

AFRL-PR-WP-TP-2006-220

**STUDIES ON Ba_2YNbO_6 BUFFER
LAYERS FOR SUBSEQUENT
 $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ FILM GROWTH**



**Srinivas Sathiraju, Paul N. Barnes, Chakrapani Varanasi,
and Robert Wheeler**

MARCH 2004

Approved for public release; distribution is unlimited.

STINFO COPY

© 2005 IEEE

This work is copyrighted. One or more of the authors is a U.S. Government employee working within the scope of their Government job; therefore, the U.S. Government is joint owner of the work and has the right to copy, distribute, and use the work. All other rights are reserved by the copyright owner.

**PROPULSION DIRECTORATE
AIR FORCE MATERIEL COMMAND
AIR FORCE RESEARCH LABORATORY
WRIGHT-PATTERSON AIR FORCE BASE, OH 45433-7251**

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

The public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Department of Defense, Washington Headquarters Services, Directorate for Information Operations and Reports (0704-0188), 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to any penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number. **PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ADDRESS.**

1. REPORT DATE (DD-MM-YY) March 2004		2. REPORT TYPE Journal Article Postprint		3. DATES COVERED (From - To) 03/10/2003 – 03/10/2004	
4. TITLE AND SUBTITLE STUDIES ON Ba ₂ YNbO ₆ BUFFER LAYERS FOR SUBSEQUENT YBa ₂ Cu ₃ O _{7-x} FILM GROWTH				5a. CONTRACT NUMBER In-house	
				5b. GRANT NUMBER	
6. AUTHOR(S) Srinivas Sathiraju, Paul N. Barnes, Chakrapani Varanasi, and Robert Wheeler				5c. PROGRAM ELEMENT NUMBER 61102F/62203F	
				5d. PROJECT NUMBER 3145	
				5e. TASK NUMBER 32	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Power Generation Branch (AFRL/PRPG) Power Division Propulsion Directorate Air Force Research Laboratory, Air Force Materiel Command Wright-Patterson Air Force Base, OH 45433-7251				5f. WORK UNIT NUMBER 314532Z9	
				8. PERFORMING ORGANIZATION REPORT NUMBER AFRL-PR-WP-TP-2006-220	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) Propulsion Directorate Air Force Research Laboratory Air Force Materiel Command Wright-Patterson AFB, OH 45433-7251				10. SPONSORING/MONITORING AGENCY ACRONYM(S) AFRL-PR-WP	
				11. SPONSORING/MONITORING AGENCY REPORT NUMBER(S) AFRL-PR-WP-TP-2006-220	
12. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution is unlimited.					
13. SUPPLEMENTARY NOTES Journal article postprint published in IEEE Transactions on Applied Superconductivity, Vol. 15, No. 2, June 2005. © 2005 IEEE. This work is copyrighted. One or more of the authors is a U.S. Government employee working within the scope of their Government job; therefore, the U.S. Government is joint owner of the work and has the right to copy, distribute, and use the work. All other rights are reserved by the copyright owner. PAO case number: AFRL/WS 04-1275; Date cleared: 16 Nov 2004.					
14. ABSTRACT In this paper, we are reporting a dielectric oxide buffer Ba ₂ YNbO ₆ (BYNO) and its performance on various substrates for a potential buffer layer for the growth of YBa ₂ Cu ₃ O _{7-x} (YBCO) coated conductors. Ba ₂ YNbO ₆ is a moderate dielectric. Using pulsed laser deposition, epitaxial BYNO films were grown at 850 °C with an oxygen pressure of 200 mTorr on single crystal MgO (100) substrate and ion beam assisted sputter deposited MgO buffered hastelloy metal substrates. The surface morphology of the BYNO films reveals out growths even though the average surface roughness is only 2–8 nm. The texture of BYNO films is ~ 8° and thickness of these layers 100 nm on metal substrates. Highly c-axis oriented YBCO films were deposited on BYNO buffered substrates. Critical transition temperatures (T _{c0}) determined from electrical transport measurements vary between 88–89 K and corresponding critical current densities (J _c) ranging from 0.5–1 MA/cm ² at 77 K.					
15. SUBJECT TERMS Buffer layer, BYNO film, coated conductors, IBAD, YBCO film					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT: SAR	18. NUMBER OF PAGES 10	19a. NAME OF RESPONSIBLE PERSON (Monitor) Paul N. Barnes
a. REPORT Unclassified	b. ABSTRACT Unclassified	c. THIS PAGE Unclassified			

Studies on Ba_2YNbO_6 Buffer Layers for Subsequent $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ Film Growth

Srinivas Sathiraju, Paul N. Barnes, Chakrapani Varanasi, and Robert Wheeler

Abstract—In this paper, we are reporting a dielectric oxide buffer Ba_2YNbO_6 (BYNO) and its performance on various substrates for a potential buffer layer for the growth of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) coated conductors. Ba_2YNbO_6 is a moderate dielectric. Using pulsed laser deposition, epitaxial BYNO films were grown at 850°C with an oxygen pressure of 200 mTorr on single crystal MgO (100) substrate and ion beam assisted sputter deposited MgO buffered hastelloy metal substrates. The surface morphology of the BYNO films reveals out growths even though the average surface roughness is only 2–8 nm. The texture of BYNO films is $\sim 8^\circ$ and thickness of these layers ~ 100 nm on metal substrates. Highly *c*-axis oriented YBCO films were deposited on BYNO buffered substrates. Critical transition temperatures (T_{c0}) determined from electrical transport measurements vary between 88–89 K and corresponding critical current densities (J_c) ranging from 0.5–1 MA/cm² at 77 K.

Index Terms—Buffer layer, BYNO film, coated conductors, IBAD, YBCO film.

I. INTRODUCTION

RECENT advancements in coated conductor research based on the ion beam assisted deposition (IBAD) buffer architecture for MgO on metallic substrates show promise [1], [2]. Using IBAD-MgO is important as a template for subsequent epitaxial growth of conducting oxide buffer layers as well as the epitaxial growth of YBCO coated conductors. The high processing speed can be obtained because the optimum thickness of the IBAD-MgO template layer of approximately 10 nm is 100 times thinner than that needed to obtain good in-plane orientation in IBAD-YSZ films. However, it has been reported that for Y123 films deposited on MgO substrate, an interlayer of barium salt is formed at the interface if the processing temperature is above 700°C [3]. Also, another problem reported with the films grown on MgO is the presence of 45° in-plane rotated grains which degrades the crystalline quality of the deposited $\text{R}\text{Ba}_2\text{Cu}_3\text{O}_x$ (RBCO) where $\text{R} = \text{Y}$, rare earths [4]. A possible solution is to use a suitable thin buffer layer on the MgO buffered

metallic substrate or a separate buffer layer for the growth of the subsequent Y123 superconductor films. So far Yittria stabilized Zirconia (YSZ) and $\text{Gd}_2\text{Zr}_2\text{O}_7$ are some of the successful buffer layers for the long length tape production using IBAD technique [5], [6]. In the present study, we report, a moderate dielectric buffer layer Ba_2YNbO_6 on the single crystal MgO as well as on IBAD MgO tem plated polycrystalline Hastelloy substrates.

BYNO is a double perovskite structure of $\text{A}_2\text{BB}'\text{O}_6(\text{ABO}_3)$ type and has moderate dielectric properties [7]. It has cubic structure with $a = 0.84$ nm which is double of the unit cell of MgO ($a = 0.42$ nm). Moreover BYNO has no reaction with YBCO up to 900°C [7], [8]. These physical properties make it very attractive buffer layer for the coated conductors. Here, we report our effort to use a single layer of BYNO as a buffer for growth of high performance YBCO films on metal substrate where an IBAD MgO template is used. BYNO acts as barrier layer to reduce the interaction between the MgO buffered substrate and the YBCO films.

II. EXPERIMENTAL

The BYNO films were prepared by the pulsed laser deposition (PLD) technique using a lambda physik 304i KrF 248 nm excimer laser. We have used in house prepared stoichiometric sintered BYNO target. The BYNO target was prepared following the conventional solid state route by thoroughly mixing high purity (99.99%) Y_2O_3 , BaCO_3 , Nb_2O_5 in the stoichiometric ratio and calcifying the mixture at 1350°C for 48 hrs with three intermediate grindings. The phase pure material was finely grounded and palletized at a pressure of 3 Tons in the form of circular discs having 1 inch diameter and 3 mm thickness. These discs were sintered again at 1400°C for 24 hrs in air. MgO (100) and IBAD MgO buffered Hastelloy substrates were used in our studies. The dimensions of these substrates are 10 mm \times 10 mm \times 0.25 mm. IBAD MgO substrates have an average texture of $6\text{--}8^\circ$. The substrates were held to the substrate heater using silver paste. The base pressure of the chamber was maintained in high 10^{-7} Torr and specimens were then heated from room temperature to 850°C . Then chamber was filled with O_2 gas of 200 mTorr and BYNO films were subsequently deposited at 850°C . Details of deposition parameters used are summarized in Table I. After the deposition, films were cooled to room temperature in 500 Torr of oxygen. YBCO films were deposited on these buffer layers using PLD. As deposited BYNO films were analyzed by detailed X-ray diffraction studies. Two theta (2θ) scans were accomplished by using a Rigaku X-ray diffractometer. A Philips MRD with four circle

Manuscript received October 3, 2004. This work was supported in part by the U.S. Air Force Office of Scientific Research during 2002–2003 and Propulsion Directorate during 2003–2004 and 2004–2005.

S. Sathiraju is with the National Academy of Sciences and Air Force Research Laboratory, Wright-Patterson Air Force Base, OH 45433 USA (e-mail: srinivas.sathiraju@wpafb.af.mil).

P. N. Barnes is with Propulsion Research, Air Force Research Laboratory, Wright-Patterson Air Force Base, OH 45433 USA (e-mail: paul.barnes@wpafb.af.mil).

C. Varanasi is with University of Dayton Research Institute and Air Force Research Laboratory, Wright-Patterson AFB, OH 45433 USA (e-mail: chakrapani.varanasi@wpafb.af.mil).

R. Wheeler is with UES Inc., and Air Force Research Laboratory, Wright-Patterson Air Force Base, OH 45433 USA (e-mail: robert.wheeler@wpafb.af.mil).

Digital Object Identifier 10.1109/TASC.2005.848703

TABLE I
FILM DEPOSITION PARAMETERS

Laser deposition Conditions	Ba ₂ YNbO ₆	YBa ₂ Cu ₃ O _{7-x}
Deposition Temperature	850 °C	750-820 °C
Oxygen Pressure	250 mTorr	230mTorr
Laser Energy	2.5 J/cm ²	2 J/Cm ²
Laser frequency	10 Hz	4 Hz
Substrate-Target distance	6 cm	6 cm
Substrates	MgO, IBAD MgO/Hastelloy	YBNO buffered substrates
Thickness of the layer	50-200 nm	250-500 nm

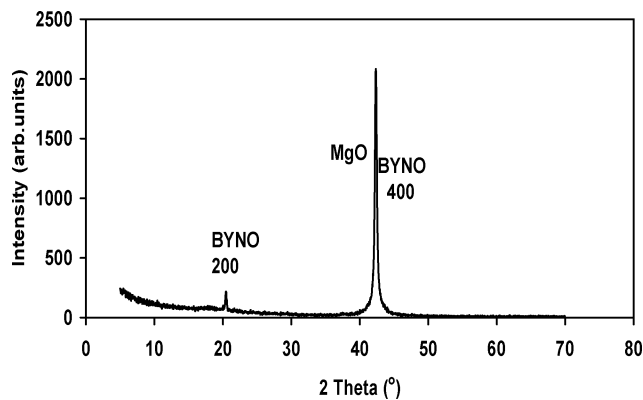


Fig. 1. X-ray diffraction pattern of BYNO film deposited on MgO (100) substrate. MgO (200) peak and BYNO (400) peak are overlapped on each other.

diffraction was used to study the crystalline alignment of the substrate, buffer layers, and the superconductor with phi scans. The microstructure of the various films was evaluated under atomic force microscopy (AFM) and Scanning Electron Microscopy (SEM) to study the surface roughness and surface morphology of the films as deposited films. Cross section of the samples was prepared using focused ion beam (FIB) method and Transmission Electron Microscopy has been used to study the interface of the layers. Transport current measurements were performed to check the critical temperature (T_c) and critical current density (J_c) of the superconducting films.

III. RESULTS AND DISCUSSION

X-ray diffraction of the BYNO film deposited at 850°C on MgO single crystal is shown in Fig. 1. All the peaks were indexed based on a cubic perovskite structure with general formula $A_2BB'O_6$ with lattice parameter 0.84 nm (PDF file no. 24-1042). Strong (400) orientation and weak (200) orientations of BYNO were observed $\sim 21^\circ$ and 42.9° of 2θ . MgO (200) orientation and BYNO (400) orientations are overlapped on each other. The in plane orientations of BYNO are $(100)_{BYNO} // (100)_{MgO}$ and $(100)_{BYNO} // (100)_{YBCO}$.

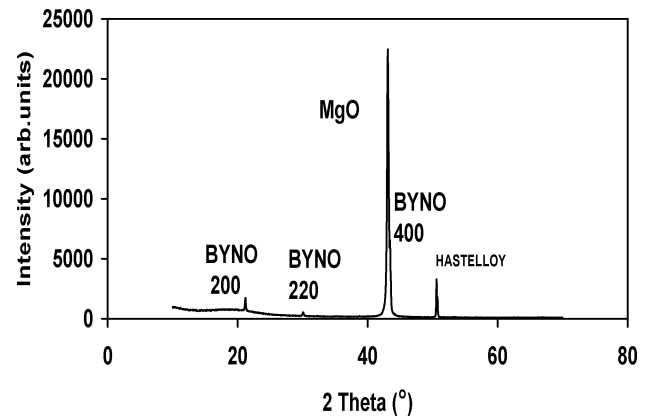


Fig. 2. X-ray diffraction pattern of BYNO film deposited on IBAD MgO buffered Hastelloy substrate.

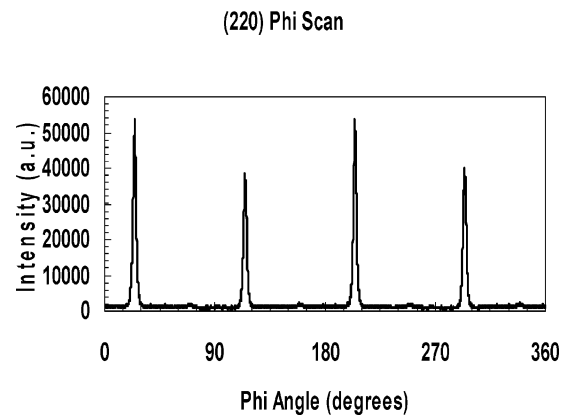


Fig. 3. X-ray Phi scan of (220) reflection of BYNO film deposited on IBAD MgO substrate.

Fig. 2 shows the X-ray diffraction of the BYNO films deposited at 850°C on IBAD MgO buffered hastelloy substrate. BYNO films deposited on IBAD MgO buffered Hastelloy metal substrate have shown highly (400) oriented and weak (200) and (220) in plane orientations do appear.

Fig. 3 Shows the phi scan of (220) reflection of BYNO film grown on IBAD MgO buffered Hastelloy substrate. The average full width at half maximum from the phi scan is around 8° . FWHM from this graph is 7.8° . The surface morphology of the BYNO films has been studied using AFM and SEM since the surface roughness of the buffer layer plays a key role in achieving high quality superconducting film [8]. BYNO film deposited on MgO single crystal has ~ 4 nm root mean square (RMS) surface roughness and BYNO film deposited on IBAD MgO substrate has a ~ 2 nm average RMS surface roughness. Table II lists the root-mean-square (RMS) surface roughness of the films deposited on various substrates.

Fig. 4 shows AFM image of the BYNO film deposited on IBAD MgO substrate. Even though average surface roughness is relatively low, our film consists of particulates and out growths. Fig. 5 shows the SEM image of the same film. Fig. 6 shows the XRD pattern of highly c-axis oriented YBCO film deposited on BYNO/IBAD-MgO/Hastelloy. Table III lists the Phi scan FWHM values of 220 orientation of BYNO film as well as

TABLE II
SURFACE ROUGHNESS OF AS DEPOSITED FILMS FROM AFM ANALYSIS

Film Architecture	RMS R_a (nm)	Thickness (nm)
BYNO/ I-MgO/Hastelloy	2	~100/10
BYNO/MgO	3	~100
YBCO/ BYNO/ IMgO/Hastelloy	8	~200/85/10
YBCO/BYNO/MgO	6	200/100

Last column indicates the thickness of the layers corresponding to the film architecture shown in first column without the substrate.

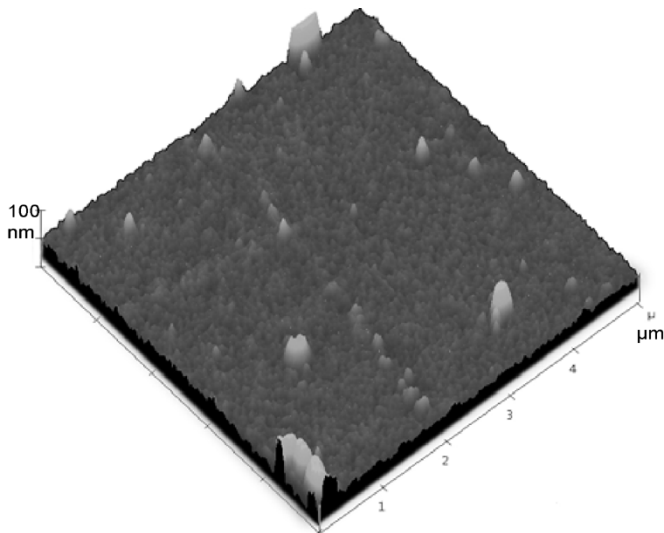


Fig. 4. Atomic Force Microscope image of BYNO film surface ($5 \mu\text{m} \times 5 \mu\text{m}$ area) deposited on IBAD MgO substrate. X-Scale is $1 \mu\text{m}/\text{unit}$ division and Z-Scale is $50 \text{ nm}/\text{unit}$ division.

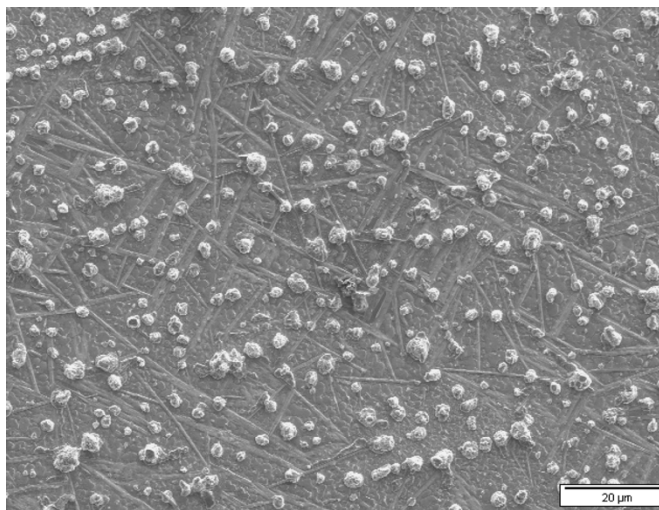


Fig. 5. Scanning electron microscope image of the same BYNO film at higher magnification.

(103) orientation of YBCO superconducting thin film. Also corresponding superconducting critical temperature (T_c) and Crit-

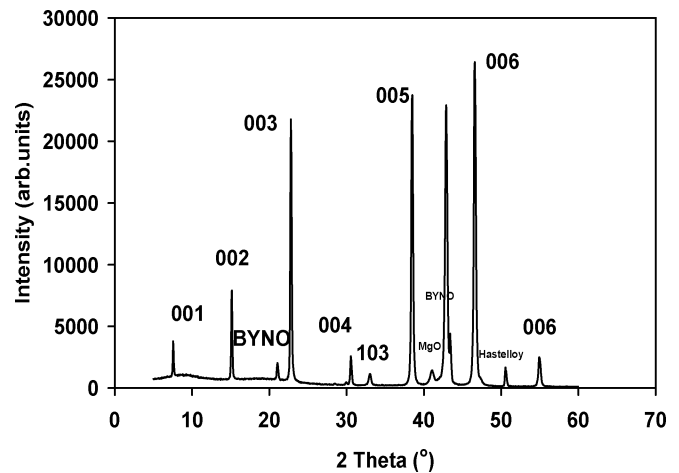


Fig. 6. Highly c-axis oriented YBCO films on BYNO/IBAD MgO buffered Hastelloy substrate.

TABLE III
TEXTURE RESULTS FROM PHI-SCANS OF BYNO AND YBCO FILMS

Base Substrate	FWHM of 85nm thick BYNO film (220) peak ($^\circ$)	FWHM of 200nm thick YBCO film (103) peak ($^\circ$)	YBCO film	
			T_c (K)	J_c At 77K (MA/cm^2)
MgO	2	4	89	1
IBAD MgO/ Hastelloy	8	8	88	0.5

Last column indicates the superconducting properties of YBCO films deposited on the BYNO buffered base substrates.

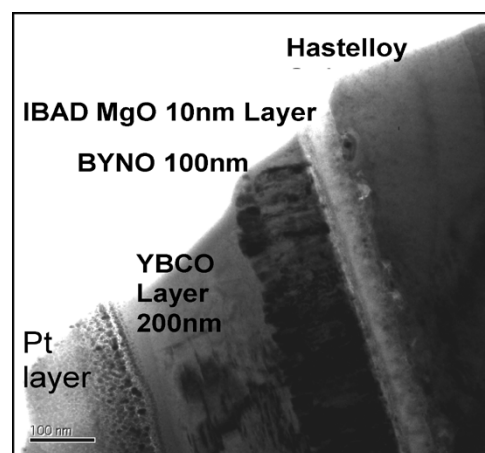


Fig. 7. Cross sectional transmission electron microscope image of the Pt protected YBCO (200 nm thick) deposited over a 100 nm thick BYNO layered IBAD MgO (20 nm thick) buffered Hastelloy substrate. Sharp interface between YBCO and BYNO layers can be seen.

ical Current density of the YBCO films deposited on the BYNO buffer layers were listed in the Table III.

Fig. 7 Shows cross sectional image of the transmission electron microscopy of YBCO film deposited on the BYNO (100

nm thick) buffered IBAD MgO (10 nm thick) layer on Hastelloy substrates. A clear boundary between layers suggests that there is no reaction between BYNO and other layers.

The YBCO film (200 nm thick) deposited here has a T_{c0} of 88 K and a J_c of $\sim 0.5 \text{ MA/cm}^2$ at 77 K. The deposition of BYNO may have been further optimized but the IBAD MgO layers were not of constant thickness for complete testing of compatibility and epitaxial growth of the BYNO buffer. Hence, the results presented here can be improved upon by varying the thickness of the BYNO buffer thickness as well as IBAD MgO template thickness. Further studies of the effect of BYNO thickness on the subsequent YBCO films, growth on RABITS Cu-Fe and Ni-based alloy substrates, and IBAD processing of BYNO are in progress.

IV. CONCLUSION

BYNO films have been successfully grown on single crystal MgO and IBAD buffered MgO/Hastelloy. Their microstructure-property relationship has been investigated and reported. It was noticed that there is no reaction between YBCO and BYNO films from cross sectional TEM studies. YBCO films deposited on BYNO buffered IBAD MgO/Hastelloy substrate has 88 K T_c and 0.5 MA/cm^2 J_c at 77 K.

ACKNOWLEDGMENT

S. Sathiraju is thankful to P. N. Arendt and Q. Jia from Superconductivity Technology Center, Los Alamos National Labora-

tory, and Los Alamos, NM for providing IBAD MgO buffered Hastelloy substrates, A. L. Campbell, Materials Lab, Air Force Research Laboratory, WPAFB, OH for AFM pictures.

REFERENCES

- [1] J. R. Groves, P. N. Arendt, S. R. Foltyn, Q. X. Jia, T. G. Holesinger, H. Kung, E. J. Peterson, R. F. Depaula, P. C. Dowden, L. Stan, and L. A. Emmert, "High critical current density $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thick films using ion beam assisted deposition MgO bi-axially oriented template layers on nickel-base super-alloy substrates," *J. Mater. Res.*, vol. 16, pp. 2175–2181, 2001.
- [2] S. Kreiskott, P. N. Arendt, J. Y. Coulter, P. C. Dowden, S. R. Foltyn, B. J. Gibbons, V. Matias, and C. J. Sheehan, "Reel-to-reel preparation of ion-beam assisted deposition (IBAD) of MgO based coated conductors," *Supercond. Sci. Technol.*, vol. 17, pp. 132–134, 2004.
- [3] M. Mukadia, Y. Takano, K. Chiba, T. Moriya, M. Kusunoki, and S. Ohshima, "A new epitaxial BaSnO_3 buffer layer for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin films on MgO substrates," *Supercond. Sci. Tech.*, vol. 12, pp. 890–892, 1999.
- [4] T. Yoshitake, W. Hattori, and S. Tahara, "Copper composition dependence of the structure and microwave properties in Y Ba-Cu-O films," *IEEE Trans. Appl. Supercon.*, vol. 9, pp. 3058–3063, 1999.
- [5] Y. Iijima, N. Tanabe, O. Kohno, and Y. Ikeno, "In-plane aligned $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin films deposited on polycrystalline metallic substrates," *Appl. Phys. Lett.*, vol. 60, pp. 769–771, 1992.
- [6] K. Kakimoto, Y. Iijima, and T. Saitoh, "Development of Y-123 coated conductors by ion-beam-assisted deposition," *Physica C*, vol. 378, pp. 937–943, 2002.
- [7] K. V. Paulose, J. Koshy, and A. D. Damodaran, " YBa_2NbO_6 synthesis, properties and compatibility with $\text{YBa}_2\text{Cu}_3\text{O}_x$," *Physica C*, vol. 193, pp. 273–278, 1992.
- [8] T. G. N. Babu and J. Koshy, " $\text{Ba}_2\text{RETaO}_6$ (RE = Pr, Nd, Eu, and Dy), a group of chemically stable substrates for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ films," *Solid State Chem.*, vol. 126, pp. 202–207, 1996.