YZN. This results in a YBCO film with nearly the same texture as the Ni RABiTS. A 103 pole figure of the YBCO layer of a complete stack is shown in Fig. 6. Only the single 001 orientation for YBCO is observed. The ϕ-scans of each of the layers of a typical stack are shown in Fig. 7. The FWHM of the 111 of the underlying RABiTS substrate is 9.2° in ϕ [Fig. 7(a)]. The ϕ-scan of the converted YSZ layer [Fig. 7(b)] has a FWHM of 10.0° in ϕ. Cube-on-cube epitaxy resulted in only a slight loss in preferred orientation of 0.8°. The ϕ-scan of the PLD-deposited CeO_2 layer [Fig. 7(c)] shows a slight improvement in texture, with a FWHM of 9.5°. The (001) YBCO layer maintains this texture, as shown in

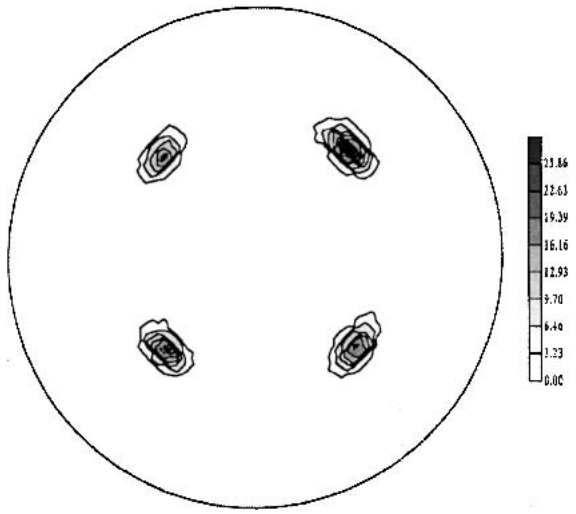

FIG. 6. YBCO 103 pole figure for a YBCO/YSZ/Ni sample. The YSZ was converted from a nitride film. Only a single biaxial orientation, corresponding to the cube-on-cube epitaxy of YSZ on the biaxially textured Ni, is evident.

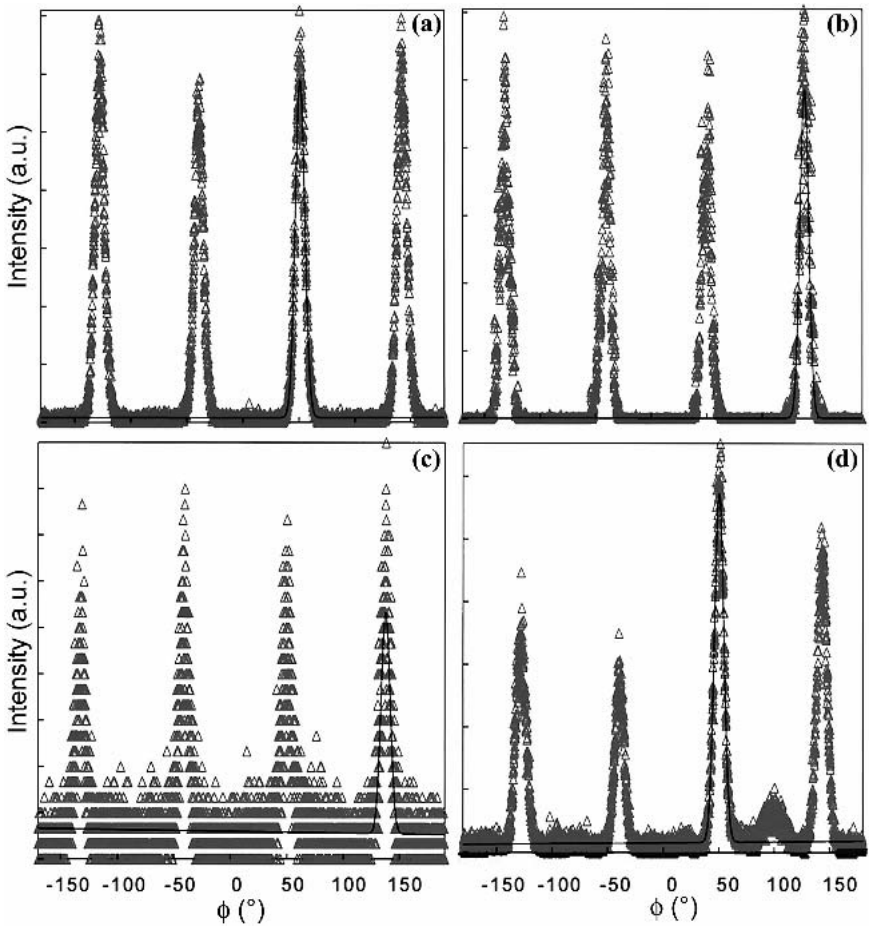

FIG. 7. ϕ-scans of a complete superconductor-tape stack; the decrease in texture about surface normal through the entire stack is only 0.3°. (a) Ni RABiTS with FWHM = 9.2°. (b) YSZ layer converted from nitride, with FWHM = 10.0°, showing the excellent retention of texture of the buffer layer. (c) CeO_2 buffer layer, with FWHM = 9.5°, an improvement of 0.5°. (d) (001) YBCO film with FWHM = 9.5°.

the ϕ-scan in Fig. 7(d), with a 9.5° FWHM. The overall decrease in texture from the Ni-RABiTS to YBCO is only 0.3°. These data demonstrate the high quality of YBCO that can be grown using an oxide buffer layer converted from a textured nitride precursor layer.

Electrical transport measurements from a 200-nm-thick complete YBCO/CeO_2/YSZ/Ni-RABiTS stack are shown in the I-V plot in Fig. 8. J_c of the film is calculated to be 1 MA/cm^2, with I_c = 6 A at 1 μV/cm criterion (film width = 3 mm).

D. Conversion mechanics

The mechanisms by which oxygen is exchanged for nitrogen are not completely understood. What is clear is that the degree to which orientation is maintained through the conversion process is determined by the first few atomic layers converted at the film surface. In other words, the YSZ grows syntaxially from the free surface, in toward the substrate. This is believed to be responsible for the improvement in texture of YSZ over YZN. The film appears to undergo competitive grain growth twice—once during YZN deposition, and once during conversion to YSZ. It has been suggested that this improvement may be in part attributable to the energetics of low-misorientation grain boundaries.

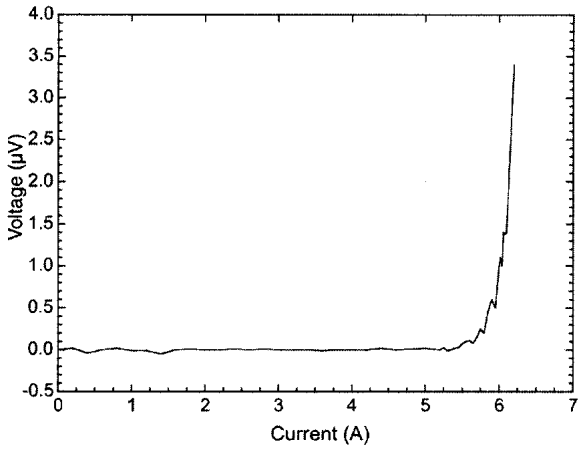

FIG. 8. I-V plot, measured on a 200-nm-thick YBCO film grown on CeO_2/YSZ/Ni-RABiTS, the same sample described in Fig. 7. J_c = 1.0×10^6 A/cm^2 at 77 K, measured by whole body transport current measurement in self-field using a 1 μV/cm criterion.

The cross-sectional TEM micrograph in Fig. 9(a) shows a partially converted film. The film is fully dense and smooth, with a thickness of about 220 nm. The boundary between the YSZ and YZN can be seen. (The Al was deposited as a protective layer during TEM specimen preparation.) The nitrogen map in Fig. 9(b), acquired by energy-filtered imaging of the same area, shows the boundary between the two phases more clearly. The conversion front is planar, indicating that the process should scale well.

Further detail of the conversion front can be seen in Figs. 10(a)–10(d). The boundary between the nitride and oxide phases is clearly visible. The local syntaxy of

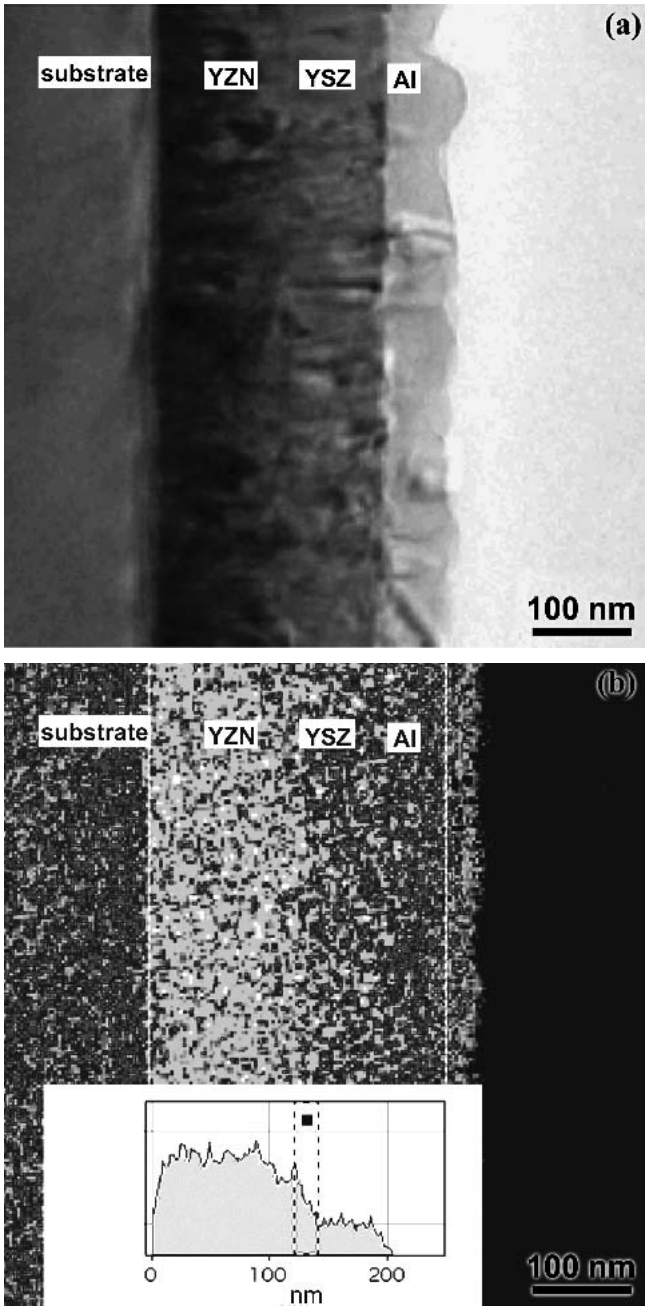

FIG. 9. (a) Cross-sectional morphology of a partially converted film, showing a fully dense and crack-free structure. (b) The energy-filtered elemental map of the same area shows the distribution of nitrogen in the film.

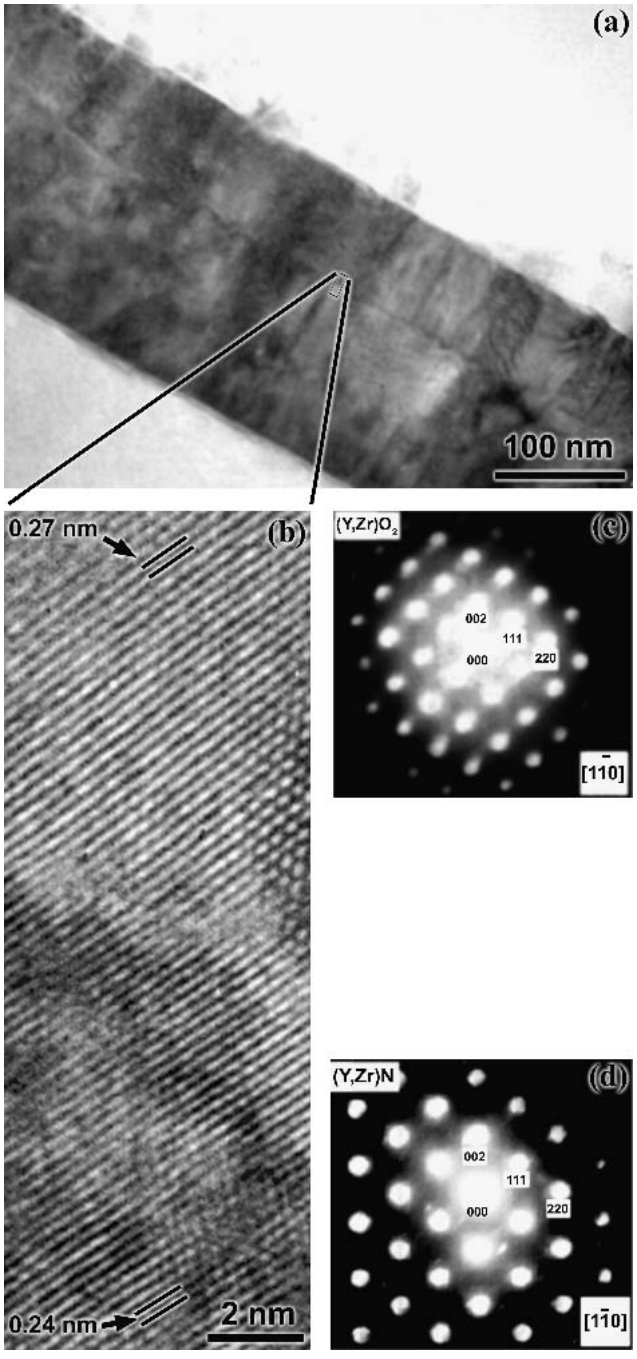

FIG. 10. Cross-sectional TEM characterization of a YZN/Si film halfway through the conversion process to YSZ. (a) The planarity of the growth front is apparent. Local syntaxy is maintained despite a 13% lattice mismatch between the two phases, as shown by the electron-diffraction patterns in (c) and (d), and by the HRTEM image of the conversion front in (b).

the material as it undergoes conversion is apparent in Fig. 10(b), and from the electron-diffraction patterns of the two grains shown, in Figs. 10(c) and 10(d). This is significant considering that YSZ is ionic and YZN is metallic/covalent, and that a 50% difference in molar volume exists between the two phases. (The difference in lattice parameter is 13%.)

In addition to texture, the nitride–oxide conversion step can affect the mechanical integrity of the YSZ film. Figure 11 shows SEM images of two YSZ/Ni-RABiTS films, produced via the process, converted to the oxide using (a) a low (ppm-range) p_{O_2}, and (b) a high (percent-range) p_{O_2}. The film in Fig. 11(a) is smooth and conforms to the surface such that the topography of the RABiTS can be discerned. YSZ films do not display any cracking, unless a very high p_{O_2} is used for oxidation, as is the case for the film in Fig. 11(b), which exhibits bubbling, compressive-stress cracking, and spallation. The difference in lattice parameter between bulk YSZ and YZN is 13%, which would be expected to result in a large compressive stress in converted films. The lack of spallation in the film oxidized using a ppm-range p_{O_2}, versus the cracked film oxidized using a percent-range p_{O_2}, indicates that oxygen deficiency may play a role in alleviating this stress. Nitrogen bubble formation due to high-rate oxidation is a second possibility. Some degree of nitrogen could be dissolved in the YSZ lattice, although its effect on lattice parameter is unknown. The conversion process involves a large molar volume change, so care must be taken to use the appropriate oxidation conditions such that large stresses are not induced in the film. Oxidation times on the scale of minutes have been shown to allow enough time for atomic reorganization and removal of nitrogen-containing gases without these defects. Gaining a fuller understanding of the oxygen–nitrogen exchange mechanism will enable optimization of the oxidation step.

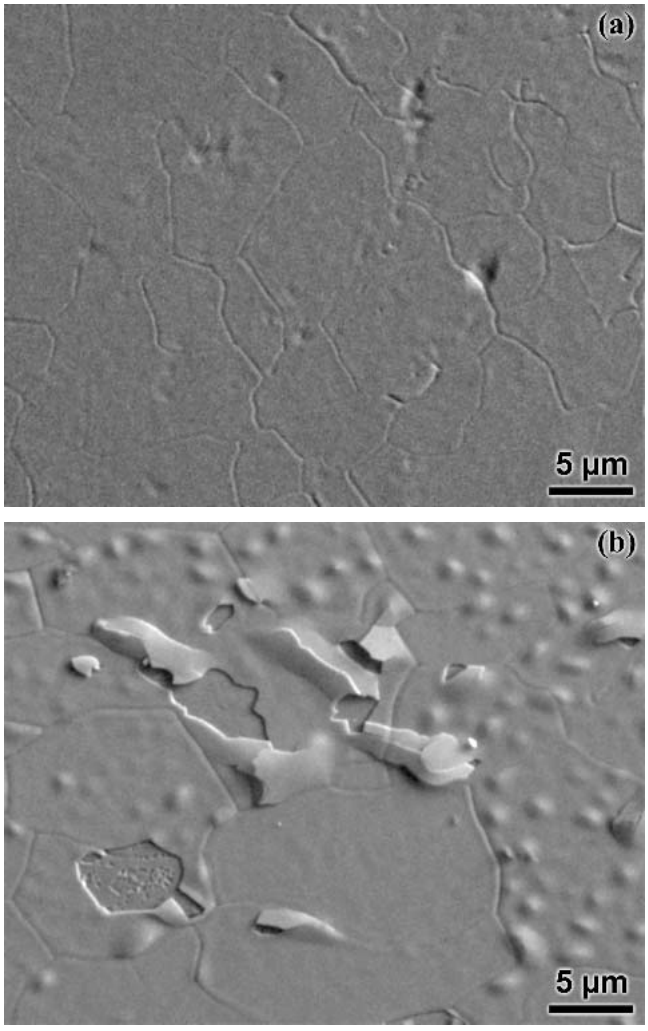

FIG. 11. SEM images of two YSZ/Ni-RABiTS films. (a) A film oxidized under optimal conditions shows a smooth surface morphology. The film in (b) was oxidized at a high p_{O_2}, resulting in blistering, cracking, and spallation of the film.

V. CONCLUSIONS

We have shown that nitrides provide a viable route to the deposition of buffer layers, which adopt the biaxial texture of the tape for Ni and Ni–Cr (13%) RABiTS, and which maintain this texture upon oxidation. The YSZ buffer layer thus produced often has a much narrower biaxial texture distribution, providing improvement over that provided by the RABiTS substrate. The texture of these buffer layers, both in- and out-of-plane, is sufficient that high-quality coated conductors can be fabricated. Pole figure analysis shows a single orientation for films. A J_c of 1 MA/cm^2 has been demonstrated for YBCO RABiTS coated conductors with a converted nitride buffer layer. Microscopic examination of partially converted nitride films show that the conversion front is planar and that the oxide is locally epitaxial with the nitride during the conversion process.

The consistent high-quality texture of YSZ films converted from YZN, and their smooth surface morphology, make the process an attractive candidate indeed. The process allows simplification of the buffer layer stack. The high rates possible for nitride deposition by reactive sputtering make this process a viable option for the economical production of second-generation HTS wires intended for a broad range of applications. In addition, this process may also be suitable for fabricating HTS on silicon and to serve as a templating technology for a broad range of device applications where growth of complex cubic oxides are desired on silicon or other substrates. Work is currently underway to address the technical and engineering challenges to scale up the process for production of buffer layers in long lengths. Efforts are also being used to gain a better understanding

of the oxygen–nitrogen exchange mechanism, and on optimizing the performance of films for use in commercial coated conductors.

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