Nucleation of epitaxial yttria-stabilized zirconia on biaxially textured (001) Ni for deposited conductors

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(Received 25 October 1999; accepted for publication 24 February 2000)

The nucleation of (001)-oriented yttria-stabilized zirconia (YSZ) directly on the (001) Ni surface is realized via nucleation on an oxygen-terminated nickel surface using pulsed-laser deposition. Under conditions where the nickel surface is either oxygen free or substantially covered with NiO, a mixed orientation of YSZ occurs. The epitaxial YSZ layer grown on a biaxially textured Ni(001) surface was used as a single buffer layer for a high temperature superconducting coated conductor architecture, yielding superconducting $YBa_2Cu_3O_7$ films with high critical current densities, J_c. This architecture eliminates the necessity for a multilayer buffer architecture, since high J_c superconducting films are achieved with no intermediate buffer layer between the (001) YSZ and the biaxially textured metal. © *2000 American Institute of Physics.* [S0003-6951(00)04516-2]

Significant effort to develop a high temperature superconducting (HTS) wire or tape has focused on the epitaxial growth of superconducting films on biaxially textured metals.[1,2] This approach is dependent on the heteroepitaxial growth of oxide buffer layer films on metal surfaces. In order to realize a HTS film possessing a high critical current on a biaxially textured metal substrate, the buffer layer architecture must satisfy a set of rather stringent requirements. The buffer layer construct must be chemically compatible with both the metal and superconductor. It must be mechanically robust so as to prevent microcrack formation at the HTS/buffer layer interface. It must also be epitaxial relative to the biaxially textured metal with a singular (001) orientation. The latter requirement is particularly challenging because the epitaxy of metal oxides on metal surfaces is determined not only by the relative lattice match between film and substrate, but by possible roles of a native oxide layer and/or intermetallic phases formed during film nucleation.

In previous work, these objectives have required multilayer combinations of various oxide buffer layers. Superconducting films with critical current densities in excess of 1 MA/cm^2 at 77 K have been achieved for epitaxial $YBa_2Cu_3O_7$ films on rolled-textured Ni substrates with the use of certain epitaxial buffer-layer constructs.[3] For example, CeO_2 has been used to nucleate an epitaxial (001)-oriented oxide layer on a biaxially textured (001) Ni surface.[4] A tendency for the CeO_2 layer to crack due to differences in thermal expansion coefficients of the oxide film and metal tape requires an additional epitaxial yttria-stabilized zirconia (YSZ) buffer layer on the CeO_2 in order to achieve crack-free superconducting films.[1]

Although effective in forming a high current superconducting tape, the use of a multilayer buffer architecture introduces significant complexity to the fabrication process. One of the most attractive materials for a single buffer layer that is mechanically robust and chemically inert is YSZ.[5–7] Unfortunately, previous efforts to grow epitaxial YSZ films with a single (001) orientation directly on rolled-textured (001) Ni tapes have been unsuccessful. In this letter, we report on the successful nucleation and growth of epitaxial (001) YSZ on the (001) Ni surface using pulsed-laser deposition. Using the (001)-oriented YSZ as a template, a $YBa_2Cu_3O_7$/YSZ/biaxially textured Ni structure with a high J_c is realized.

The substrates used in this study were biaxially textured (001) Ni tapes (50 μm thick) produced by cold rolling and annealing.[8] The starting purity of the Ni tape was 99.99%. For texture development, the cold-rolled metal tape was enclosed in a tantalum envelope and annealed in vacuum with a base pressure of 2×10^{-6} Torr at 800–1000 °C for 60 min. The tantalum envelope serves as a local oxygen getter during the anneal. The degree of in-plane and out-of-plane texture for the substrates as determined by x-ray diffraction (XRD) rocking curves was approximately 8°. Note that exposure to air almost immediately oxidizes the nickel surface. A 10 mol % yttria-stabilized ZrO_2 ceramic target was used to deposit the YSZ film. A KrF excimer laser beam was incident on the rotating target surface at an angle of 45 °C. The laser energy density was 2.6 J/cm^2 with a deposition rate of 0.25 nm/s. The target-to-substrate distance was 7 cm. The base pressure of the growth chamber was 10^{-6} Torr, assumed to consist primarily of water vapor. The YSZ nucleation experiments were performed in the temperature range of 600–800 °C.

For the experiments reported here, an initial nucleation layer (~5 nm) of YSZ was deposited under various conditions, followed by the additional epitaxial growth of a 200 nm thick YSZ film with $P(O_2)=10^{-4}$ Torr. The relatively thick epitaxial film grown on the nucleated template layer effectively reflects the crystalline orientation of the nucleated layer. Three ambient conditions were considered during nucleation. First, nucleation was carried out on an oxide-free surface by introducing sufficient hydrogen partial pressure

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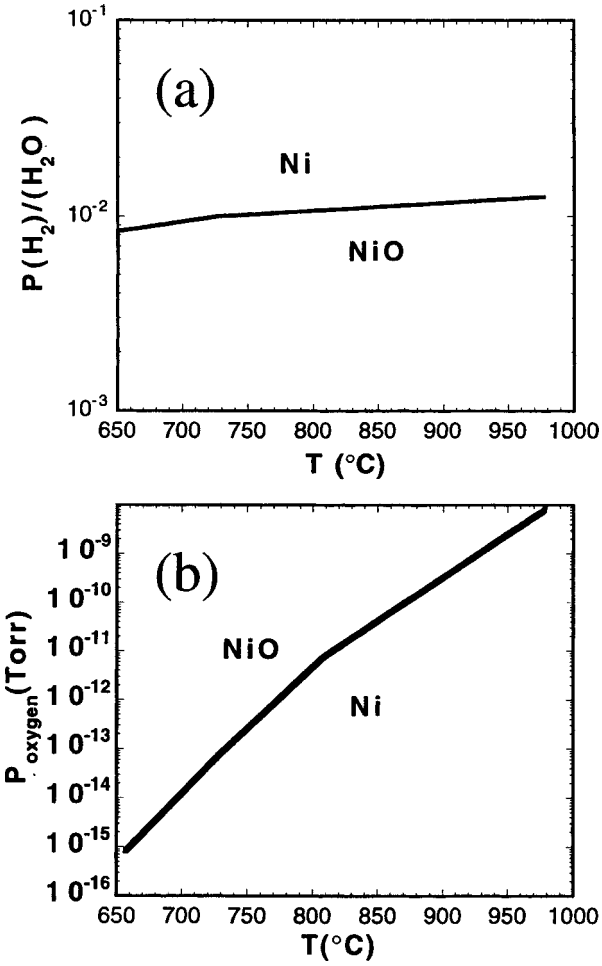

FIG. 1. Stability curves for NiO as a function of (a) $P(H_2)/P(H_2O)$ and (b) $P(O_2)$.

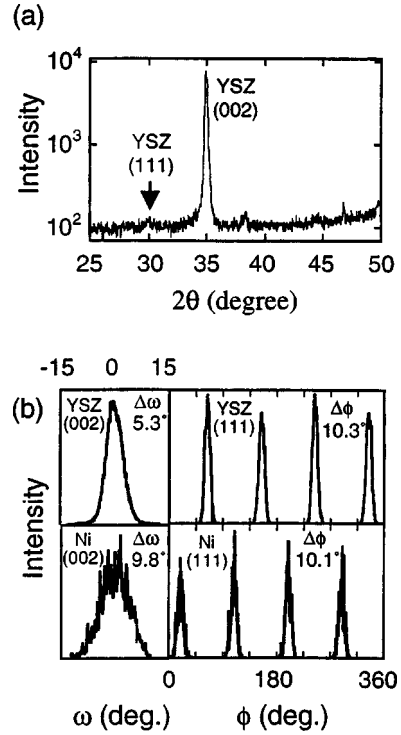

FIG. 2. θ–2θ x-ray diffraction scan (a) along the surface normal for a (001) YSZ film deposited on a biaxially textured Ni substrate at 800 °C. The nucleation layer was deposited at a background pressure of 2×10^{-6} Torr with no intentional oxygen flow. (b) Out-of-plane ω scans and in-plane ϕ scans of the YSZ peaks indicating that the film is in plane aligned and epitaxial.

(up to 200 mTorr 4% H_2/96% Ar mixture) so as to reduce any NiO on the Ni surface as determined from the thermodynamics of NiO reduction (case A). Figure 1(a) shows the stability curve for NiO in the presence of hydrogen and water vapor.[9] For $P(H_2)/P(H_2O)>10^{-2}$ at $T>600$ °C, NiO will be reduced, yielding an oxide-free surface. Second, nucleation was performed under base pressure conditions (case B). For a vacuum system with a base pressure of $\sim10^{-6}$ Torr, the residual oxygen pressure will be at least 10^{-8} Torr. Using Ellingham diagrams for binary oxides, the equilibrium oxygen partial pressure for NiO heated to 600–800 °C should be in the range of 10^{-16}–10^{-11} Torr as illustrated in Fig. 1(b).[9] Under the above conditions, oxygen termination of the surface would be expected. Surface studies have shown that ~3 monolayers of NiO rapidly form on a (001) Ni surface when exposed to oxygen at room temperature.[10–12] Based on the Gibbs free energy for NiO thermal decomposition, desorption of this native NiO layer from the nickel surface under conditions used in case B during film nucleation would not be anticipated.[9] Last, nucleation was carried out in an oxygen partial pressure of 5×10^{-6} Torr or greater (case C). Under these conditions, significant NiO coverage of the Ni surface is expected. Previous studies on the oxidation of the (001) Ni surface indicate that the native NiO formed at ambient temperatures is polycrystalline with multiple orientations.[10,12] However, the formation of a fine-grained (001) oriented epitaxial NiO layer has been observed after moderate vacuum annealing of an initially mixed oriented NiO thin layer using reflection high energy electron diffraction (RHEED).[12]

For nucleation of YSZ in the presence of sufficient hydrogen to yield an oxide-free nickel surface (case A), polycrystalline films with mixed orientation were realized. This differs from the case for CeO_2, where (001) epitaxy is achieved via nucleation on an oxide-free nickel surface.[1]

In contrast to nucleation in the presence of hydrogen, nucleation of YSZ at a base pressure of 10^{-6} Torr (case B) results in a singular (001) epitaxial relationship. Figure 2(a) shows a θ–2θ XRD scan along the surface normal for an epitaxial (001) YSZ film that was nucleated on the Ni (001) substrate at 800 °C in a background pressure of 2×10^{-6} Torr with no intentional oxygen or hydrogen flow. This film is essentially 100% (001) oriented with an intensity ratio of the (111) to (200) peaks less than 0.01. Figure 2(b) shows ω scans and ϕ scans of the YSZ peaks, indicating that the film is in plane aligned and epitaxial with respect to the biaxially textured Ni substrate. Similar results were obtained for films deposited at temperatures as low as 600 °C. For the case where an oxygen partial pressure greater than 5×10^{-6} Torr is introduced during either substrate heating or film nucleation (case C), a significant or dominant YSZ (111) oriented component was evident in the x-ray diffraction data. An oxygen partial pressure of 10^{-5} Torr or greater during nucleation yields nearly 100% (111)-oriented YSZ.

This study shows that epitaxial (001) YSZ can be nucleated on a Ni (001) surface that is apparently oxygen terminated under limited oxidation conditions that do not lead to

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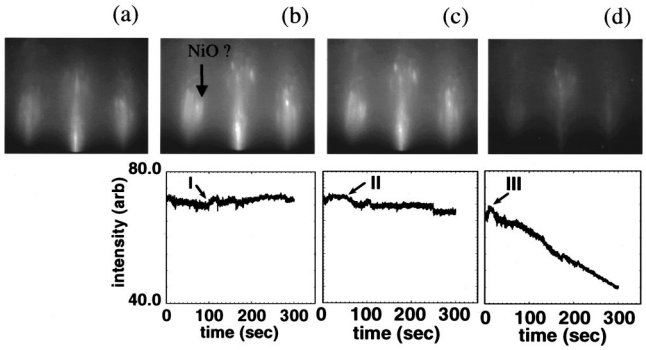

FIG. 3. RHEED pattern of a biaxially textured (001) Ni substrate taken along the Ni(110) direction at (a) 800 °C in $P(H_2)=8\times10^{-7}$ Torr, and at 700 °C in (b) $P(O_2)=1.5\times10^{-7}$, (c) 5×10^{-7}, and (d) 1.6×10^{-6} Torr. Plots of the specular intensity are also shown, with the points of oxygen introduction noted (I, II, and III).

significant polycrystalline NiO formation. It is conceivable that the YSZ (001) nucleates on (001) NiO that orders on the surface. However, the Gibbs free energy for YSZ is significantly larger in magnitude than that for NiO.[9] Thermodynamically, the reduction of NiO by YSZ is favored. Note that similar results have also been observed for the epitaxial growth of YSZ on (001) Si, where a thin amorphous SiO_2 layer is beneficial to achieving (001)-oriented epitaxy.[13–15] In order to elucidate the surface conditions at nucleation, we have investigated the progression of the Ni (001) surface as a function of background pressure conditions using RHEED. A biaxially textured Ni substrate was heated to 800 °C in $P(H_2)=8\times10^{-7}$ Torr with a base pressure of 1×10^{-8} Torr, then cooled to 700 °C and exposed to oxygen. Figure 3(a) shows the initial RHEED pattern at 800 °C in $P(H_2)=8\times10^{-7}$ Torr. The individual spots presumably represent diffraction from individual Ni grains. Figure 3(b) shows the RHEED pattern with $P(O_2)=1.5\times10^{-7}$ Torr. Also shown is a plot of the intensity for the (001) Ni specular reflection as the oxygen partial pressure is increased. At $P(O_2)=1.5\times10^{-7}$ Torr, one expects the nickel surface to be oxidized. Note that the RHEED patterns before and after the introduction of $P(O_2)=1.5\times10^{-7}$ Torr are quite similar. There appears to be evidence for NiO (100), although the diffusely scattered nature of the pattern makes this difficult to delineate. As $P(O_2)$ is increased to 5×10^{-7} Torr, a slight decrease in the specular reflection intensity is observed. At $P(O_2)>10^{-6}$ Torr, the RHEED pattern becomes weak and polycrystalline in nature. Clearly a polycrystalline oxide forms on the surface for $P(O_2)>10^{-6}$ Torr. This is the oxygen pressure that yielded (111) YSZ nucleation. Note that the polycrystalline NiO RHEED pattern remained if the oxygen was then removed to $P(O_2)<10^{-8}$ Torr, indicating that the oxidized surface is relatively stable once formed.

With the epitaxial growth of high-quality YSZ on biaxially textured nickel, a single buffer layer architecture for the growth of $YBa_2Cu_3O_7$ becomes possible. A 0.25 nm thick $YBa_2Cu_3O_7$ film was deposited on the 250 nm YSZ layer at a deposition temperature of 750 °C and oxygen pressure of 200 mTorr. A θ–2θ XRD scan along the surface normal for the $YBa_2Cu_3O_7$/YSZ/Ni structure shows only *c*-axis oriented $YBa_2Cu_3O_7$, with the intensity ratio of the YSZ (111) to (200) peaks less than 0.02. X-ray diffraction ω scans and ϕ scans for the YSZ and $YBa_2Cu_3O_7$ peaks show that the layers are in plane aligned and epitaxial with respect to the biaxially textured Ni substrate. Scanning electron microscopy images of the (001) YSZ/Ni films show no evidence for microcracking in the YSZ layer. The critical current density at 77 K for the $YBa_2Cu_3O_7$ film was 0.5 MA/cm^2, which is the highest reported value for any epitaxial $YBa_2Cu_3O_7$ film on biaxially textured metal substrate with a single buffer layer separating the superconductor from the biaxially textured metal substrate. Note that, for this sample, two in-plane variants of the $YBa_2Cu_3O_7$ film, rotated 45°, are present. This is commonly observed for $YBa_2Cu_3O_7$ epitaxial films deposited on a (001) YSZ surface, and is due to the lattice mismatch between $YBa_2Cu_3O_7$ and YSZ.[15] Minimizing or eliminating one of the in-plane variants by optimization of film growth temperature and oxygen partial pressure should lead to a significant increase in the critical current density for this architecture. A reproducible approach to eliminating one of these in-plane variants is to deposit a thin, epitaxial capping layer of CeO_2 or Y_2O_3 layer on the YSZ layer to improve the lattice match with $YBa_2Cu_3O_7$.[16] Incorporating this capping layer into the above architecture resulted in a relatively simple biaxially textured superconducting structure with a critical current density greater than 1 MA/cm^2.

This research was sponsored by the Office of Sciences and the Office of Power Technology, U.S. Department of Energy under Contract No. DE-AC05-96OR22464 with Lockheed Martin Energy Research Corp.

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